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### ANTOINE LAVOISIER



PORTRAIT OF LAVOISIER AND HIS WIFE

After a portrait by David

## ANTOINE LAVOISIER

# THE FATHER OF MODERN CHEMISTRY

by

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# DEDICATED TO THE MEMORY OF

MY FATHER,
JAMES McKIE (1854-1934)

WHO FOUGHT WITH WOLSELEY
AT TEL-EL-KEBIR

### INTRODUCTION

THE FIRST great synthesis of physical principles was achieved towards the end of the seventeenth century, and culminated in Newton's famous Philosophiæ Naturalis Principia Mathematica. This was the establishment of the mechanical laws relating to the motion and interaction of large visible masses. What Dr. Douglas McKie sets forth in the present book, with such exact and comprehensive scholarship, is the story of the first great synthesis of chemical principles, which occurred towards the end of the eighteenth century, and culminated in Lavoisier's great Traité Elémentaire de Chimie. The thinker and intellectual leader in this movement was Antoine Laurent Lavoisier. Setting forth on his voyage of discovery with a wider vision and a greater synthetic imagination than any of his contemporaries or predecessors, he was able to combine with the results of his own famous experiments the discoveries of Black, Priestley, Scheele, and Cavendish, and in twenty years of continuous experimental work and intense thinking to clear away the mists of Greek and mediæval speculation and make the first great and solid beginning of a real science of chemistry, of the interactions and exchanges which occur in the invisible world of atoms and molecules. Not even in the *Mécanique Céleste* did the glory of the French intellect, in all its power of lucid and penetrating thought, achieve a sublimer triumph.

All this splendid story is described by Dr. McKie in a manner which entitles him not only to a very high place amongst the historians of science but also to the sincere gratitude of all lovers of scientific truth and humane scholarship.

The writer of this Introduction will never forget the feelings of admiration for the power of the human mind and spirit which he experienced when he first read Lavoisier's great Traité. It should be read by every student of science and learning, for it is one of the immortal landmarks in the history of civilisation. Will not the Institut de France issue a cheap facsimile edition, which could be placed in the hands of every young student? History is the central spine of all true culture, and ignorance of it the source of much of that vulgarity which disfigures the thought and feeling of the modern world. Fortunately there exists at the present time a welcome renaissance of public interest in history. Many brilliant writers—and who amongst them is greater than André Maurois?—are devoting their lives and talents to this work. That the literæ humaniores include as an essential part the history of scientific thought and discovery is a characteristic

feature of this new birth of learning. Amongst a talented group of young Englishmen who are thus extending the green fields of humane letters Dr. Douglas McKie has won for himself an honoured place.

F. G. DONNAN

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### PREFACE

IN DESCRIBING here the work done by Lavoisier a century and a half ago that led to the foundation of modern chemistry, we have attempted to appreciate the difficulties of that great labour, those difficulties that Hooke had in mind when he wrote in his Micrographia that "the footsteps of Nature are to be trac'd, not only in her ordinary course, but when she seems to be put to her shifts, to make many doublings and turnings, and to use some kind of art in endeavouring to avoid our discovery": and in hailing Lavoisier, rather than Boyle, as the "Father of Modern Chemistry," we have departed from tradition without any desire of denigration and with a full sense of the services which Boyle rendered to experimental science and especially to mechanical philosophy. But Boyle failed to convince chemists against Aristotelian and Paracelsan theory: and medicine, not science, remained their object. "If chymistry was once too wild and extravagant," wrote Francis Home in 1756, "it has been for many years too tame and confined. It seldom ventures further than the composition of a medicine, as if that were all the service it could be of to mankind " (Experiments etc., Edin., 1756, p. 8). Of modern chemistry, Boyle might be regarded as a distinguished but distant ancestor: between him and Lavoisier there stand Black, Priestley, Cavendish and Scheele, but in every sense, it seems to us, Lavoisier is thoroughly entitled to be regarded as the father and founder of modern chemistry. His execution at the age of fifty-one, at the very zenith of his powers, in the mad passions of the Revolution need remind us only that, when Lavoisier's head rolled from the guillotine, his great contemporary Priestley, our own countryman, was on the high seas, bound for America to escape a political persecution that had already nearly cost him his life.

For the biographical material in Chaps. I and IX we are extensively indebted to Grimaux's Lavoisier, 1743–1794 (Paris, 1888): all other sources are acknowledged in the text. Certain abbreviations have been used: Grimaux's Lavoisier is quoted as Grimaux, Berthelot's La Révolution Chimique–Lavoisier (Paris, 1890) as Berthelot, Meldrum's The Eighteenth Century Revolution in Science—The First Phase (Calcutta, 1929) as Meldrum, the collected edition of Lavoisier's works (Paris, 6 vols., 1864–93) as Œuvres, and the journal edited by the Abbé Rozier under the title of Observations sur la Physique as Observations, while the reprint in two volumes (Paris, 1777) of the early issues is quoted as Introduction.

Finally, the author's warmest thanks are due to his former teacher, Professor F. G. Donnan, C.B.E., F.R.S., Professor of Chemistry in the University of London and Director of the Chemical Laboratories in this College, for kindly contributing the Introduction.

D. McK.

University College, London. October 1935.

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### CHAPTER I

### BIOGRAPHICAL

Antoine Laurent Lavoisier was born in Paris in the Cul-de-sac Pecquet<sup>1</sup> on Monday, August 26, 1743; and he was christened in the parish church of St. Merry on the day of his birth. His ancestors belonged to Villers-Cotterets and had risen in the social scale from humble origins. Antoine Lavoisier, who died in 1620, was a postillion in the King's service at Villers-Cotterets; his son, Antoine, was a postmaster, and his grandson, also Antoine, a sheriff's officer, while his great-grandson, Nicolas, became a merchant. Antoine, son of Nicolas, was attorney to the bailiwick of Villers-Cotterets, and married in 1705 Jeanne Waroquier, daughter of a notary, by whom he had a son, Jean Antoine, who studied law at Paris and qualified as an advocate. In 1741 Jean Antoine succeeded his uncle, Jacques Waroquier, who had bequeathed him his fortune, as attorney to the Parliament of Paris; and on June 14, 1742, he married Émilie Punctis, daughter of Clément Punctis, advocate to the Parliament and secretary to the Marquis de

<sup>&</sup>lt;sup>1</sup> Now a through-way, the Passage Pecouay, connecting the Rue des Blancs-Manteaux with the Rue de Rambuteau, near the Archives Nationales. The house in which Lavoisier was born is no longer standing.

Châteaurenaud, vice-admiral of France. The first child of this marriage was Antoine Laurent Lavoisier, born in those easy middle-class circumstances to which his family appear to have risen from the moment when great-grandfather Nicolas turned to trade and thereby enabled his son and grandson to marry the daughters of wealthy lawyers. But the first and the last of the Lavoisiers both bore the name of Antoine; and to the end of his life the subject of this biography preserved his family relationship with the village of Villers-Cotterets, where his earliest known forbear had ridden the horses of King Louis XIII.

In 1748 Mme. Lavoisier died, and Antoine Laurent and his young sister, Marie Marguerite Emilie, born in 1745, moved with their father to the Rue du Four St. Eustache<sup>1</sup> to the house of their grandmother, Mme. Punctis, now a widow through the death of Clément Punctis in 1747. The two children were taken charge of by their aunt, Constance Punctis, then aged twenty-two, with a devotion that apparently led her to refuse marriage and that could have been surpassed only by the love of their own mother. The young Lavoisier presently entered the Collège Mazarin, founded under the will of the great cardinal who had governed France during the minority of Louis XIV; and he gained the second prize for rhetoric in a general competition in 1760.2 But in this year death again

<sup>&</sup>lt;sup>1</sup> Now the Rue de Vauvilliers, between the Bourse and the Halles.

<sup>2</sup> The prizes in this competition, which originated in a bequest dating from 1746, were presented at the Sorbonne: Robespierre won a number of them and seems to have distinguished himself at Latin (L'Intermédiare des Chercheurs et Curieux, Paris, 1886, p. 480).

struck the family, and Lavoisier lost his sister when she was barely fifteen years old.

At first Lavoisier had literary ambitions and spent some time in the writing of essays for the competitions organized by the provincial Academies. But he had developed a taste for science, possibly by attendance at the scientific lectures said to have been given at the Collège Mazarin, and, although he studied law on leaving college, becoming a Bachelor in 1763 and a Licentiate in 1764, he worked at the same time at mathematics and astronomy under the Abbé de Lacaille,1 botany under de Jussieu,2 mineralogy and geology under Guettard<sup>3</sup> and chemistry under Rouelle.<sup>4</sup> And he studied also anatomy and meteorology. His interest in meteorology led him at this time to begin making barometrical observations several times every day and to engage relatives and correspondents in various places to do likewise with the object of discovering the laws that govern the movements of the atmosphere. This work was continued throughout his life and death alone prevented him from attempting to co-ordinate observations made at

<sup>&</sup>lt;sup>1</sup> Nicholas Louis de Lacaille (1713-62), Professor of Mathematics at the Collège Mazarin, where he had an observatory. Spent four years on astronomical work at the Cape of Good Hope.

cal work at the Cape of Good Hope.

<sup>2</sup> Bernard de Jussieu (1699-1777).

<sup>3</sup> Jean Étienne Guettard (1715-86), one of the founders of modern geology and the originator of geological maps.

<sup>4</sup> Guillaume François Rouelle (1703-70), demonstrator in chemistry at the Jardin des Plantes from 1742 to 1768. Classified salts according to their crystalline form, but did little original work owing to his great interest in teaching. His pupils included, besides Lavoisier and others, the chemists Cadet, Macquer and Bayen. A lovable and eccentric personality, the subject of many amusing stories, enthusiastically admired by Diderot, Rouelle exercised by his teaching a great influence on the development of chemistry exercised by his teaching a great influence on the development of chemistry in France.

stations ranging from Paris to Aleppo and even to Baghdad. His anatomical studies are an indication of interests that were ultimately to lead him to fundamental discoveries in physiology. These busy days did not permit him to indulge in many social activities and indeed Lavoisier, a serious youth of nineteen, decided to cut himself off from such light affairs, excusing himself on the grounds of ill-health and actually living for some months on an exclusive diet of milk. His friends really believed him to be unwell and one of them, de Troncq, soundly advised him in 1763 that a year longer on earth was worth more than a hundred years in the memory of men.

Thus at the age of twenty, Lavoisier enjoyed the company of some of the most distinguished French scientists of his day. Indeed, as Grimaux put it, from being his masters, they had become his friends. Moreover, Mme. Punctis preferred solitude and quiet in her house in the Rue du Four St. Eustache and only great intimates were admitted there. Among these was Guettard who, it seems to us, exercised such a powerful influence over Lavoisier that he may almost be said to have brought about Lavoisier's adoption of a scientific career. Guettard had first won distinction as a botanist; but, applying himself afterwards to geology and mineralogy, he conceived the idea of constructing mineralogical maps.2 His travels in France and

<sup>&</sup>lt;sup>1</sup> In 1790 Lavoisier drew up a set of rules for predicting changes in the weather and suggested the publication every morning of daily weather forecasts (Œwres, III, p. 765).

<sup>2</sup> Really geological maps, the term mineralogy not having acquired at that time its present significance.

several voyages abroad, undertaken with this end in view, soon taught him that life was too short for him to hope to carry out his work alone and in 1763 he accordingly invited Lavoisier to collaborate with him in carrying out and continuing this heavy task. Lavoisier accepted the invitation and spent the years 1763 to 1766 in collecting the necessary information from the various provinces bordering on Paris, usually spending his vacations with his relatives at Villers-Cotterets. His note-books for this period show him to have held personal views about the data he was amassing and not to have been a mere laborious compiler. Indeed, his first original research dates from this period and was evidently inspired by facts that had come to his notice in his work with Guettard; for, in 1764, he studied the various kinds of gypsum, the substance commonly known as plaster of Paris, and presented the results to the Academy on February 25, 1765, in a memoir<sup>1</sup> that was the first of a long series extending over thirty years and including some of the classic papers in the history of chemistry. In this first memoir Lavoisier studied a number of varieties of gypsum, determined their solubilities in water, explained the binding of plaster by showing that gypsum on heating lost a quantity of water, that it took this up again when re-mixed with water and that this re-combination or rather re-crystallization was the cause of its solidification. He was careful in his conclusions to go no further than the facts

<sup>1</sup> Mémoires de Mathématiques et de Physique Présentés à l'Académie Royale des Sciences, par divers Savans, & lús dans ses Assemblés, 1768, 5, 341.

warranted and to avoid hypothesis. Observing that overburnt gypsum would not bind with water to form plaster, he indicated that he could guess at various causes for this, even probable causes, but that he regarded such speculation as out of place in chemistry, a science where experiment must control all advance.

Another problem also attracted Lavoisier's attention at this time. In 1765 the Academy offered a prize of 2,000 livres to be awarded in the following year for an essay on the best means of lighting the streets of a large town at night, special consideration to be given to the degree of illumination and the easiness and economy of the method proposed. Lavoisier entered the competition and studied various kinds of lamps burning candles or oil, elliptical and hyperbolic reflectors, numerous oils, wicks of various shapes, and the relation between intensity of illumination and consumption of oil. The Academy divided the essays submitted into two classes, namely, a first class consisting of those that were purely practical in approach, and a second class consisting of those in which the scientific principles of mathematics and physics had been applied. The prize was divided between Bailly, Bourgeois and Le Roy, whose essays belonged to the first class. Among the competitors of the second class, only Lavoisier was named: but his essay was specially mentioned and he was awarded a gold medal by the King. This was presented to him by the president at the public assembly on April 9, 17661;

<sup>&</sup>lt;sup>1</sup> Hist. Acad. R. Sci., 1766, p. 165.

and the attention of the public was drawn to the young scientist who had won recognition at the Academy at the early age of twenty-three.<sup>1</sup>

Guettard now received official recognition and patronage for his work; his plan for a mineralogical atlas of France was adopted by the Government; and in 1767 he was despatched to carry out the work for the provinces of Alsace and Lorraine. Again he called on Lavoisier as collaborator; and again the invitation was accepted. Guettard was a man of hot temper and brusque manner, suffering no contradiction. He had been educated by the Jesuits and their expulsion was a subject that constantly roused his fury. His manner of dealing with his fellows may be glimpsed from his reply to one who thanked him for support in being elected to the Academy: "You owe me nothing," said Guettard. " If I had not thought it just to give you my vote, you would not have got it, because I don't like you." But he had almost a father's love for Lavoisier; and his rudeness was for those in power and authority, while his kindness to his inferiors, his wit and his lightheartedness won the friendship of younger contemporaries.

On June 14, 1767, at three o'clock in the afternoon Lavoisier set out with Guettard for the Vosges. He was a healthy young man of twenty-four with fifty louis in his pocket and a good horse between his knees; he was a keen scientific observer, anxious to see new places and thoroughly aware that he was

<sup>&</sup>lt;sup>1</sup> Journal des Sçavans, September, 1766, p. 627: "Le Public a vû avec plaisir cette distinction si flatteuse pour un jeune Auteur, & dont il n'y avoit pas eu encore d'exemple dans l'Académie des Sciences."

going to collect a mass of interesting facts; he was escorted by his trusty servant Joseph; and he was accompanying one of the most distinguished scientists of the France of his day. Life must have seemed good to him at that moment; but it was clouded by his departure from his family, now limited by the death of Mme. Punctis in 1766 to his father and Mlle. Punctis. Indeed he wrote to them that evening from Brie-Comte-Robert to assure them that the travellers were happy and well and to say, as he was invariably to do in subsequent letters, that the horses were fit and well foddered. This was the first letter of many: and in the Rue du Four St. Eustache, as Mlle. Punctis said later on, the post was looked for like the coming of the Messiah.

The anxiety of Lavoisier's relatives grew as the travellers left the country known to them and especially as they approached the mountains of the Vosges and the mines of St. Marie, which it was intended that they should visit. On June 25, Mlle. Punctis wrote "Je commence à vous perdre de vue et m'en alarme. Je crains pour vous la chaleur qui commence vivement, je crains les armes que vous avez sur vous, quoiqu'elles peuvent vous être d'une grande utilité pour les bêtes et gens, et je crains les mines<sup>1</sup>; mon cœur n'est soulagé qu'en vous engageant, par notre tendre amitié, à user encore de plus de prudence, s'il est possible, que vous ne vous étiez promis," and, abandoning the formal for the familiar tutoiement, "Notre crainte est

<sup>&</sup>lt;sup>1</sup> The mines were, however, flooded when the party arrived and descent was impossible.

que tu ne reçoives pas toutes les lettres que nous t'écrivons, et ton père propose, si tu juges convenable, pour qu'on y fasse plus d'attention à la poste, de mettre : A M. Lavoisier, envoyé par le roi dans les Vosges. Nous espérons aujourd'hui recevoir de tes nouvelles ; il nous en faut souvent pour soutenir ton absence. Nous attendons le facteur comme le Messie. Tu sais nos conventions ; cela nous suffit, mais ne nous néglige pas, car notre situation serait à plaindre ; c'est notre soutien. . . . Porte-toi bien, mon cher enfant, ménage-toi bien, pense un peu à moi seulement pour te conserver, et crois à la tendresse sincère de ta meilleure amie."

But Lavoisier could do no more than his best to allay the fears of his relatives: he was often far from the highroad, and in those days Paris was a month from Strasbourg and in some parts the fourhorse mail-coach took an hour to cover a league. His yearning for his home was keen and at times he had to persuade himself that the journey was a pleasant one and that there was good work on hand. Meanwhile, Mlle. Punctis kept him posted in all the news of this bourgeois Parisian household of the mid-eighteenth century; and, with his father, consoled herself for Lavoisier's absence by arranging his library, adding to it the numerous books that he had sent home, and unpacking the cases of mineralogical specimens that frequently arrived. Occasionally there were instruments or clothes to be sent on. But when Lavoisier's father, on going to meet his son at Bourbonne-les-Bains, was asked to bring

<sup>&</sup>lt;sup>1</sup> Grimaux, p. 15.

with him some gold-fish for Mme. de Brioncourt, whose hospitality the travellers had enjoyed, and to get them from the Palais-Royal through Marianne, Guettard's housekeeper, he protested that it was une vilaine commission and that it would mean carrying the bowl of gold-fish the whole way in his hands, and that without the certainty of getting them there alive.

Lavoisier's detailed diary of his travels shows that between five and six o'clock every morning before setting out on the day's work he recorded the readings of the thermometer and barometer; that he repeated these observations several times during the day and made another on arrival at his destination for the night; that he noted the nature of the soil, the relief of the land and its vegetation, visited mines, iron-works and bleaching-works: that he gathered information about the various kinds of stone quarried and what plaster was used; that he took the temperature and density of the various river waters and mineral waters and even of the water supplied at the various inns at which he stayed; and that each evening he completed his diary, dealt with his letters and wrote down his expenses. And at the same time he was collecting specimens of minerals for Bertin, the Minister who had recognized this work, and also forming a collection of his own. And there were other activities as well as these. At Basle, for instance, there was much to interest the travellers; for among its

<sup>&</sup>lt;sup>1</sup> Guettard had charge of the natural history collection of the Duke of Orléans at the Palais-Royal and had apartments there.

citizens were Daniel Bernouilli, Dassonne, Raillard, Bruchner, and there was the tomb of Maupertuis to visit. Here, too, the expedition's barometer was unfortunately broken; but happily M. Jacques Bavière at the sign Aux trois pots rouges insisted on presenting his own barometer in its place, thereby unwittingly saving himself from utter biographical extinction. At Strasbourg they met the chemists Spielmann<sup>1</sup> and Ehrmann; and here too Lavoisier spent 500 livres in buying books on chemistry published in Germany and not known in Paris. And so the journey wore on, sometimes comfortably and sometimes not, as for instance in one village, where the only accommodation was a kind of granary, in which there was drying a stock of onions that, in Lavoisier's words, "tainted" the travellers.

At last they arrived at Bourbonne-les-Bains, where the elder Lavoisier had been anxiously and impatiently awaiting his son. The meeting over, they went on to Chaumont, whence the father departed for Villers-Cotterets, while Guettard and Lavoisier continued on their way to Paris. Mlle. Punctis received her nephew on the evening of October 19; and the latter, even on this occasion, did not forget to record the reading of the barometer before retiring to sleep off in the comfort of his home the fatigues of a tiresome journey. Two days later Lavoisier joined his father at Villers-Cotterets, where he continued his meteorological

<sup>&</sup>lt;sup>1</sup> J. R. Spielmann, author of *Institutiones Chemiæ* (Strasbourg, 1763 and 1766). A French version, *Instituts de Chimie*, was published at Paris in 1770.

observations. In mid-November father and son returned to Paris and the family was once again united.

On his return to Paris Lavoisier busied himself with arranging the masses of data that he had collected, setting out his numerous analyses of water in a lengthy memoir which, however, was not published until after his death.1 Along with Guettard he was involved in the heavy labours of preparing from their joint observations the mineralogical atlas of France, sixteen of the 230 proposed sheets of which were already engraved in 1770. But the expenses proved too great and there were delays. The retirement of Guettard led to Monnet, the inspector-general of mines, being given charge of the work. In 1780 Monnet published the atlas in incomplete form in the name of Guettard and himself,2 ignoring all Lavoisier's contributions except the first sixteen sheets and making use of the wealth of observations for which Lavoisier was alone responsible. Lavoisier was galled at what he described as Monnet's "impudence"; and this incident made an enmity between the two that persisted, Monnet attacking Lavoisier even after his death in a book entitled Démonstration de la fausseté des principes des nouveaux chymistes (Paris, 1798). Yet Guettard had praised Lavoisier's work at the Academy (Observations, 1775, 5, 357); and Lavoisier had been associated with Dupain-Triel in an attempt to draw up at his own expense a mineralogical

<sup>&</sup>lt;sup>1</sup> Œuvres, III, p. 145. <sup>2</sup> Allas et description minéralogiques de la France, entrepris par ordre du Roi par MM. Guettard et Monnet, Paris, 1780.

map of France, work for which even in 1793, the last year of his life, he demanded the utmost national recompense for his friend Dupain-Triel. Lavoisier's interest in geology persisted throughout his life, and a memoir dealing with some of this early work appeared as late as 1789 (Mém. Acad. R. Sci., 1789, p. 351).

Since 1766 Lavoisier had been nominated by his friends for election to the Academy; and in 1768, when the chemist Baron died, it seemed very probable that Lavoisier would be chosen in his place. But the metallurgist Jars appeared as a formidable rival, the Minister, de Saint-Florentin, being anxious that Jars should be elected in recognition of the many important services that he had rendered to the State. Jars was thirty-six; he had been in charge of the development of the argentiferous lead mines at Poullaouen in Brittany and the coal-mines of Ingrande in Anjou, and later he had served on various official missions, visiting the mines of Saxony, Austria, Carinthia, Bohemia, the Hartz, Sweden, and Norway, and the factories of Holland and England, whence he introduced into France the manufacture of red-lead 1

Lavoisier, on the other hand, was well known to many of the Academicians for his interest and enthusiasm in scientific work. For his essay on the lighting of towns he had, as we have already seen, been specially awarded a gold medal by the King; and he had read four memoirs to the Academy,

<sup>1</sup> De Fouchy, Éloge de Jars (Mém. Acad. R. Sci., 1769, p. 173)

two on gypsum<sup>1</sup> and two on hydrometry,<sup>2</sup> which had been very well received. Indeed, Nollet and Macquer, reporting on one of them, had written: "Son mémoire est celui d'un homme qui a beaucoup de connaissances physiques et qui les employe avec une grande sagacité."3 His collaborative work with Guettard was also well known to the Academicians and had shown him to be a young man of considerable scientific promise.

At this time the Academy consisted of members of several different grades with varying privileges. Firstly, there were twelve honoraires, chosen from the nobility, and these alone could serve as presidents or vice-presidents. Then there were eighteen pensionnaires, after whom came twelve associés and then twelve adjoints. Besides these, there were the associés libres, the associés étrangers, the pensionnaires vétérans and the associés vétérans. Voting in the affairs of the Academy was restricted to the honoraires and the pensionnaires. The two associés, in whose branch of science the vacancy fell, had however some voice with the three pensionnaires in drawing up the list of candidates. The adjoints had few rights in this almost feudalistic assembly except bare membership; in the meetings they sat on benches behind the chairs of the associés, but, if there was a vacant chair, it might be occupied by an adjoint.

<sup>&</sup>lt;sup>1</sup> The first was published in 1768 (see p. 23 above, footnote); the second was published posthumously (*Œuvres*, III, p. 128).

<sup>2</sup> Read on March 23, 1768, and published posthumously (*Œuvres*, III, pp.

<sup>427</sup> and 145).

3 Quoted by Meldrum from the Registers of the Academy for May 11, 1768 (Isis, 1933, 19, 332).

The election to the vacancy caused by Baron's death took place on May 18, 1768, and Lavoisier obtained a majority of the votes.1 The Academy, however, merely recommended and the final choice lay with the King. The names of the two nominees were submitted to the King by the Minister, de Saint-Florentin, who advised the election of Jars and, in deference to the wishes of the majority of the Academy, the provisional establishment of a new place as adjoint, to be given to Lavoisier, on the understanding that there would not be an election when the next vacancy among the adjoints came to be filled.2 Jars and Lavoisier were formally admitted on June 1, 1768. The Secretary's minute of the proceedings, including the reading of the Minister's letter and the admission of the new Academicians, reads as follows: "J'ai, M. rendu compte au Roy de l'élection faite à l'Académie le 18 de ce mois pour remplir la place vacante par le mort de M. Baron. Sa Majesté a mis en considération que le nombre des voix pour MM. Lavoisier et Jars étaient presquégales que cependant le S. Jars était beaucoup plus âgé que le S. Lavoisier et que d'ailleurs il a été employé dans plusieurs circonstances par ordres de sa Majesté pour des objets intéressants à Son Service et même à l'Etat. Sa Majesté a jugé à propos de le choisir pour remplir la place vacante et attendu que le S. Lavoisier est aussi un sujet très distingué Sa Majesté

Ct.

<sup>&</sup>lt;sup>1</sup> Among Lavoisier's supporters were de Jussieu, who had taught him botany, and Duhamel du Monceau, the former of whom had belonged to the Academy for forty-three and the latter for forty years.

<sup>2</sup> Jars died suddenly the following year and the situation was regularized.

l'a pareillement nommé pour être dès à présent adjoint dans la même classe et lorsqu'il viendra à vaquer une autre place dans cette classe il n'y sera point fait de nouvelle élection. Vous voudrez bien informer l'Académie de cette disposition de Sa Majesté.

"Après la lecture de laquelle MM. Jars et Lavoisier ont pris place."

Thus Lavoisier entered the Academy at the early age of twenty-five; and the news of his election brightened the convalescence of his father, then recovering from a serious illness, while the feelings of Mlle. Punctis may perhaps best be appreciated from a letter written to her by de La Voye: "Je vois la joie briller dans vos yeux, en apprenant que ce cher neveu, l'objet de toutes vos complaisances, est nommé à l'Académie des sciences. Quelle satisfaction que dans un âge si tendre, où les autres jeunes gens ne songent qu'à leurs plaisirs, ce cher enfant ait fait de si grands progrès dans les sciences, qu'il obtienne une place que l'on n'obtient ordinairement, après beaucoup de peine, qu'à plus de cinquante ans."<sup>2</sup>

The expedition with Guettard had opened a chapter in Lavoisier's life: the election to the Academy opened another and a greater one. He was, at twenty-five, an active young man of considerable scientific experience: and he had at his hand ample private means to exploit his great gifts. The sources of his fortune were later on to bring

<sup>&</sup>lt;sup>1</sup> Quoted by Meldrum from the Registers of the Academy for June 1, 1768 (Isis, 1933, 19, 333).

<sup>2</sup> Grimaux, p. 30.

upon his head the opprobrium of his countrymen and, indeed, to drag him to the guillotine; but there is no shadow of suspicion that Lavoisier ever prospered unjustly or that he did not put his wealth to the best uses. Yet in many incidents in his life Lavoisier gives the impression of being a thoroughgoing man of affairs; and it seems to us not improbable that Monnet's disregard of his rights in the publication of the mineralogical atlas, the first important original work on which he had been engaged, led Lavoisier to a very clear awareness of when it was necessary to fight for his own hand and some aptness in the manner of his doing it.

Soon after his election Lavoisier was called upon to take part in the preparation of various scientific reports, either on matters of public interest or on memoirs submitted for the acceptance of the Academy; and, while the details of this work do not concern us here, it is not out of place to indicate that he was almost continually engaged in this kind of work and that he gave much thought, time and attention to it. During his long association with the Academy, Lavoisier drew up either alone or in collaboration with other members appointed for the purpose reports on the water-supply of Paris, prisons, animal magnetism, the adulteration of cider, the site of slaughter-houses, Montgolfier's "Aerostatic Machine," bleaching, tables of specific gravities, hydrometers, the theory of colours, lamps, meteorites, smokeless grates, tapestry making, the engraving of coats-of-arms, paper, fossils, an invalid

chair, a water-driven bellows, tartar, sulphur springs, the cultivation of cabbage and rape seed and the oils extracted thence, a rasp for tobacco, the working of coal-mines, white soap, the decomposition of nitre, the manufacture of starch, expressed oils, the distillation of phosphorus, the storage of fresh water on board ship, fixed air, a reported occurrence of oil in spring-water, lead ore, zeolite, marble, various machines, dyeing, the removal of oil and grease from silks and woollens, the preparation of nitrous ether by distillation, ethers, a reverberatory hearth, a new ink and inkpot in which it was only necessary to add water in order to maintain the supply of ink (some hopeful inventor had evidently sought the approval of the Academy), thermal springs, steel, vegetable rouge, specular iron ore, spathic iron ore, crystallization, the purification of oil, the art of the pewterer, salt, volatile marine acid, the decomposition of sal ammoniac, Chinese ink, glacial vitriolic acid, glass, the effects of mephitic fluids on animals, the decomposition of nitrous (nitric) acid, metallic soaps, a factory for watch-glasses, the production of volatile alkali, the precipitates of iron, the estimation of alkali in mineral waters, a powder-magazine for the Arsenal, the mineralogy of the Pyrenees, wheat and flour, cess-pools and the air arising therefrom, the alleged gold in the ashes of plants, arsenic acid, the parting of gold and silver, the base of Epsom salt, the winding of silk, the solution of tin used in dyeing, volcanoes, putrefaction, fireextinguishing liquids, alloys, the rusting of iron,

a proposal to use inflammable air in a public firework-display (this at the request of the police), coal measures, dephlogisticated marine acid, lampwicks, the natural history of Corsica, the mephitis of the Paris wells, the alleged solution of gold in nitric acid, the hygrometric properties of soda, the iron and salt works of the Pyrenees, argentiferous lead-mines, a new kind of barrel, the manufacture of plate-glass, fuels, the conversion of peat into charcoal, the construction of corn-mills, the manufacture of sugar, the extraordinary effects of a thunderbolt, the retting of flax, the mineral deposits of France, plated cooking vessels, the formation of water, the coinage, barometers, the respiration of insects, the nutrition of vegetables, the proportion of the components in chemical compounds, vegetation and other subjects.

Another development in Lavoisier's activities originated in this period; for in March, 1768, a few days after his nomination for the Academy, he entered the Ferme, the company of financiers who for a certain sum, fixed every six years and paid annually to the Government in advance, purchased the privilege of collecting the national taxes with, of course, the chance of profit or the risk of loss on the transaction. His entry into this association, which was ultimately to stink in the nostrils of his fellow-countrymen—the conditions of the poorest among whom he constantly bore in mind and did much to improve—and indeed to bring him to the guillotine, was prompted by the highest motives in that he sought only an opportunity to increase

the fortune left him by his mother so that he might have at his disposal more ample means for the prosecution of his scientific researches. Accordingly, he became adjoint to the Farmer-General Baudon,1 buying from the latter one third of the interest in the lease of Jean Alaterre. His friends at the Academy did not entirely approve of this, but Fontaine consoled them with the remark: "Ah, well! the dinners that he will give us will be all the better." The work of the Ferme was heavy, involving long journeys into various parts of France. But Lavoisier did not neglect his scientific work on these excursions, continuing his barometrical and mineralogical observations during his travels. He was absent from Paris for months at a time, but allowed nothing to interfere with his scientific work; for example, he was away on a tour of inspection from July 18, 1760, to January 7, 1770, and on August 9, 1770, he was again travelling and did not return to Paris until February, 1771.

In the course of his work in the Ferme, Lavoisier was soon brought into contact with the Farmer-General Jacques Paulze, whose daughter Marie Anne Pierrette, born in 1758, he married in 1771. The influential Abbé Terray, uncle to Mme. Paulze, who had died in 1761, had urged the marriage of his grand-niece, then thirteen years old, to the Comte d'Amerval, a penniless aristocrat aged fifty. But M. Paulze's refusal on the ground of differences

<sup>&</sup>lt;sup>1</sup> When Baudon died, Lavoisier became titulary Farmer-General (1779) on condition of paying Mme. Baudon a proportion of the profits for the remaining period of Baudon's interest. In 1780 a new lease to the Ferme was drawn up under Necker and Lavoisier became a Farmer-General.

in age and fortune, together with his remark that ma fille a pour lui une aversion décidée; je ne lui ferai certainement pas violence, considerably irritated the Abbé. Moreover, all blandishments about presentation at Court by the Baronne de la Garde, sister to the Comte d'Amerval, failed to convince Mlle. Paulze, who explained that she regarded the suitor for her hand as an espèce d'ogre. Paulze, indeed, now stood in some danger of losing his control of a department of the Ferme; for Terray was Controller-General of Finance. But a courageous protest from one of Paulze's colleagues prevented the removal of a talented and upright official. To avoid further possible harassment from Terray, Paulze decided to marry off his daughter as soon as possible. A marriage with Lavoisier, arranged in November, 1771, took place on December 16, 1771, in a very distinguished company that included, besides the formidable Abbé, many high state dignitaries and academicians. Lavoisier was twenty-eight and his bride was fourteen: from the time of their marriage until Lavoisier's appointment to the Régie des Poudres, they lived at a house in the Rue Neuve des Bons Enfants.1

Lavoisier's father died suddenly on September 15, 1775, at his country house at Le Bourget, a premature death that greatly grieved his son. In March of that year Lavoisier had been appointed to the Régie des Poudres and he and his wife went to live at the Arsenal, where they remained until 1792.

<sup>&</sup>lt;sup>1</sup> This house is now demolished: it stood at the corner of the Rue des Bons Enfants and the Rue Neuve, near the Banque de France.

Mlle. Punctis died in 1781 and left her fortune to the nephew on whom she had lavished years of devotion.

Lavoisier's marriage was notably a happy one. Mme. Lavoisier, alert and intelligent, rapidly equipped herself to take an interest and, indeed, to share in her husband's scientific pursuits. She learned English and translated into French many chemical memoirs, including the classic papers of Priestley and Cavendish, and Kirwan's Essay on Phlogiston. She was soon able to help Lavoisier in his experiments. Many entries in his laboratory notebooks are in her handwriting; she drew the figures illustrating his Traité Elémentaire de Chimie (1789) and two of her sketches, showing some experiments on respiration, are extant. In every sense, Mme. Lavoisier carried enough guns for her husband.

In prosecuting his various interests, Lavoisier reserved six hours every day, from six to nine o'clock in the morning and from seven to ten o'clock in the evening, for his scientific work. One day a week was entirely set apart for experiments. "C'était pour lui," wrote Mme. Lavoisier, "un jour de bonheur; quelques amis éclairés, quelques jeunes gens fiers d'être admis à l'honneur de coopérer à ses expériences, se réunissaient dès le matin dans le laboratoire; c'était là que l'on déjeunait, que l'on dissertait, que l'on créait cette théorie qui a immortalisé son auteur; c'était là qu'il fallait voir, entendre cet homme d'un esprit si juste, d'un talent si pur, d'un génie si élevé; c'était dans sa

conversation que l'on pouvait juger de la hauteur de ses principes de morale."1

As Lavoisier's fame grew, his laboratory at the Arsenal became a meeting-place for the leaders of scientific thought, not only of France but also of other countries: Priestley, Blagden, Franklin, Tennant, Watt and Arthur Young were received there at various times. In fitting up the laboratory Lavoisier spared no expense, employing his increasing fortune almost prodigally in order to obtain the best instruments that Parisian craftsmen could produce; and he allowed some of his younger contemporaries, Gengembre, Hassenfratz, Adet and others, to work there under his direction. Academicians, ministers and state officials all flocked there. Yet, while thus busily occupied, Lavoisier was also preparing reports for the Academy, giving technical advice to the government, carrying out his duties in the Ferme, acting as Régisseur des Poudres, serving on the Committees for Agriculture and for Weights and Measures, collecting further meteorological data and maintaining relations with numerous widely scattered correspondents. At the same time he was preparing the classic memoirs that revolutionized the science of chemistry. Moreover, throughout his administrative work, he displayed a spirit of great humanity, constantly being concerned with improving the well-being of his countrymen, reforming the state of agriculture, developing industry and striving to lighten the burden of taxation on the poorer classes. It appears too that he helped some of the

<sup>&</sup>lt;sup>1</sup> Grimaux, pp. 44-5.

younger scientists by providing the expenses for their researches; and that he gave considerable sums to the inhabitants of Blois and Romorantin when the bad harvest of 1788 had made corn so expensive that they could not pay the price demanded in the market. And some of those whom he helped forgot him in the day of trouble.

During the vacations of the Academy, Lavoisier found time to rest from his strenuous life either at Le Bourget or at the Château de Fréchines, which he bought in 1778 and where he carried out experiments in agriculture in the hope of improving the lot of the peasants, founded a free school and generally played the part of a kindly squire to his people. And, as Grimaux pointed out, his name nowhere appears in the chronique scandaleuse of eighteenth century France.

Before passing on to discuss Lavoisier's scientific work, a word must be said about his labours in the Ferme and in the Régie des Poudres. As a Farmer-General, his work was above reproach and he was concerned with many reforms and improvements during the years that preceded the Revolution; his administration was just and he was no robber of his fellows; and he abolished the infamous tax of pied forchu to which the Jews were subject in Clermontois. But his plan for the erection of a wall surrounding Paris, designed to prevent smuggling and therefore to protect honest traders from unfair competition, brought him much criticism. The architect who was commissioned to erect the wall

<sup>&</sup>lt;sup>1</sup> Between Blois and Vendôme, Loir-et-Cher.

spent money lavishly on magnificent barriers. The citizens of all classes objected to the arrangement as designed to put them into subjection to the Ferme. The Duc de Nivernais, Marshal of France, bluntly expressed the soldier's opinion that the originator of the scheme ought to be hanged; the wits of the day coined the mot that le mur murant Paris rend Paris murmurant; and others chanted that

"Pour augmenter son numéraire Et raccourcir notre horizon La Ferme a jugé nécessaire De nous mettre tous en prison."

An anonymous pamphlet (by de Dulaure) entitled Réclamation d'un citoyen contre la nouvelle enceinte de Paris, élevée par les fermiers-généraux (1787) contended that the Ferme was preventing the fresh air from entering Paris and stifling its citizens by preventing the dispersal of the foul exhalations constantly discharged into its atmosphere; and, moreover, put the whole blame on Lavoisier. "Tout le monde assure," it ran, "que M. Lavoisier, de l'Académie des sciences, est le bienfaisant patriote à qui l'on doit l'ingénieuse et salutaire invention d'emprisonner la capitale des Français. Après la mort de cet académicien, le confrère chargé de l'éloge du savant défunt fera charitablement de retrancher ce trait de son histoire. La Ferme peut lui élever une statue sur les murs qu'il a inventés, mais l'Académie doit rougir de sa confraternité. On rapporte qu'un

<sup>&</sup>lt;sup>1</sup> Grimaux, p. 80.

maréchal de France, le duc de N . . . . . . , á qui l'on demandait son avis sur cette enceinte, répondit : Je suis d'avis que l'auteur de ce projet soit pendu. Par bonheur pour M. Lavoisier, cet avis n'a pas encore été suivi." As Grimaux writes, this act of justice was misrepresented as arbitrary and Lavoisier was depicted as an enemy of the people. Grimaux, from a study of the evidence, concluded that from 1768 to 1786 Lavoisier profited by nearly 1,200,000 livres from the Ferme, which in comparison with other cases does not appear to be excessive. The Ferme was suppressed by decree of the National Assembly on March 20, 1701, and the business of liquidating its affairs was entrusted to six of Lavoisier's seniors, Lavoisier thus terminating a connection that only a malicious fate could ever have led him to begin.

Lavoisier's work in the Régie des Poudres is of more scientific interest than are his activities in the Ferme, however much credit is due to him in the latter for the fairness and equity of his administration and for the reforms that he introduced into a system which, towards its last days, was tending to approach the modern system of national taxation. In the provision of gunpowder for the needs of the nation, it was the story of the Ferme over again, the supply being entrusted to a company of financiers who engaged to provide a million livres of powder every year. The highest price that the company was allowed to pay for saltpetre did not admit of its profitable manufacture and the producers had to be indemnified by the Government, who moreover

had also to disburse sums to cover the costs of accidents and explosions. If the company's supply was short of the stated amount in any one year, they were not liable to make good the deficit in the following year; and in emergency they could not be called on to supply more than the amount named in the contract. The Government were thus forced in time of war to buy supplies from other countries at very high prices, a circumstance that is said to have led them to accept the Peace of Paris in 1763 concluding the Seven Years' War. The contract of the Ferme des Poudres was for six years; and the company had no motive or interest in increasing the national output of saltpetre. Moreover, the salpetriers enjoyed costly privileges that were a public burden. They had the right of free lodging and conveyance. They were allowed to dig anywhere in search of saltpetre,1 but they could be bought off-to their own profit and consequent decrease in the supply; and they could demand wood for fuel at a miserably cheap price. Thus, when all these items were considered, the cost to the nation was absurdly high, while the supply was continually decreasing, and the company, which had a monopoly of the sale of saltpetre, sporting powder, blasting powder and the low grade powder sold to other countries, was reaping a profit as high as 30% of the capital invested.

<sup>&</sup>lt;sup>1</sup> The saltpetre was formed in soil that was much trodden by cattle and impregnated with their excreta, in stables, sheep-folds, pig-pens, dovecotes, etc. The material was dug out, dried, mixed with lime and wood ashes, and lixiviated with hot water. The extract yielded saltpetre on evaporation.

Lavoisier drew up a report for Turgot on the Ferme des Poudres, recommending a complete change in the system; and in 1775 a decree replaced the Ferme by the Régie des Poudres. Four régisseurs were appointed as state officials for the manufacture of gunpowder. Of these one was Lavoisier. The régisseurs were to advance 4,000,000 livres, at 1 % above legal interest over a period of four years, to compensate the Ferme and to be repaid this as rapidly as the new system prospered, in such a way that the sum sunk in the enterprise should not exceed what was really necessary. For their services they received a fixed salary of 2,400 livres with increasing allowances on the amount of powder and saltpetre sold. The total annual income soon rose to the order of 16,000 to 17,000 livres. The burdens on the nation were removed, the state profited from the monopoly of the sale of powder and the national output of saltpetre was increased. The villages were released from compulsory cartage and allowed to bargain for lodging and wood. In 1778 the Régie obtained restriction of the right to dig1 to stables, sheep-folds and dovecotes: and yet the output increased, the villages having no more interest in buying themselves off and the salpetriers, being better paid, having every advantage in an increased output. In ten months the profits were 900,000 livres; in two years 2,000,000 livres of the advance was repaid.

But even now the supplies were not sufficient and

<sup>1</sup> Cf. MS. note by Lavoisier, Œuvres, V, p. 665.

the régisseurs had to supplement production by foreign purchases. Accordingly their plans to increase the national output were now extended with the object of developing in France such artificial nitre-beds1 as were already established in other countries and of which little was known in France. Research was necessary and Turgot, on Lavoisier's advice, instructed the Academy to announce a prize to be awarded in 1778 for an essay on the best method of making saltpetre and to draw up a detailed programme, setting out clearly all possible information relating to the formation of saltpetre with a list of works to be consulted and experiments to be tried. Without the usual delay of eight days, the Academy promptly appointed a commission, consisting of Macquer, Sage, Baumé, Montigny, d'Arcy and Lavoisier, to carry out this task. The programme was drawn up in a month and 3,000 copies were distributed; and the assistance of possible authorities was invited correspondence. All information that could be obtained was published in a Receuil de mémoires et d'observations sur la formation et sur la fabrication du salpêtre (Paris, 1776).

Meanwhile Lavoisier himself drew up an Instruction sur l'établissement des nitrières et sur la

<sup>&</sup>lt;sup>1</sup> Artificial nitre-beds were constructed by reproducing the conditions in which the nitre or saltpetre was formed in nature (see p. 45 above, footnote). Animal excreta, decaying vegetable matter and old mortar or calcareous earth were mixed in dry ditches, dug for the purpose and roofed to keep off the rain. Occasionally the material was turned over to allow air to penetrate the mass and a little water was added. After some months the mass with added wood ashes was lixiviated with hot water and saltpetre was obtained in much the same way as already described for the natural product.

fabrication du salpêtre,1 which was published in 1777 as a joint work of the régisseurs and was evidently of much value, since it was reprinted in 1794. 1777 also Lavoisier communicated to the Academy a memoir on the ash used by the Parisian saltpetre manufacturers and on its use; and together with Clouet he visited Roche-Guyon, where the Duc de la Rochefoucauld had noted the occurrence of saltpetre in the chalk, confirmed the presence of saltpetre and detected also calcium nitrate, indicated the method of extraction and recommended the most suitable methods for establishing artificial nitre-beds in the neighbourhood.

In 1778 there was enough powder in the magazines for two or three campaigns, but for all that, Lavoisier, still anxious to increase output, spent in company with Clouet and at their own expense three months in Touraine and Saintonge in search of further natural supplies: and the commissioners also began at the Arsenal experiments on nitrification that continued until 1783 under Lavoisier's direction. Details of these are given in Lavoisier's laboratory note-books.2

Meanwhile, none of the essays submitted for the prize due to be given in 1778 were considered of sufficient merit; and a larger prize of 8,000 livres was announced for 1782. This was won by the brothers Thouvenet. The commissioners were ordered by the Academy to collect and edit all the essays presented, together with other accessible

<sup>1</sup> Œuvres, V, p. 391.

Cf. Grimaux, p. 88, footnote 2, and Berthelot, p. 283.

material and a historical introduction, but almost the whole of this work fell to Lavoisier as secretary of the commission.<sup>1</sup>

In these various ways the manufacture of saltpetre in France greatly increased. In seven years it rose from 1,600,000 to 2,500,000 livres and in 1788 it had reached 3,770,000 livres. Moreover, the quality of the gunpowder was much improved; for before 1775 its range was from 136 to 155 metres, whereas it was now from 224 to 253 metres, and there were complaints in England about the superiority of the French powder. The price was low: and in 1788 the magazines contained 5,000,000 livres. In fourteen years it was estimated that a saving of 28 million livres had been effected.

Research in the refining and analysis of saltpetre continued to interest Lavoisier long after he had severed his connection with the Régie des Poudres and as late as 1793 he prepared a memoir on this subject. His interest in the manufacture of gunpowder nearly cost him his life in 1788 when an experiment with a powder containing the new substance potassium chlorate, which had recently been discovered by Berthollet, was being carried out at Essones. Lavoisier was accompanied by Mme. Lavoisier, Berthollet, Le Tort, Chevraud and Mlle. Chevraud. The workmen carrying out the mixing of the powder were protected by a wooden screen that divided them from the mortars; the visitors were looking on from the other, that is, the unprotected

<sup>&</sup>lt;sup>1</sup> Mém. Math. et Phys. . . . par divers Savans, 1786, Vol. XI. <sup>2</sup> Œuvres, V, p. 614.

side. Lavoisier remarked sharply on the danger of their position and the party moved off. A moment later the mixture exploded, and Mlle. Chevraud and Le Tort were killed, the other members of the party narrowly escaping the fate of their friends. The workmen, protected by the screen, were unharmed. Lavoisier informed the Minister of the accident of which he had nearly been the victim, moi et ce que j'ay de plus cher, adding: "Si vous daignés, Monsieur, occuper un instant le Roy du récit de ce triste événement et des dangers que j'ay courus, permettez-moi de vous prier d'assurer en même temps Sa Majesté que ma vie est à elle et à l'État et que je serai toujours prêt à en faire le sacrifice, toutes les fois qu'il pourra en résulter quelque avantage pour son service, soit pour la reprise du même travail sur la nouvelle poudre, travail que je crois nécessarie, ou de toute autre façon."1

On another occasion, this time during the troubles of 1789, Lavoisier as Régisseur des Poudres was in a danger of another kind. In the Arsenal magazines there were stored 10 milliers of poudre de traite, or export powder, a powder of low quality exported to other countries, and it was decided to send this to Essonnes and replace it with poudre à mousquet, musket powder, as likely to be more needed having regard to the state of affairs in Paris. In fact, the poudre de traite had been on its way from the Metz factory to Rouen and Nantes for shipment; but the panic-struck provincial officials smelt treachery

<sup>&</sup>lt;sup>1</sup> Grimaux, p. 93. For Lavoisier's MS. note on the explosion, see Œuvres, V, 741.

and, patriotically ensuring that the cargo did not find its way into the hands of their country's enemies, sent it to Paris to encumber the Arsenal magazines. Here the real trouble took place. No munitions of war were allowed to leave Paris without permission of the commander of the Garde Nationale, Lafayette. The Marquis de La Salle, Lafayette's chief of staff, signed the authority for the movement in his chief's absence, the powder to be loaded at Port-St.-Paul in a boat escorted by four gardes nationaux. The local inhabitants misconstrued the loading of the powder and a mob went to inform Lafavette who, in ignorance of what was going on and not troubling to find out the true state of affairs, ordered the re-magazining of the powder. Suspicion and rumour now ran their full course: the Régisseurs des Poudres were traitors, robbing the citizens of the powder and shot that were their sole means of defence. Even the escort was clapped under arrest. Next day there was ample protest, Lavoisier explaining the reasons for moving the powder and the arrested guards being released, but the assembly of the local commune felt that the public mind must be calmed and sent two representatives, de La Rozière and Franchet, to conduct an enquiry at Port-St.-Paul, where the boat was then surrounded by a mob of guards and citizens from the surrounding districts. The representatives ordered the powder to be taken back into the Arsenal and then proceeded with their examination. Four barrels, chosen at random, were opened: the contents proved to be poudre de traite. The necessary documents were now signed and all seemed in order, except the surging mob at the gates of the Arsenal demanding the arrest of Lavoisier and Le Faucheux the younger, one of the régisseurs, both of whom were therefore escorted to the Hôtel de Ville by the angry crowd. Here again Lavoisier explained the affair to the assembly, who set him and his colleagues at liberty. Still the cry of treachery to Paris arose from the mob, and de La Salle was the next quarry for their vengeance; for, had he not signed the authority? Lafayette was helpless. The crowd called for de La Salle's death. He escaped for the moment but was later arrested and spent some weeks in prison. Bailly, mayor of Paris, and Mme. Bailly on their return from Versailles at 11 o'clock that night heard of the disorder and being told that Lavoisier and his wife had been arrested went to the Hôtel de Ville to save their friends, but by midnight Lafayette had dispersed the mob. Two days later 10,000 livres of poudre à mousquet from Essonnes were magazined at the Arsenal and the public clamour died down. Lavoisier continued his work with the Régie des Poudres until 1791, his services thus extending over a period of sixteen years during which he placed the manufacture of saltpetre and the supply of gunpowder on a sound and secure footing, displaying throughout his great organizing, financial and economic abilities. From this brief sketch of some

<sup>&</sup>lt;sup>1</sup> The two other regisseurs were Le Faucheux the elder, then a feeble old man, and Le Clouet, who was still suffering from the wounds he had received on July 14, when his uniform as Captain of the Garde Nationals had led the crowd to mistake him for Delaunay, Governor of the Bastille.

of the activities of his many-sided life, we shall pass on to consider his work in chemistry, first taking some account of the state of chemical thought at a time when Lavoisier as a young chemist first entered that arena from which he was to depart one of the immortals of science.

## CHAPTER II

## LAVOISIER'S CHEMICAL HERITAGE

ATTHE TIME when Lavoisier was beginning his scientific studies at the Jardin des Plantes, chemical thought was still thoroughly mediæval: chemistry was still the mere handmaid of medicine and lagged far behind mechanics, which had already been set on its modern road nearly a hundred years earlier, when in 1687 Newton through the pleadings and at the expense of his friend Halley published his Philosophiæ Naturalis Principia Mathematica, the greatest of all books in the history of human thought. To understand and to appreciate the achievements by which Lavoisier in his later years transformed chemistry into one of the great sciences, rivalling mathematics, mechanics, astronomy and physics in its dignity and importance, helping man to a better understanding of the world around him, providing lavishly for his daily needs and contributing greatly to the progress of his civilization, we need to devote some space to consider the chemical milieu into which Lavoisier was born before detailing the steps by which he slowly effected this remarkable revolution and did for chemistry what Newton had already done for mechanics.

In the earliest civilizations, man was undoubtedly

familiar with many chemical facts; for the arts of cooking, baking, brewing, dyeing, making pottery, extracting metals from their ores and the art of that primitive scientist, the medicine-man of early times, are all essentially chemical. Notably in ancient Assyria and Egypt, these arts and crafts, including the making and colouring of glass and the working of metals, had all reached a high stage of development. But, as far as we know, there was until the time of the Greeks no attempt to explain the many extraordinary changes that matter is observed to undergo in various conditions. Then under Empedocles in the fifth and Aristotle in the fourth century B.C., there arose a theory that all things were made of varying proportions of four elements, earth, air, fire and water, the elements themselves being formed from a prime or first matter. This theory was, however, the mere speculation of philosophers who, through the class distinctions of their time, were far above debasing themselves by gaining any solid acquaintance with such valuable chemical facts as were at that time easily observable among the craftsmen labouring at those essentially chemical arts that were already established among every civilized people, including their own. When political changes later on drove the philosophers of Rome and Byzantium to seek refuge in Alexandria, there occurred a merging of Greek speculative thought and the practical chemical knowledge of the ancient Egyptians, handed down over many centuries; and here for the first time chemical theory and chemical fact met on the same

ground. Accordingly, there arose at Alexandria in the early centuries of the Christian era the first active chemical theory, namely, that one kind of matter was transmutable into another: for, since the Aristotelian theory of the four elements supposed that all things were made up of these four elements combined in various proportions, it followed that, if in a given substance these proportions were altered, then a new substance would be formed. The experimental philosophers of Alexandria therefore sought to convert base metals, such as lead, into the precious metal gold. Although these attempts invariably ended in failure, the theory was not rejected; but it was then thought that the transmutation could be effected only by some special substance, called the Philosophers' Stone; and this then became the object of these primitive researches. Another substance, the Elixir of Life, similar in its special properties to the Philosophers' Stone, was thought to be capable of curing all diseases and therefore of conferring immortality on those who possessed it: it also formed part of the search.

Such a hypothesis as we have just been describing is not so highly absurd as it sounds to modern ears. It was indeed far from absurd in the early centuries of this era. There were many well-known and thoroughly authenticated facts to support it. For example, if water was evaporated, an earthy residue was left at the bottom of the vessel when all the water had been driven off, just as "fur" is left in a kettle; at the time we are speaking of, this was

plain evidence that water was convertible into earth, since it was not suspected that the so-called "earth" might have been present already in solution in the water or might have been formed by the action of the water on the material of the evaporating vessel. This alleged change was not finally disproved until 1770 in experiments carried out by Lavoisier. Again, when lead from certain sources is heated for a very long time, a small residue of silver is left; here again was evidence of transmutation, since what was originally lead had now become silver, the experimenters of that time being unaware that some lead ores are argentiferous and that their lead had already contained the silver into which they supposed it to have been transmuted by the long-continued action of the fire. Many facts similar to these were known, and it is not surprising that the Alexandrian chemists believed in the transmutability of matter: it would be much more surprising if they had not.

This science, which had already been named chemeia, passed with the passing of political power to the Arabs, under whom it acquired the name of al-kimia, which subsequently became alchemy when the Arabic power passed and Arabic culture penetrated Western Europe. But, both under the Arabs in their day and among the peoples of Western Europe during Renaissance times, the Aristotelian theory of the four elements and its logical consequence, the transmutability of matter, completely ruled chemical thought and guided chemical experiment. The thirteenth, fourteenth and

fifteenth centuries were the great ages of alchemy in the West; and Aristotelian theory found no serious critic until the rise of that wayward genius, Paracelsus (1493-1541), whose strident tongue in the early part of the sixteenth century called chemists to a new service, urging them to forgo their search for the Philosophers' Stone and the Elixir of Life and to seek instead new chemical, that is, mineral as opposed to vegetable, medicines. Two results followed. Firstly, the change of emphasis from the Philosophers' Stone and the Elixir of Life to the preparation of physicians' drugs reduced chemistry to becoming the mere handmaid of medicine; and during this period, often referred to as the iatro-chemical age, chemistry would hardly deserve to be called a science, if it were not for the few enthusiastic workers, such as Glauber, Libavius and van Helmont, gifted with some wider vision than their contemporaries. Secondly, in turning the attention of chemists in a new direction, Paracelsus, while still accepting the four elements of Aristotle as the ultimate elements of all bodies and believing accordingly in the transmutability of matter, taught that their more immediate constituents were the tria prima or three principles, salt, sulphur and mercury not, of course, the substances ordinarily known by these names, but principles or elements resembling those substances in their general properties; and, while a change from four to three elements in the composition of a world has not much significance, what was important was the partial

break with a theory and tradition that had lasted for nearly two thousand years. The older and the newer beliefs, however, lived on side by side.

Therefore, when the Hon. Robert Boyle (1627ai) in the middle of the seventeenth century turned to study the chemical thought of his day, he found it necessary to take account of two opinions, each closely resembling the other; and he criticized both of them severely in his Sceptical Chymist (London, 1661). In this work Boyle first of all disposed of the widely accepted belief that fire resolved all bodies into their ultimate elements, pointing out that by the action of fire bodies were resolved sometimes into less than three, and sometimes into more than four, other substances, and arguing that these products were rather new compounds, formed by re-arrangement of the component particles of the original substances, than the true elements of bodies. He then pleaded for a closer relation between experimental fact and theoretical speculation and, after an extensive and detailed criticism of current chemical ideas, formulated his famous definition of the chemical element, namely, that a chemical element is a substance that analysis cannot resolve into anything simpler. In actually defining the element, Boyle wrote that he "must not look upon any body as a true Principle or Element, but as yet compounded, which is not perfectly Homogeneous, but is further Resoluble into any number of Distinct Substances how small soever" (op. cit., p. 236), adding in a later passage: "And, to prevent mistakes, I

must advertize You, that I now mean by Elements, as those Chymists that speak plainest do by their Principles, certain Primitive and Simple, or perfectly unmingled bodies; which not being made of any other bodies, or of one another, are the Ingredients of which all those call'd perfectly mixt Bodies are immediately compounded, and into which they are ultimately resolved " (ibid., p. 350). Boyle's definition is still the basis of modern chemical theory; and Boyle, as being the first to propound this definition clearly, has been hailed as the "Father of Modern Chemistry." But, since he was unable to devise experiments to apply or illustrate his definition, which thus remained a mere sterile intellectual analysis, and since he was therefore unable to draw up any list of elements, it seems to us that there is more justice in applying the title to Lavoisier, who, as we shall see below, by applying Boyle's definition drew up the first table of the chemical elements and erected, partly on this definition and partly on the discoveries of his time, the modern system of chemistry.

Boyle's criticism, despite what is frequently stated by historical writers, had little or no effect on the progress of chemical thought: for, in one guise or another, the four elements and the three principles lived on for another century and even in 1750 chemistry was still actively concerned with the medicinal properties of substances. This neglect to apply the fruits of Boyle's genius is largely due to the rise of another theory, the

phlogiston theory, propounded in its earliest form only a few years after Boyle published his *Sceptical Chymist* and rejected only when Lavoisier formulated his theory of combustion, which received its most complete expression in his classic *Traité Elémentaire de Chimie*, published in 1789, a work that is properly to be regarded as the *Principia* of chemistry.

The phlogiston theory was the current theory of chemistry when Lavoisier began his chemical studies; and, since the theory which was accepted in its place was the entire handiwork and the undivided property of Lavoisier and since his contribution to chemistry can only be understood by reference to the system that his labours overthrew, it is necessary for us to consider that system briefly.1 The phlogiston theory originated with Johann Joachim Becher (1635-82) and was greatly extended by Georg Ernst Stahl (1660-1734). Becher's main contribution to the theory appeared in his Physica Subterranea (Frankfurt, 1669), in which he expressed the view that all sub-terrestrial or inorganic bodies were compounded of varying proportions of four elements, namely, water and three different earths, one of which earths, terra pinguis, oily or combustible earth, he supposed to be present in all combustible bodies and to be expelled from them on combustion, combustion and the action of fire on bodies being then and previously, as we have seen when discussing Boyle's criticisms, looked upon as leading to the

<sup>&</sup>lt;sup>1</sup> A fuller account is given by J. H. White, The History of the Phlogiston Theory, London, 1932.

decomposition of those bodies into their simpler constituents. Stahl extended these views, more particularly in Specimen Beccherianum (Leipzig, 1703), Fundamenta Chymiæ (Nuremberg, 1723) and Exberimenta Observationes, etc. (Berlin, 1731), and during the eighteenth century until Lavoisier developed his attack almost all the leading chemists of Europe accepted this theory. Thus they generally supposed that all combustible bodies contained an extremely subtle matter, an inflammable principle or element, which, following the example of Stahl, they called phlogiston. When a body was burnt, its phlogiston was not destroyed but escaped into the atmosphere. The more combustible a body was, that is, the less ash it left on being burnt, the more phlogiston it must have contained originally. Therefore, such substances as oils, fats and charcoal contained a high proportion of phlogiston in their composition. These could in certain conditions restore to a burnt body the phlogiston that it had lost in the process of burning. Thus, for instance, the calces of metals, produced by heating the metals in air, were bodies that had lost their phlogiston. It was known that these metallic calces could be re-converted to the metals from which they had been formed by heating them with charcoal; and it was therefore concluded that the phlogiston of the charcoal had, by its addition to the calces in this process, re-constituted the metals, so that a metal was a compound of its calx and phlogiston. It was, however, equally well known that, when a metal was calcined, its weight increased and that the calx produced was heavier than the metal from which it had been formed. Yet, in the current theory, the metal had suffered a loss of phlogiston on calcination. In some places it is stated that the phlogistic chemists explained this increase of weight accompanying a loss of phlogiston as due to phlogiston possessing the property of levitation rather than gravitation, but White (op. cit., p. 82) has shown that this explanation received scant acceptance. The gain in weight was generally explained as Boyle had explained it, in his Essays of Effluviums (London, 1673), as the result of the fixation in the metals of material particles of fire which, since they were able to traverse the pores of glass, could produce the increase of weight even when the metals were calcined in sealed glass vessels.1 Moreover, in these Essays Boyle had disputed the contention that in calcination a part of the metal was expelled and only its earth and fixed salt left behind. "These notions," he wrote, "are not well fram'd, and do not universally hold," as was to be seen in his experiments. "For," he added, "it does not appear by our Tryals, that any proportion, worth regarding, of moist and fugitive parts was expell'd in the Calcination; but it does appear very plainly, that by this Operation the Metals gain'd more weight than they lost." He noted further that he had frequently obtained lead by merely exposing its calx to the action of heat, so that it was evident that calcination had

<sup>&</sup>lt;sup>1</sup> A detailed analysis of Boyle's studies on calcination has been made by the author in *Science Progress*, 1934, 29, 253.

not deprived it "of the suppos'd radical moisture requisite to a Metal." But Boyle's sound contention was forgotten by the dogmatic phlogistic chemists of the succeeding century, although his explanation of the cause of the gain in weight of metals on calcination was accepted in one form or another almost until the time when Lavoisier propounded that accepted nowadays, and his observation of the conversion of a calx into a metal by the action of heat alone without the addition of phlogistic matter passed unheeded, being re-discovered by Bayen in 1774.1

Meanwhile, before Boyle had published his criticisms and before the phlogiston theory had been formulated, the iatro-chemist Johann Baptista van Helmont (1577-1644) had in his posthumous Ortus Medicinæ (Leyden, 1648) reverted to a form of a much older theory, originally propounded by Thales of Miletus in the sixth century B.C., that all things were made from water, van Helmont recognizing only one exception, in that he did not suppose that air was formed from water. And he had argued from an experiment, which Singer has pointed out was described earlier by Nicholas of Cusa (c. 1430) in his De Staticis Experimenti, that water was transformable into wood; and, since he had already converted wood into ashes and gas (a term which he was the first to use). he found much to confirm his revival of an ancient hypothesis. The experiment to which we refer

<sup>&</sup>lt;sup>1</sup> See p. 195 below.

<sup>2</sup> See E. J. Holmyard, Chemistry to the Time of Dalton, London, 1925, p. 54, footnote 1.

## LAVOISIER'S CHEMICAL HERITAGE 6

and which had much influence on seventeenthand eighteenth-century chemists was described by van Helmont thus: "But I have learned by this handi-craft operation, that all Vegetables do immediately, and naturally proceed out of the Element of water onely. For I took an Earthen Vessel, in which I put 200 pounds of Earth that had been dried in a Furnace, which I movstened with Rain-water, and I implanted therein the Trunk or Stem of a Willow Tree, weighing five pounds; and at length, five years being finished, the Tree sprung from thence, did weigh 169 pounds and about three ounces: But I moystened the Earthen Vessel with Rain-water, or distilled water (alwayes when there was need) and it was large, and implanted into the Earth, and least the dust that flew about should be co-mingled with the Earth, I covered the lip or mouth of the Vessel, with an Iron-Plate covered with Tin, and easily passable with many holes. I computed not the weight of the leaves that fell off in the four Autumnes. At length, I again dried the Earth of the Vessel, and there were found the same 200 pounds, wanting about two ounces. Therefore 164 pounds of Wood, Barks, and Roots, arose out of water onely" (Ortus Medicinae, Leyden, 1648, p. 109; English trans. by J. Chandler, Oriatrike, London, 1662, p. 109). This experiment provided ample confirmation of the transmutability of matter, especially in an age when such eminent chemists as Becher and Stahl were still taking a lively interest in the search for the Philosophers' Stone.

Thus, in the middle of the eighteenth century, the scientific mind was still in the grip of some form or other of the ancient belief in a world composed of a small number of elements, in the interconvertibility of the various forms of matter and in the existence of a fire-element. The complaint voiced in 1712 by John Freind, Professor of Chemistry at Oxford, was still justified. "We own," he wrote, "Chymistry has made a very laudable Progress in Experiments; but we may justly complain, that little Advances have been made towards the Explication of 'em. The Stock of these Materials is very large and splendid, but the Rationale of them is still intirely wanting. The Three, or as others will have it, the Five Principles of Paracelsus, the Alkali and Acid of Tachenius, are perhaps not openly acknowledg'd, but are at least too much allow'd of by our modern Chymists, who in words may disapprove of that way of Reasoning, but mean the same thing in other Terms. Nor is there one to be found, among so great a Number of these Writers, who does not fall into those very Hypotheses, which he condemns in others; or at least there is not one, who has laid down such Fundamental Principles of Chymistry, upon which a just and rational Explication may be built. For in reality they have not Examin'd what the true Nature and Mechanism of Bodies is, but have only Describ'd it such as they would have it to be; insomuch that they have assign'd Laws and Properties to Bodies, that agree neither to the Rules of Mechanism, nor yet with themselves. No Body has brought more Light into this Art than Mr. Boyl, that famous Restorer of Experimental Philosophy: Who nevertheless has not so much laid a new Foundation of Chymistry, as he has thrown down the old; he has left us plentiful Matter, from whence we may draw out a true Explication of things, but the Explication it self he has but very sparingly touch'd upon." (Chymical Lectures, London, 1712, pp. 2-4)

Outside the great corpus of chemical fact that had been accumulated through many centuries of practical experience and beyond Boyle's definition of the chemical element, which had fallen dead from his hands nearly a hundred years earlier, there was, as the first half of the eighteenth century sped on its way, nothing to indicate that the science of chemistry was not still in the darkness of mediævalism. There was less mysticism and greater clarity of expression in its literature, but the ancient beliefs resisted all challenge, remained essentially the same and, while perhaps outwardly changing their garments, curiously wove into those fabrics the multitudes of new facts that were continually being discovered. Nature was still supposed, by the chemists at least, to be extraordinarily simple.

But in 1754 there appeared the first gleam of the dawn; for in that year the young Joseph Black (1728-99), in presenting his inaugural dissertation at the University of Edinburgh for the degree of M.D., advanced an explanation of the differences between the mild and caustic alkalis that, slowly but inevitably winning acceptance, gave a completely new turn to chemical thought. The origins

of this work, De Humore Acido a Cibis Orto, et Magnesia Alba (Edin., 1754), subsequently expanded in 1755 into Experiments on Magnesia Alba, Quicklime, and some other Alcaline Substances and published a year later in Essays and Observations, Physical and Literary, Read before a Society in Edinburgh (Edin., 1756, II, p. 157), are curious. Walpole, the Prime Minister, suffering from the stone, thought that a remedy invented by Mrs. Joanna Stephens had brought him relief; and Mrs. Stephens, receiving a reward of £5,000, parted with her secret which was published in the London Gazette on June 19, 1739. The published recipe read as follows:

"My medicines are a Powder, a Decoction, and Pills. The powder consists of Egg-shells<sup>2</sup> and Snails,<sup>3</sup> both calcined. The decoction is made by boiling some Herbs<sup>4</sup> (together with a Ball, which consists of Soap,<sup>5</sup> Swines'-Cresses, burnt to a Blackness, and Honey) in water. The Pills consist of Snails calcined, Wild Carrot seeds, Burdock seeds, Ashen Keys, Hips and Hawes, all burnt to a Blackness, Soap and Honey."<sup>6</sup>

<sup>&</sup>lt;sup>1</sup> A translation by Crum Brown was recently published by Dobbin (J. Chem. Education, 1935, 12, 225 and 268).

<sup>&</sup>lt;sup>2</sup> Egg-shells and Snails calcined in crucible surrounded with coal for 8 hours. Then left in earthenware pan to slake in a dry room for 2 months. The Shells thus become of a milder taste, and fall into powder.

<sup>&</sup>lt;sup>3</sup> Snails left in crucible until they have done smoking, then rubbed up in a mortar. Take 6 parts of Egg-shell to 1 of Snail-powder. Snails ought only to be prepared in *May*, June, July and August.

<sup>&</sup>lt;sup>4</sup> Herbs of decoction: Green Chamomile, Sweet Fennel, Parsley and Burdock; leaves or roots.

<sup>&</sup>lt;sup>5</sup> Soap: Best Alicant Soap.

<sup>&</sup>lt;sup>6</sup> Quoted from Sir William Ramsay, The Life and Letters of Joseph Black, M.D., London, 1918, p. 22.

This remedy had considerable caustic properties. The "Powder" was evidently lime. Medical opinion, particularly that of Cullen, the distinguished professor of medicine at Glasgow, was very properly against the use of such caustic remedies on account of their severe action on the body tissues. In fact, at this time the solvents recommended and used for urinary calculi were all caustic and included not only lime-water, or a solution of quick-lime, but also caustic potash and caustic soda, the soap-boiler's ley. Black, studying under Cullen at Glasgow during the years 1749 to 1751, was therefore led to experiment on magnesia alba in order to find out if it could be applied as a milder lithontriptic than the caustic alkalis then in use. The caustic alkalis were prepared from the mild alkalis (the carbonates of modern times) by boiling their solutions with slaked lime, which was prepared by adding water to the quicklime produced by "burning" or heating limestone. This causticization of the mild alkalis by means of quicklime was a process of some antiquity; and it was thought, firstly, that the quicklime acquired its causticity by the absorption of some kind of igneous matter from the fire in the process of "burning," and, secondly, that, in the causticization of the mild alkalis by boiling them with slaked lime, this igneous matter was transferred from the lime to the mild alkalis which thereby were rendered caustic. This, as is evident, is a thoroughly phlogistic explanation, as might be expected at that time.

Black therefore set out to study the properties of

the substance magnesia alba in the hope of discovering a new sort of lime and lime-water. The magnesia alba had been obtained by Friedrich Hoffmann (1660-1743), Professor of Medicine in the University of Halle, by adding alkali to mother of nitre, the bitter liquor left after the crystallization of saltpetre from the water used to extract the latter from nitrous earths, or by calcining the residues left after evaporation of mother of nitre. He also prepared it in similar ways from the residual liquor left after the evaporation of waters containing common salt. Black first prepared his magnesia from bittern, the bitter saline liquor left in the pans after sea-water has been evaporated in the process of extracting common salt from it, but finding this not always easy to procure, he then prepared it by adding a solution of pearl ashes (potassium carbonate) to a solution of Epsom salt, "which," he wrote, "is separated from the bittern by crystallization, and is evidently composed of magnesia and the vitriolic acid," and, aware that the precipitate of magnesia alba then produced contained some "vitriolated tartar" (potassium sulphate), formed by the combination of the acid and the alkali, he added a large quantity of hot water to the precipitate, agitated the mixture, allowed the magnesia to settle, decanted off as much of the water as possible and then washed the precipitate many times with cold water to ensure complete removal of the "vitriolated tartar." "The first affusion of hot water," he wrote, "is

<sup>&</sup>lt;sup>1</sup> Alembic Club Reprint No. 1, p. 7. All references given below to Black's work refer to this reprint.

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intended to dissolve the whole of the salt, and the subsequent additions of cold water to wash away this solution."1

Black then found that the magnesia dissolved in acids "with violent effervescence, or explosion of air"; and, after some other simple experiments that do not concern us here, he then proceeded to find out "whether this substance could be reduced to a quick-lime." He heated an ounce of it in a crucible for an hour: it lost 7 of its weight. The residue dissolved in acids and gave salts exactly as did the original substance before it was heated, "but what is particularly to be remarked," he wrote, "it is dissolved without any the least degree of effervescence": and it liberated volatile alkali from sal ammoniac on boiling, but it had no action on the neutral solution of calcareous earths in acids or on lime-water, whereas the original substance precipitated calcareous earths from such solutions and lime from lime-water. "Observing magnesia to lose such a remarkable proportion of its weight in the fire," wrote Black, "my next attempts were directed to the investigation of this volatile part." Three ounces of magnesia were then heated strongly in a glass retort and Black collected in a receiver attached to the retort a small amount of slightly acid water that did not sensibly effervesce on the addition of acids. The magnesia had lost more than the half of its weight, but still effervesced with acids, though not so strongly as it did before it was heated. He concluded correctly that the magnesia had not been heated to a sufficiently high temperature to ensure its complete calcination. "But even from this imperfect experiment," he wrote, "it is evident, that of the volatile parts contained in that powder. a small proportion only is water; the rest cannot, it seems, be retained in vessels, under a visible form.1 Chemists have often observed, in their distillations, that part of a body has vanished from their senses, notwithstanding the utmost care to retain it; and they have always found, upon further inquiry, that subtile part to be air, which having been imprisoned in the body, under a solid form, was set free and rendered fluid and elastic by the fire. We may therefore safely conclude, that the volatile matter, lost in the calcination of magnesia, is mostly air; and hence the calcined magnesia does not emit air, or make an effervescence, when mixed with acids "2

In the next experiment Black calcined two drams of magnesia in a crucible: it was reduced to 2 scruples and 12 grains. The residue was dissolved "in a sufficient quantity of spirit of vitriol" and from this solution the magnesia was precipitated by the addition of alkali. The precipitate was washed and dried; it weighed I dram and 50 grains; it had recovered all the properties it had lost by calcination, including the property of effervescing with acids and had "acquired besides an addition of weight nearly equal to what had been lost in the fire; and, as it is found to effervesce

<sup>&</sup>lt;sup>1</sup> The phrase "cannot, it seems, be retained in vessels, under a visible form" is taken from van Helmont. Cf. p. 79, footnote 1, below.

<sup>2</sup> A.C.R., No. 1, p. 16.

LAVOISIER'S CHEMICAL HERITAGE 73 with acids, part of the addition must certainly be air."

Black added: "This air seems to have been furnished by the alkali from which it was separated by the acid; for Dr. Hales has clearly proved, that alkaline salts contain a large quantity of fixed air, which they emit in great abundance when joined to a pure acid. In the present case, the alkali is really joined to an acid, but without any visible emission of air; and yet the air is not retained in it: for the neutral salt, into which it is converted, is the same in quantity, and in every other respect, as if the acid employed had not been previously saturated with magnesia, but offered to the alkali in its pure state, and had driven the air out of it in their conflict. It seems therefore evident, that the air was forced from the alkali by the acid, and lodged itself in the magnesia."2

Black then gradually added dilute oil of vitriol to 2 drams of magnesia contained in a flask of known weight until the magnesia was completely dissolved. He noted the weight of the acid used and from the figures found that the magnesia "lost 1 scruple and 16 grains by the ebullition." Another 2 drams of magnesia were first strongly calcined, being thereby reduced in weight to 2 scruples and 12 grains, and then subjected to the same process. Almost exactly the same amount of acid was required for the solution of the calcined magnesia as had been necessary to dissolve the uncalcined material, "and no weight was lost in the experiment."

<sup>1</sup> Ibid., p. 17. <sup>2</sup> Ibid.

"We now perceive the reason," wrote Black, "why crude and calcined magnesia, which differ in many respects from one another, agree however in composing the same kind of salt, when dissolved in any particular acid; for the crude magnesia seems to differ from the calcined chiefly by containing a considerable quantity of air, which air is unavoidably dissipated and lost during the dissolution.

"From our experiments, it seems probable, that the increase of weight which some metals acquire. by being first dissolved in acids, and then separated from them again by alkalis, proceeds from air furnished by the alkalis. And that in the aurum fulminans, which is prepared by the same means, this air adheres to the gold in such a peculiar manner, that, in a moderate degree of heat, the whole of it recovers its elasticity in the same instant of time; and thus, by the violent shock which it gives to the air around, produces the loud crack or fulmination of this powder. Those who will imagine the explosion of such a minute portion of fixed air, as can reside in the aurum fulminans, to be insufficient for the excessive loudness of the noise, will consider, that it is not a large quantity of motion communicated to the air, but rather a smart stroke which produces sound, and that the explosion of but a few particles of fixed air may be capable of causing a loud noise, provided they all recover their spring suddenly, and in the same instant.

"The above experiments lead us also to conclude, that volatile alkalis, and the common absorbent earths, which lose their air by being joined to acids, but shew evident signs of their having recovered it, when separated from them by alkalis, received it from these alkalis which lost it in the instant of their joining with the acid."

"Black here made an enormous stride," wrote Ramsay<sup>2</sup>; "he had weighed a gas in combination." We may note, too, in passing, that Black's advance had been made without isolating the "fixed air" and that he had recognized the existence of this substance and rigorously and quantitatively demonstrated by means of the balance the part it played in the constitution of the mild alkalis.

Black now extended these results generally to the calcareous earths and the mild alkalis, arguing that in their ordinary condition these substances contained a large quantity of fixed air, separable from them only by the agency of fire, and this in some cases only incompletely, and by the action of acids. "I also imagined," he wrote, "that, when the calcarious earths are exposed to the action of a violent fire, and are thereby converted into quicklime, they suffer no other change in their composition than the loss of a small quantity of water and of their fixed air. The remarkable acrimony which we perceive in them after this process, was not supposed to proceed from any additional matter received in the fire, but seemed to be an essential property of the pure earth, depending on an attraction

<sup>&</sup>lt;sup>1</sup> Ibid., pp. 19-20. The reference in this passage to the probability that air from the alkalis provided the increase of weight observed in metals, when they were dissolved in acids and then precipitated by alkalis, may well have helped Lavoisier into the confusion discussed in Chapter IV below.

<sup>&</sup>lt;sup>2</sup> Op. cit., p. 25.

for those several substances which it then became capable of corroding or dissolving, which attraction had been insensible as long as the air adhered to the earth, but discovered itself upon the separation."

An unseen breach had at last been effected in the stubborn defences of the phlogiston theory; the causticity of lime was now shown not to depend on the absorption of matter from the fire.

Combination with fixed air therefore rendered quicklime mild; and the removal of fixed air from calcareous earth produced the caustic substance, quicklime. When lime-water was added to a solution of a mild alkali, the fixed air left the latter and combined with the lime, which was thus rendered mild and precipitated, while the alkali, now deprived of its fixed air, was rendered caustic.

In a later experiment Black rapidly slaked 8 grains of finely powdered quicklime, freshly prepared from chalk, in 2 drams of boiling distilled water and at once threw the mixture into 18 ounces of distilled water. A slight sediment, weighing about one-third of a grain, subsided. Other experiments gave a like result, but Black had expected a much larger quantity of sediment, produced by the combination of the dissolved lime with the air that was known to be invariably contained in water. To discover whether or no water retained this air when saturated with lime, he prepared a saturated solution of lime in water and he then placed 4 ounces of this in one

<sup>&</sup>lt;sup>1</sup> A.CR., No. 1, pp. 22-3.

vessel and 4 ounces of common water in another vessel of the same size under the receiver of an airpump: "and, upon exhausting the receiver, without heating the vials, the air arose from each in nearly the same quantity: from whence it is evident, that the air, which quick-lime attracts, is of a different kind from that which is mixed with water. And that it is also different from common elastic air, is sufficiently proved by daily experience; for lime-water, which soon attracts air, and forms a crust when exposed in open and shallow vessels, may be preserved, for any time, in bottles which are but slightly corked, or closed in such a manner as would allow free access to elastic air. were a vacuum formed in the bottle. Ouick-lime therefore does not attract air when in its most ordinary form, but is capable of being joined to one particular species only, which is dispersed thro' the atmosphere, either in the shape of an exceedingly subtile powder, or more probably in that of an elastic fluid. To this I have given the name of fixed air, and perhaps very improperly; but I thought it better to use a word already familiar in philosophy, than to invent a new name, before we be more fully acquainted with the nature and properties of this substance, which will probably be the subject of my further inquiry."1

Here Black had made a very striking and important advance, as important indeed as that which he had already achieved with regard to the constitutional difference between the mild and caustic alkalis; for, by showing that the "air," to which this difference was due, was "different from common elastic air," he had for the first time in the history of chemistry differentiated an "air," or as we would now say a "gas," from the ordinary air of the atmosphere. This differentiation of a gas from ordinary air was to have a great influence on Lavoisier's work, as we shall see later. For the moment, it will be more appropriate for us to consider at this point certain studies that had already been carried out on the presence of air in bodies and to what extent these studies had progressed before Lavoisier entered upon his classic researches.

Van Helmont had pointed out that coals, although strongly heated by means of a furnace, did not burn if contained in a closed vessel, but were consumed if the vessel was open. "Every coal," he wrote, "... although it be roasted even to its last day in a bright burning Furnace, the Vessel being shut, it is fired indeed; but there is true fire in the Vessel, no otherwise than in the coal not being shut up; yet nothing of it is wasted, it not being able to be consumed, through the hindering of its efflux. Therefore the live coal, and generally whatsoever bodies do not immediately depart into water, nor yet are fixed, do necessarily belch forth a wild spirit or breath. Suppose thou, that of 62 pounds of Oaken coal, one pound of ashes is composed. Therefore the 61 remaining pounds, are the wild spirit, which also being fired, cannot depart, the Vessel being shut, I call this Spirit, unknown hitherto, by

the new name of Gas, which can neither be constrained by Vessels, nor reduced into a visible body. . . . "1

This passage marks the first use of the word gas in the literature of chemistry. Van Helmont's suggestion of the use of this term passed unheeded and his successors in pneumatic chemistry wrote of "airs," "different kinds of air," "factitious airs" and so on, gas not coming into its proper use until Lavoisier and Macquer re-introduced it one and a half centuries later.

The particular gas with which van Helmont was concerned in the above-mentioned case, namely, in the combustion of charcoal, he called spiritus sylvestris or gas sylvestre. He noted that it was produced also in the fermentation of grapes—its presence, he thought, made wines sparkling and effervescent—in the action of vinegar on the shells of certain fish (i.e., the action of acetic acid on carbonates), in caverns, mines or grottoes (as in the Grotto del Cane near Naples), in mineral waters, such as those of Spa, or in intestinal putrefaction. He called it gas sylvestre sive incoercibile, because it could not be kept in the closed vessels in which it was produced but shattered them in pieces (sylvestris, wild, savage). But, in the state of chemistry in his time, van Helmont failed to distinguish this gas from the gases produced by the solution of silver in aqua fortis (nitric oxide), by the burning of sulphur (sulphur dioxide), by the action of aqua fortis on

<sup>1</sup> Op. cit., p. 106; English trans., p. 106. The original passage defining Gas reads: Hunc spiritum incognitum hactenus, novo nomine gas voco, qui nec vasis cogi, nec in corpus visibile reduci potest.

sal ammoniac (hydrochloric acid gas) and by the action of heat on saltpetre (oxygen and oxides of nitrogen), and he named all of these gas sylvestre. However, he noted also the existence of gas pingue, obtained from the large intestine and by the fermentation of animal excreta; and he found that this was inflammable, while gas sylvestre extinguished a flame; and, although he called the gas formed by the action of aqua fortis on sea-salt or sal ammoniac by the name of gas sylvestre, he also referred to it as gas salium—"There was produced," he wrote, "even in the cold, a gas the release of which shattered the vessel." Here was the labour and the difficulty: for van Helmont recognized that these breakages, which must have been well known in chemistry for centuries, were due to the production of gases, of material bodies in a state in which they could not be restrained within sealed vessels, and yet he was unable to devise any method for their collection or isolation.

Later, Boyle's studies, described in his New Experiments Physico-Mechanicall, Touching The Spring of the Air, and its Effects, etc. (Oxford, 1660), had emphasized a growing interest in the mechanical properties of air; and van Helmont's primitive and somewhat imprecise indication of the existence of different kinds of gases was lost in the impressive array of orderly experiments set out in the various editions of Boyle's experimental and unspeculative treatise. Yet, when Boyle himself published an extensive work dealing with the "air" obtained from ale, fermenting bread, flesh, fruits and

vegetables, he observed that his product, which he called "factitious Air," was "noxious to the life of Animals," and "prejudicial to Fire" (A Continuation of New Experiments Physico-Mechanical, Touching the Spring and Weight of the Air, and their Effects. The Second Part, London, 1682). The significance of these observations did not apparently strike Boyle; and he seems to have regarded his "factitious Air" as a mere variety of common air, which is not to be wondered at since it was a well-known fact that common air could easily be brought, by respiration or burning, into a condition in which it was fatal to life and fire.

This suggestion about Boyle's notion of his "factitious Air" appears all the more reasonable in the light of the later writings of Newton and Hales. In the famous Queries that he appended to his Opticks, Newton, who found that Nature "seems delighted with transmutations" and had adopted atomistic views about the structure of matter, held that "Dense Bodies by Fermentation rarify into several sorts of Air "1 and that the particles expelled from bodies by heat and fermentation, once they were beyond the reach of the body's attraction, "take up above a Million of Times more space than they did before in the form of a dense Body. Which vast Contraction and Expansion seems unintelligible, by feigning the Particles of Air to be springy and ramous, or rolled up like Hoops, or by any other means than a repulsive Power. The Particles of Fluids which do not cohere too strongly,

<sup>&</sup>lt;sup>1</sup> Opticks, 4th edn., London, 1730, p. 349.

and are of such a Smallness as renders them most susceptible of those Agitations which keep Liquors in a Fluor, are most easily separated and rarified into Vapour, and in the Language of the Chymists, they are volatile, rarifying with an easy Heat, and condensing with Cold. But those which are grosser, and so less susceptible of Agitation, or cohere by a stronger Attraction, are not separated without a stronger Heat, or perhaps not without Fermentation. And these last are the Bodies which Chymists called fix'd, and being rarified by Fermentation, become true permanent Air; those particles receding from one another with the greatest Force, and being most difficultly brought together, which upon Contact cohere most strongly. . . ."1

Hales, continuing Boyle's work and much impressed with Newton's comments and having observed that air was "plentifully inspired" by vegetables, heated a great variety of substances, animal, vegetable and mineral, and collected and measured the amounts of air given off in this way, including also the "air" released in various chemical actions. But, to Hales, as apparently to Newton and in a sense to Boyle, it was all "air"; and he was unaware that he had prepared a number of chemically distinct gases, disposing of his products without examining them once he had measured the amounts produced (Vegetable Staticks, London, 1727). Hales supposed that the air, the presence of which in bodies he had so amply proved, acted as a kind of cement or "bond of union" between

<sup>&</sup>lt;sup>1</sup> Ibid., pp. 371-2.

the ultimate particles of solids and in so functioning conferred on them their characteristic coherence.

From the position taken up by Hales, Black, as we have seen, had made a decisive advance by recognizing the existence of a gas, "fixed air," chemically different from common air. The next gas to be thus differentiated was "inflammable air," hydrogen. The production of an inflammable gas by the action of dilute spirits of salt on iron had been known from the time of Boyle (New Experiments, touching the Relation betwixt Flame and Air, London, 1672, p. 64). To Boyle the product was merely a kind of fume of a sulphureous nature, sulphureous because it was inflammable. In 1766, however, the Hon. Henry Cavendish (1731-1810) studied the two "factitious airs," namely, Black's "fixed air" and this other "air," to which he gave the name of "inflammable air." Cavendish measured the densities of fixed and inflammable airs and the solubility of fixed air in water. Thus two "airs" or gases had now been differentiated from the common air of the atmosphere.

This, in broad outline, was the state of chemistry in the period from 1765 to 1770, when Lavoisier was embarking on his researches and when, in Vernon Harcourt's apt description, the phlogiston theory, "at first but a conjectural attempt to generalize the phenomena of combustion," had

<sup>1</sup> Phil. Trans., 1766., 56, 141.

<sup>&</sup>lt;sup>2</sup> Presidential Address, British Association, Birmingham Meeting, 1839 (B.A. Report, 1839, p. 8).

"gradually made itself a coat of patch-work out of the successive discoveries of half a century, and arrived at playing as many feats in philosophy, as the harlequin in a pantomime."

## CHAPTER III

## EARLY RESEARCHES: THE NATURE OF WATER, ITS ALLEGED CONVERSION INTO EARTH. THE COMBUSTION OF THE DIAMOND

LAVOISIER had become interested in the nature of water through the shortage in the watersupply of Paris. Déparcieux1 had suggested the use of water from the river Yvette and had developed a detailed scheme for carrying out his proposal,<sup>2</sup> a scheme that appears to have had the support of the Academy. After the death of Déparcieux in 1768, Lavoisier, who was known to be much interested in the problem, was called on by the Academy to report on various other suggestions<sup>3</sup> and to defend the plan of Déparcieux when it was criticized by the Carmelite Father Félicien of St. Norbert.4 The proposal to use the waters of the Yvette brought up the question of their purity, and this at a time when the methods of chemical

<sup>&</sup>lt;sup>1</sup> Antoine Déparcieux (1703-68).

<sup>&</sup>lt;sup>2</sup> Mém. Acad. R. Sci., 1762, p. 337; 1766, p. 149; 1767, p. 1.

<sup>3</sup> Mém. Acad. R. Sci., 1771, p. 17; Œuvres, III, pp. 221, 255 and IV,

<sup>&</sup>lt;sup>4</sup> Lavoisier's reply to these criticisms, read to the Academy in July, 1769, was printed in the *Mercure de France* for October, 1769 (Œuvres, III, p. 208).

analysis were very unsatisfactory. Lavoisier, as we have noted in describing his tour with Guettard, had used sensitive hydrometers, thus preferring to test the purity of water by determining its density; and he had presented two memoirs on hydrometry to the Academy in 1768,<sup>1</sup> and he had determined the density of distilled water at various temperatures.<sup>2</sup> Altogether the subject of water, its purity and its density was, as Meldrum pointed out,<sup>3</sup> one that Lavoisier was greatly interested in.

When Déparcieux read his memoir of 1766 at the Academy, the question was raised as to whether or no the examination of water by evaporation to dryness involved error through the conversion of some of the water into earth. In 1767 Le Roy argued with regard to this that earth was essential to the constitution of water and that it was some of this constituent earth, not transmuted water, that was left behind in the distillation of water. Le Roy's memoir was not published in the Mémoires but an account of it was given, in the Histoire,4 the introductory section printed at the beginning of each annual volume of the Mémoires. This account. written by Defouchy, throws light on the contemporary attitude of the Academy to the doctrine of the four elements. It reads: "In ancient times four elements were recognised, that is to say, four primitive and stable materials which entered more or less into the composition of all substances; these primitive materials were air, water, earth and fire:

<sup>&</sup>lt;sup>1</sup> Œwres, III, pp. 427 and 145. <sup>2</sup> Œwres, III, pp. 156, 438, 451; II, p. 774. <sup>3</sup> Isis, 1933, 19, 330. <sup>4</sup> Hist. Acad. R. Sci., 1767, p. 14.

this idea has been adopted by nearly all physicists. We say nearly all because there certainly can be found some moderns who have rejected it and maintained that these substances, that were taken to be primitive and unalterable elements, were themselves composed of other substances and could change their form. Their aim, above all, is to show that the water which we have at hand, so to say, more than any other elementary substance, could be converted into earth and consequently is neither simple nor stable. One can readily see that this assertion, if it were true, would upset all accepted ideas and would destroy, beyond recovery, all the certainty that one can expect of chemical analysis, because one could never be sure that the substances obtained by the decomposition of a mixture, instead of being the materials that were in it, were not the results of the process. Those, however, who have impugned the unalterability of water, if one may make use of this term, have not done so without the support of some weighty reasons."1

The position therefore was that the unalterability of water had not been proved. And Defouchy added, in a passage that clearly appreciates the constant problem of chemistry in obtaining pure substances, that those who thought that water could be freed from the earth that Le Roy argued was necessary to its constitution "suppose that, after a certain number of distillations, all the earth that might be associated with the water may be separated from it; but nothing is more gratuitous than this

<sup>&</sup>lt;sup>1</sup> Meldrum's translation (*Isis*, 1934, 20, 398).

supposition; it is supremely difficult to obtain single substances, and chemists continually meet with this difficulty: we have shown, moreover, that some parts of earth and even of glass may be fine enough to be carried off with the molecules of water to which they are adherent and that consequently it is impossible that distillation should completely separate one from the other." The problem now bore down on Lavoisier; for, in his work on hydrometry, he had proposed to take the density of distilled water as his standard of reference and doubt was here cast upon its constancy. Accordingly, Lavoisier set out to determine the effect of repeated distillations on the density of water and communicated his results to the Academy in two memoirs entitled On the Nature of Water and on the Experiments Alleged to Prove its Transmutability into Earth.2 The first gave a historical survey of the work of previous investigators and the second described the author's own experiments.

In the historical memoir Lavoisier stated that the question of the transmutability of one element into another, particularly of water into earth, was of too great an interest and had attracted too many distinguished authors for him to neglect to give some account of the work previously carried out on it: and, he added, in a sentence that admirably instances his approach to scientific problems, he did not propose to deal with the views of the philosophers of former centuries about the elements,

<sup>&</sup>lt;sup>1</sup> Translated by Meldrum (Isis, 1934, 20, 398). <sup>2</sup> Mém. Acad. R. Sci., 1770, pp. 73 and 90.

since they threw little light on the question, but that he wished to deal with facts—je veux parler des faits. The experiments alleged to prove this transmutation, Lavoisier pointed out, were of two kinds. They either referred to the growth of vegetables supplied with water only, or they dealt with continued distillations or other chemical manipulations of water. With regard to the former, he referred to van Helmont's experiment, of which we have already taken account, and to similar experiments reported by Boyle (The Sceptical Chymist, London, 1661, pp. 106-12; and The Origine of Formes and Qualities, 2nd edn., Oxford, 1667, pp. 164-9) and by others. From the observations reported by these workers, it might perhaps rightly be concluded that the earth in which plants grew was merely an accidental circumstance attending vegetation and that it contributed nothing of its own substance to the growth of the plants or the formation of their solid parts; and it could even be supposed that vegetables effected a real transmutation of water into earth. But, argued Lavoisier, some clearer proof was necessary; for, although it was recognized that the combination of two substances could produce a third substance with entirely different properties from either of the original substances, the conversion of a mass of water, without addition to or loss of its substance, into a mass of earth ought to be admitted only after the most stringent experimental tests, more especially because, in the growth of vegetables, it was a question of the change of water not only into earth but also into the various

other proved constituents of plants, namely, oils, resins, juices, etc., and thus into a number of substances other than earth. Moreover, most of the experimenters had used common water, spring water or river water, which were known to contain calcareous earth, selenite and other salts. Among those who turned to other sources, van Helmont had used rain-water or distilled water without describing what steps he had taken to ensure that the rainwater was pure, while it was now known that, even when the greatest precautions were taken in its collection, it contained salt, admittedly in a quantity far too small to account for the weight of 164 pounds gained by van Helmont's willow, but it might be doubted whether all this weight was due to earth. Eller had, admittedly, used distilled water, but he had found only 7 to 8 grains of earth, and it might be that this small amount had been derived from the substance composing the vessel in which the water was contained, a possibility to be made clear presently by experiments. Further, the work of Hales and others had shown that plants absorbed through their leaves a considerable amount of atmospheric air, which it was known contained various volatile substances. Indeed, Bonnet had described the air as a fertile terrain, whence the leaves of plants drew abundant nourishment of all kinds. And the experiments of Hales had proved that air entered largely into the composition of vegetable substances. Thus the plants that had been grown in water only might have derived the

<sup>&</sup>lt;sup>1</sup> Mém. Acad. Berlin, 1746, p. 46.

principles discovered in them by analysis from two sources, firstly, from water itself and the small proportion of earth proved to be contained in all the kinds of water used by previous investigators, and secondly, from the air and the substances of all kinds with which it was charged. Thus the experiments with plants did not in any way prove the conversion of water into earth.

Turning to the chemical experiments alleged to prove this change, Lavoisier pointed out that Borrichius, after obtaining common salt and an earthy powder containing some proportion of sulphur from the separate evaporation of rainwater, snow-water and hail-water in glass vessels, had subjected a sample of water to a large number of repeated distillations and claimed that by continuing these operations the whole of the water could be changed into earth. Boyle, apparently unaware of this work, had reported that he had been informed of similar experiments, in which the same sample of water had been distilled 200 times in a glass alembic with the production of an insoluble earth (Origine of Formes, etc., 1667 edn., pp. 258-62). Boerhaave had opposed these conclusions, arguing that the earth found in these experiments had been derived from the dust that continually floated in the air. But Geoffroy1 had distilled the same sample of water twenty times in well sealed apparatus and constantly found that earth was produced: and Marggraf<sup>2</sup> in a memoir on the earth found in pure water had decisively removed all possibilities of its

<sup>&</sup>lt;sup>1</sup> Mém. Acad. R. Sci., 1738, p. 208. <sup>2</sup> Mém. Acad. Berlin, 1756, p. 20.

having come from atmospheric dust, obtaining the same earth by distilling water in a glass retort sealed to its receiver and further showing that the same result was obtained by subjecting water to strong agitation in closed vessels. Then Le Roy, as we have noted above, argued in 1767 that the earth was originally contained in the water in intimate combination and that a little of it was separated in each distillation, which had thus led to the false belief that the water had been changed to earth. Lavoisier thought that if this were so, repeated distillations ought gradually to remove the earth, the amount of which ought to diminish successively with each distillation, and that there ought to be a limit beyond which it was not obtainable, whereas in the experiments reported by Boyle and Marggraf earth was still produced from the water after repeated distillations. There was thus, wrote Lavoisier at the conclusion of the historical memoir, considerable doubt and uncertainty about the origin of the earth produced in the distillation of water.

Before going on to describe Lavoisier's own work on this problem it is interesting to note that, when discussing the experiments named above on the growth of vegetables and the possibility of air playing a part in such process, Lavoisier argued that the lower layers of the atmosphere, in which plants grow, were undoubtedly highly composite and that it was probable that the air that formed the basis of the atmosphere was not a simple substance, not an element, as the older philosophers supposed. The

appearance of the opinion that air was not an element, in a memoir dated 1770, suggests that it was added at a later date before the printing of the memoir in 1773. Later, in dealing with Lavoisier's work on calcination, we shall see that it was only slowly and with great difficulty that he arrived at a clear idea of the composition of air.

In the experimental memoir, Lavoisier explained that he had originally begun this research with the object of determining what degree of purity water could be brought to by repeated distillations and what changes it underwent during these operations; but that his experiments insensibly led him further than he had expected and, indeed, to a decisive result on the problem of whether, as former philosophers and some contemporary chemists thought, water could be changed into earth. Accordingly, he collected a quantity of rain-water in large glass or glazed earthenware vessels, in an open space at a distance from trees and buildings, after the rain had been falling for some time and had therefore removed from the air the various foreign bodies that continually floated in it. Tests with the hydrometer showed that this water was slightly heavier than water from the River Seine that had been distilled once; and evaporation of a large sample indicated that it contained small proportions of a light insipid earth and sea-salt, the base of the latter being partly earthy and partly fixed alkali. The water collected

<sup>&</sup>lt;sup>1</sup> This suggestion has been confirmed by Meldrum, who has shown that no trace of such opinions occurs in either of the MSS. prepared for the Academy in 1769 and 1770 (*Isis*, 1934, 20, 410-11).

<sup>2</sup> A mixture of calcium and sodium chlorides.

in the receiver from this evaporation was still slightly heavier, very slightly heavier, than the distilled water from the Seine. Here, Lavoisier noted that, since the rain-water had been distilled in glass and the Seine water in metal apparatus, the rain-water had apparently dissolved some of the material of the vessel, the probability of which would be evident from the later experiments.

The distilled rain-water was now re-distilled eight times and its specific gravity determined at the end of each distillation against that of the distilled Seine water as standard. Lavoisier was surprised to find that, although a quantity of earth was separated in each distillation, the specific gravity of the water did not sensibly decrease, or, at least, its decrease was not at all in proportion to the amount of earth separated. From this result, argued Lavoisier, one of two things must follow. Either the earth separated in the distillation was such that it could be held in solution in water without increasing its specific gravity, or, at least, without increasing it as much as other substances did; or else it was not in the water when its specific gravity had been determined but had been formed during the distillation, in short, it was a product of the operation. To decide the question, the best method appeared to be to repeat the same experiments on weighed amounts of water in hermetically sealed vessels of known weight; for, if it was the matter of fire that passed through the glass and combined with the water to form the earth, an increase in the total weight

of the water, earth and vessel should be observed after a large number of distillations. Such an explanation, the passage through glass of material particles of fire, had, as we have noted above, been already applied by Boyle to explain the gain in weight of metals on calcination and was generally accepted. Boyle had experimented mostly with open vessels; but, in the few experiments which he attempted to make with sealed vessels, he weighed the metal before heating it and weighed the calx after extracting it from the vessel, and in no case did he weigh the sealed apparatus before and after heating, as Lavoisier now proposed to do in the problem under consideration. Moreover, Lavoisier here noted that an increase in the total weight of the apparatus and its contents would not occur if the earth was produced from the water or from the vessel in which the water was contained; but that, if the earth was produced from one of these sources, there would be a decrease in the weight of one of them exactly equal to the weight of the earth separated.

To carry out his proposed experiment, Lavoisier needed an apparatus that was at once an alembic and a receiver; and he knew that it would be difficult to make, and certainly fragile. However, realizing that what he proposed to do, namely, to distil water many times on itself, closely resembled what the alchemists had called "cohobation," he resorted to the apparatus that they had used for that process, an apparatus that in those more romantic days of chemistry had been picturesquely named a "Pelican."

Lavoisier rightly thought that long cohobation in a vessel of this construction would be equivalent to repeated distillation. He therefore arranged to have one made of a suitable size with an opening at the top fitted with a well-ground glass stopper: and he bought from Chemin, ajusteur de la Monnaie, an official of the Mint, a very sensitive balance, the most accurate then known, made by Chemin

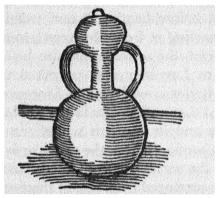


Fig. 1. A Pelican. (From J. French, The Art of Distillation, London, 1653, p. 27.)

himself. This would turn with less than I grain when carrying a load of 5 to 6 livres.

The pelican was carefully washed with some of the distilled water, dried and weighed, the recorded weight being the mean of two weighings, actually of two double weighings in each of which the pelican was weighed first in one pan of the balance and then in the other and a mean value taken. A quantity of the water that had been distilled eight times was poured into the pelican, which was then warmed on a sand-bath, care being taken to withdraw the stopper from time to time in order to permit the escape of some of the included air, the expansion of which by the heat might otherwise cause a breakage of the apparatus. When Lavoisier was satisfied that the air had expanded sufficiently, he inserted the stopper firmly in the pelican, which was then removed from the sand-bath and afterwards weighed on the same balance as before. The stopper was then sealed into the pelican by applying a coating of thick lute, made from clay, boiled linseed oil and amber, and covering this with a wetted bladder, bound into position with many turns of stout thread. The apparatus was now put back on the sand-bath, being set at such a depth that the level of the water in the pelican stood sufficiently above the level of the sand outside for any changes that might occur during the course of the experiment to be easily observable.

All was now in readiness on October 24, 1768: and it is perhaps justifiable to remind ourselves at this point what sort of enterprise Lavoisier was here engaged upon. His method, as we have already seen, aimed at tracing down the source from which the earth was produced by examining the change in detail by means of the balance. It was a method that had been initiated by Black with striking success in his study of magnesia alba and other alkaline substances; and Black's influence on Lavoisier is more than obvious in this place. Lavoisier was originally and primarily concerned GL.

here with testing the reliability of water as a standard substance in hydrometry. But there were other, more ultimate consequences. The result might show that the earth produced in such distillations had come from the water, in which case an ancient theory would be supported; it might extend Boyle's explanation to another field, and in this event, if fire combined with water to give earth, there would be another buttress for that theory; and it might prove that the earth came from the vessel and not from the water or from the combination of fire-matter with water, a result that would disprove a scientific belief held in one form or another for over two thousand years and dismiss the transmutability of matter from all serious scientific consideration.

A six-wicked lamp, constantly fed with olive oil and trimmed regularly every twelve hours, was lit and set below the sand-bath on October 24, 1768: it was kept burning there for a hundred and one days, the temperature of the water in the pelican being thus maintained during that period at from 60° to 70° on Réaumur's scale. Lavoisier watched the experiment very carefully for the first few days, but nothing remarkable appeared to be happening. He began to give up hope of its turning out successfully, when on November 20, he noticed some small particles circulating very rapidly in the water in considerable amount, but so fine as to be nearly imperceptible. A lens showed them to be very fine,

<sup>&</sup>lt;sup>1</sup> In the original and in the collected works this reads December: it should obviously be November from what follows.

thin, irregularly shaped flakes of greyish earth. These gradually grew larger, but from December 15 to 20 they appeared to increase neither in size nor in number. Presently they began to settle at the bottom of the vessel and at the end of January there appeared to be none circulating in the water.

On February 1, 1769, Lavoisier, satisfied that a considerable amount of earth had now collected and fearing the possibility of an accident that might rob him of the result of this long experiment, decided to discontinue the heating. The lamp was extinguished and, when the apparatus had cooled, Lavoisier carefully removed the covering and lute from the pelican and re-weighed it without removing the stopper. Its weight was identical with what it had been before the heating, the quarter of a grain of difference being within the limits of precision of the balance. Thus, the continuous digestion of the water for over a hundred days had produced neither increase nor decrease in the total weight of the apparatus. Lavoisier then tried to unstopper the pelican, none too easy a task since the air inside was now at reduced pressure. However, immediately he had succeeded in raising the stopper sufficiently to establish communication between the air outside and the air inside, a whistling noise was heard as the external air rushed in in very considerable quantity. This proved that air had not entered the vessel during the progress of the experiment, which had thus been carried out precisely as if the vessel had been hermetically sealed.

Since the total weight of the apparatus had shown

no increase, it was clear that neither the matter of fire nor any other external body had penetrated the substance of the glass and combined with the water to form the earth: but it remained to be determined whether the earth had been produced by the destruction of a part of the water or a part of the glass; and all that was therefore now necessary, as Lavoisier pointed out, after the precautions he had taken, was to determine whether it was the vessel or the water that was contained in it that had suffered a decrease in weight. Accordingly, he emptied the water and the earth from the pelican into a glass flask, carefully dried the pelican and weighed it. It had lost 17th grains in weight. It was evident therefore that the earth that had appeared in the distillation had been produced by the dissolvent action of the water on the glass vessel. To make the proof complete, it remained to compare the weight of the earth produced with the decrease of weight of the pelican; for, if the earth had come from the latter, these weights should be equal. Lavoisier therefore carefully dried and weighed the earth deposited by the water at the bottom of the vessel: it weighed only 40 grains, whereas the pelican had lost 174 grains of its mass. But Lavoisier had already suspected that the water still held a quantity of earth in solution. Tests with the hydrometer confirmed his suspicions, since its specific gravity was greater than that of distilled Seine water. Accordingly, Lavoisier poured the water into a glass alembic and evaporated it. In the last stages of the evaporation, he transferred the water to a glass dish and carried the evaporation to dryness. The residue of earth weighed 15% grains. Thus the total earth produced was 20th grains, or 3 grains more than the loss of weight of the pelican. Reflecting on possible sources of the additional 3 grains, Lavoisier realized that, when the water was removed from the pelican, it had been poured into a glass vessel and subsequently distilled in a glass alembic; and neither operation could have occurred without the dissolution of some small particles of the substance of both these vessels; and, moreover, it was also probable that a small amount of water had combined with the earth in its crystallization1 and had helped to increase its weight. The excess of 3 grains was therefore apparently explicable, not by an extraneous hypothesis, but by the very result established by the data of the experiment.2

The research proved, apart from the results originally sought, that water was not transmutable into earth and that the earth produced by the evaporation or distillation of water came from the substance of the glass vessels used in these operations. Lavoisier had thus, in rigorous quantitative experiments, carried out on the model set up by Black, refuted a firmly established but false theory

<sup>1</sup> He had already shown that part of the residue was saline.

a This explanation does not now bear scrutiny; for, Dobbin, having pointed out that the alkali—soda or potash—and the lime, dissolved from the glass, would absorb carbon dioxide and thus increase the weight of the residue, Meldrum calculated that such an increase would be of the order of 3 grains and concluded that this was the predominant cause of the excess weight detected by Lavoisier (Isis, 1934, 20, 404). In confirmation of this, it might be pointed out that, when Lavoisier dropped a little of the earth into acid, a slight effervescence was produced.

that had held even his distinguished contemporaries in its grip. Moreover, he had shown also that pure water could be obtained by distillation once or twice from a metal vessel and that it could therefore be used as a standard substance.

Lavoisier's numerical results may be set out as follows:

			Livre	s Onces	Gros	Grains1
Weight of the empty pelican, October 24, 1768	•		1	10	7	21.50
Weight of pelican and water,						
October 24, 1768 .	•		5	9	4	41.50
Weight of water .			3	14	5	20.00
Weight of pelican and water,						
February 1, 1769 .			5	9	4	41.75
Weight of pelican alone,						
February 1, 1769.			I	10	7	4.12
Loss of weight of pelican		•				17.38
Weight of earth						20.40

The general conclusions, as set out by Lavoisier, were as follows:

- 1. That most, if not all, of the earth separated from rain-water by evaporation was due to the solution of the material of the vessels used in collecting and evaporating the water;
- 2. That rain-water contained barely  $\frac{1}{20}$  of a grain of sea-salt per livre and could therefore be regarded as highly pure for most chemical operations;
- 3. That the difference in specific gravity of rainwater, Seine water and spring-water, distilled

<sup>1 1</sup> livre=16 onces; 1 once=8 gros; 1 gros=72 grains. As a comparison with modern weights, 1 livre=0.93 English lb.
2 Determined in the experiments mentioned on p. 93 above.

once only, and water that had been distilled eight times was almost insensible, so that springwater, Seine water or rain-water distilled once or twice from a metal alembic at a moderate heat could be regarded as absolutely pure for even the most refined chemical work;

- 4. That water neither changed its nature nor acquired any new property by repeated distillation and was far from being reducible, as Stahl had supposed, to such a tenuity that it could pass through the pores of glass;
- 5. That glass was soluble in water and that, as in the case of all salts, there was a limit or saturation point beyond which no further solution was possible; and,
- 6. Finally, that the earth obtained from water by earlier experimenters was nothing but glass produced by the evaporation, so that the experiments on which such investigators had founded their opinions, far from proving the convertibility of water into earth, led on the contrary to the conclusion that water was unalterable.

There was, however, an unresolved discrepancy in the results; for the earth obtained was infusible, or at least it was infusible in a fire that was more than hot enough to melt the hardest glass, whereas, since it indubitably came from the glass, it ought to have the essential property of the latter. Lavoisier could not explain this, but proposed to carry out

further experiments. And an apparent but slight decrease in the density of the distilled water as compared with the standard (distilled Seine water) Lavoisier ascribed, in terms of his general explanation, to an increase in the density of the latter through its solvent action on the substance of the glass vessel in which it was kept.

The memoir describing the results of these experiments was read by Lavoisier to the Academy at the rentrée publique of November 14, 1770. But the experiments had been completed early in 1769 and the memoir was initialled by the Secretary of the Academy on May 10, 1769. This and the historical memoir appeared in the volume of the Mémoires for 1770, which was published in 1773. Meanwhile, an anonymous Dissertation describing the experiments had appeared in the newly published Observations in August, 1771 (Introduction, I, 78). Meldrum,2 from an examination of the two MS. versions that Lavoisier prepared for reading to the Academy and from other circumstances, concluded that the delay of eighteen months from May 10, 1769, to November 14, 1770, was due to the

¹ The problem had already been solved by Scheele previous to 1768, but his work was not published until 1777 (Chemische Abhandlungen von der Luft und dem Feuer, Upsala, 1777). Scheele made a qualitative, but not a quantitative, study of the earth formed by the continued boiling of water in glass vessels. Separate examination of the water and the earth showed that the former gave ammonia with sal ammoniae and a precipitate with vitriolic acid and metallic solutions, turned syrup of violets green and gelatinized in air, while the latter "behaved like silex, mixed with very little lime" (Collected Papers of Carl Wilhelm Scheele, trans. by L. Dobbin, London, 1931, pp. 88-9). Lavoisier, however, may probably have observed the infusibility of the 4 th trains of insoluble matter without carrying out any further test on the 15½ grains obtained by evaporation: the former was silica, insoluble and infusible, but the latter contained both silica and alkali and was fusible.

§ Isi, 1934, 20, pp. 405-6.

fact that Lavoisier's memoir placed Defouchy, the Secretary of the Academy, in a difficulty because Le Roy's memoir of 1767, on which Lavoisier's was a great improvement, was not yet published, because Lavoisier at the age of twenty-six was claiming the solution of a long-standing problem that had defeated scientists of the highest reputation, and because there were discrepancies to account for in Lavoisier's results; and moreover Lavoisier wished to read his memoir at a rentrée publique, as Le Roy had done. Defouchy apparently got over the problem of printing Le Roy's memoir by giving it instead an unusually lengthy notice in the Histoire (1767, pp. 14-22). But Lavoisier was anxious to get his work published; and Meldrum<sup>1</sup> showed, from a comparison of the anonymous Dissertation with the MS. read at the rentrée publique, that it was highly probable that Lavoisier came to an understanding about the matter with Rozier, the editor of the Observations, and was thereby able to obtain publication of his results in 1771.

In the next year, 1771, Lavoisier submitted to the Academy some calculations and observations on a proposal to establish a steam-pump for supplying Paris with water (Mém. Acad. R. Sci., 1771, p.17): and in 1772 he presented a memoir on the use of spirits of wine in the analysis of mineral waters (ibid., 1772, II, p. 555).

A more important development in Lavoisier's chemical interests occurred in 1772; for in that year he reported to the Academy the results of certain

<sup>1</sup> Ibid., pp. 407-9.

experiments carried out on the diamond in collaboration with Macquer and Cadet in the latter's laboratory. The results were read to the Academy on April 29, 1772, and published in the May issue of the Observations (Introduction, II, 108). The apparent destruction of the diamond when strongly heated had been established by various workers from Boyle onwards; but it was now disputed whether this was a real evaporation of the diamond into vapour or a kind of combustion resembling that which occurred with phosphorus or a mere decrepitation into minute particles. The first experiments showed that diamonds, heated very strongly in a retort for three hours, showed a decrease in their weight and lost their polish. But the jeweller Maillard, convinced that the diamond would not evaporate if it was out of contact with air, offered three diamonds to the experimenters on the condition that, while the stones might be subjected to as great a heat as possible, he himself was to be allowed to protect them from contact with the air. The proposal was accepted. Maillard therefore placed the diamonds in a clay pipe filled with powdered charcoal. The pipe was closed with lute and put into a crucible, which was covered with chalk and then luted inside a device consisting of one crucible inverted over another. When the heating had gone on for two hours, it was felt that the furnace was not very satisfactory and Macquer suggested the use of the furnace in which he had succeeded in fusing various refractories. This was therefore

brought to Cadet's laboratory and the diamonds were subjected to violent heating in it for another two hours. After the apparatus was cool, the crucibles were taken out: they had been completely deformed by the heat and the chalk and the earth of the lute had vitrified. The pipe was fused into a solid mass with the melted substances. To recover the diamonds, the fused mass had to be broken up: and when this was done, the diamonds were recovered unchanged in weight or in polish, except for a slight superficial darkening. The diamond therefore was volatile only in air: and in that case its disappearance was due either to a kind of combustion or to its reduction into a very fine powder. Which of these explanations was correct, the experimenters proposed to decide by other tests.

These other tests were carried out in 1772 jointly by Lavoisier, Macquer, Cadet and Brisson. The first account of them was read to the Academy by Macquer on September 14, 1772, and appeared in the Observations in the December following (Introduction, II, 612). It appears from this report that Cadet and Brisson had obtained from the Academy permission to borrow from the Academy's collection the great lens of Tschirnhausen, formerly in the possession of the Duc d'Orléans, and previously used by Homberg and later by Geoffroy to study the action of heat on various metals placed at its focus, and to use this instrument to make a general, thorough and orderly experimental study of such phenomena as Homberg and Geoffroy had

merely begun to investigate.1 Further, the Academy invited Macquer and Lavoisier to collaborate with Cadet and Brisson and specially enjoined the two former to attend to the order and completeness of the work. The experimenters were given permission to carry out their studies in the Jardin de l'Infante so that they might take full advantage of the noonday sun, and a room in the Louvre was placed at their disposal for the necessary instruments and materials. The Comte de la Tour d'Auvergne lent them at the same time a second lens, also by Tschirnhausen, of the same diameter, namely, thirty-three pouces, as that owned by the Academy and with a much shorter focal length. The experiments were carried out in the month of August, 1772. From the brief account given in the Observations, it appears that the experimenters reported few details to the Academy, that they wished to repeat and vary most of their experiments, of which they had made a large number, and that Trudaine had offered them the use of a lens which was then under construction and which would be much larger and more powerful than any lens then existing. Trudaine's lens was however not available until 1774.

Meanwhile Lavoisier, Macquer, Cadet and Brisson carried out other experiments on the destruction of the diamond, and Lavoisier, in 1772, presented to the Academy two memoirs, one historical and the other experimental, on the

<sup>&</sup>lt;sup>1</sup> Burning-lenses provided experimenters with heat at much higher temperatures than other contemporary means afforded.

destruction of the diamond by fire.1 The second of these memoirs described the various experiments on the destruction or evaporation of the diamond in the focus of the great lens of Tschirnhausen, in which it was established that when a diamond, suitably supported in a bell-jar of air confined over water, was heated by means of the lens, the diamond lost in weight, the air was diminished in volume (by 8½ in 60 pouces) and gave a precipitate of chalk on the addition of lime-water. Similar experiments over mercury gave the same result, there being of course in this case no decrease in the volume of the confined air until the lime-water was added. The diamond was thus similar in its properties to such combustibles as charcoal, candles. spirits of wine, ether, etc., the air in which any of these had been burnt thereby acquiring the property of precipitating lime-water. It was now clear that the diamond was a combustible body and that in combustion it was "reduced" to fixed air.

The second volume of the Mémoires for 1772 did not appear in print, however, until 1776 and it is evident that Lavoisier made additions to the work between its presentation and its publication; for his laboratory note-books give clear evidence that some of the experiments were carried out in October 1773,3 and the memoir itself includes an experiment dated August 14, 1773. Lavoisier's laboratory note-book for the period from August, 1773, to March, 1774, has been lost, but a single sheet

<sup>1</sup> Mem. Acad. R. Sci., 1772, II, pp. 564 and 591. Published in 1776.

2 Lavoisier wrote à peu près prouvé (nearly proved).

<sup>8</sup> Berthelot, p. 270.

inserted in a later note-book refers to an experiment with the diamond carried out on October 22, 1773, which is actually described in the memoir of 1772. Further, the views expressed in the preceding paragraph belong to a date even later than 1773, since the memoir was not published until 1776, that is, until a time when, as we shall see below, Lavoisier had carried his ideas on combustion a considerable stage further. This work, although bearing the date 1772 in the Mémoires of the Academy, must be regarded as achieving its final form in 1776. We shall see in the next chapter how far Lavoisier was, even at the end of 1773, from holding such clear views as those expressed in the memoir considered above.

These experiments on the burning of diamonds did, however, initiate an important phase in Lavoisier's chemical researches; for in the year 1772 he had already been studying this problem of combustion with Macquer and Cadet, their preliminary report, as shown above, being read to the Academy on April 29, 1772, and a further account being given on September 14, 1772. Later in the year, Lavoisier directed his attention to the combustion of phosphorus with results that we shall describe in the next chapter.

## CHAPTER IV

## COMBUSTION AND CALCINATION: THE OPUSCULES PHYSIQUES ET CHYMIQUES

TO THE HISTORIAN of chemistry, whose duty it is to follow in detail the stages through which Lavoisier's thought passed as he gradually and laboriously re-ordered the tangle of chemical data into a coherent and consistent theory, the phase we are now about to consider bristles with difficulties: and a correct account of it can be given only by careful attention to Lavoisier's own notes and memoranda, his memoirs as read to the Academy in their unrevised form and as printed much later after revision, the papers in the Observations, the contents of his Opuscules Physiques et Chymiques (Paris, 1774) and other sources. And here opportunity cannot be forgone to pay tribute to the scholarly and accurate study of this period of Lavoisier's chemical activities already carried out by Meldrum, on whose work, as all who are familiar with it will recognize, the contents of this chapter largely depend.

On September 10, 1772, Lavoisier wrote in a note-book:

"Experiments on Phosphorus on September 10, 1772. I bought from M. Mitouard an ounce of good German phosphorus for 45 louis cost of manufacture. I put a small piece of it in a bottle. The phosphorus became luminous. It fumed but without becoming hot. I brought it near the fire. It was instantly kindled with crackling. The medicine bottle was not broken. Emboldened by this success, I wanted to find out in the same apparatus whether phosphorus absorbed air in burning. I bound a bladder with a very tight thread to the neck of the bottle into which I had previously introduced 15 grains of phosphorus. I had made another little hole at the top of the bladder and I had expressed all the air the hole in the bladder having been well" (the rest is missing, the following page in the note-book being blank1).

This brief note, as far as we know, marks Lavoisier's first important step in the study of combustion. The experiments on the diamond, reported by Macquer to the Academy on September 14, 1772, and described in the preceding chapter, do not include any observation about the function of air, while Lavoisier's own memoir of 1772, also discussed in the preceding chapter, includes the observation that fixed air was produced in the combustion of the diamond. This memoir, as we have already seen, appeared however in the second volume of the *Mémoires* for 1772, which was published

<sup>&</sup>lt;sup>1</sup> Translated from the original, first published by Meldrum in 1932 (Archeion, 1932, 14, 15). As the original is roughly drafted, a crude translation as given above seems justified here.

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in 1776, and had certainly been revised in the meantime; and moreover, as we have also seen, some of the experiments described in the published memoir belong, not to 1772, but to 1773. The note of September 10, 1772, on phosphorus is therefore of considerable interest, although the observation that phosphorus fumed and became luminous was not new, nor did Lavoisier suppose it to be. What is of importance is that Lavoisier, by some train of thought not now apparent, decided to find out whether phosphorus absorbed air in burning.1 He went on to study this problem and presented two notes to the Academy, the first being presented on October 20, 1772, and the second, a sealed note, being deposited with the Secretary of the Academy on November 1, 1772, and subsequently opened and read at the Academy on May 5, 1773.

The note of October 20, 1772, reads as follows:

"Memoir on the acid of Phosphorus and on its combinations with different substances, saline, earthy and metallic. If phosphorus is exposed to the open air, there continually rises from it an emanation or smoke, slightly visible by day, luminous in the dark. This vapour is nothing but a small part of the acid combined with much phlogiston, and if it could be collected successfully under a glass bell or any other apparatus it would be recognised as a volatile acid spirit of phosphorus.

"Contact with the open air is essential to this operation, because the vapour of the phosphorus in

<sup>&</sup>lt;sup>1</sup> Hales had already shown that phosphorus absorbed considerable amounts of air on burning (op. cit., p. 169).

being converted into a volatile acid spirit of phosphorus absorbs a small part of the air, so that it is proved that air enters naturally into the composition of this compound and that it is combined in it and fixed in it in the same way as happens in a great number of chemical combinations.

"If instead of allowing the phosphorus to be consumed in the open air it is put dry and without water in a closed vessel of small capacity. Then if a degree of heat a little higher than boiling water is imparted to the phosphorus, it burns quietly giving a beautiful flame accompanied by a thick smoke. The phosphorus is decomposed. The phlogiston leaves it. A very large amount of air is absorbed and combines with the white vapour.

"If the vapour or smoke is collected by means of a bell or otherwise, a kind of white sublimate is obtained which is nothing but the acid of phosphorus in an absolute degree of concentration and nearly the same as glacial oil of vitriol. These flowers or sublimate are resolved in a few hours, and merely by the moisture of the air, into a very strong odourless acid which has almost the same appearance as oil of vitriol.

"A singular phenomenon is that the quantity of acid obtained from the phosphorus by this last operation is greater in weight than that quantity of phosphorus which has produced it. This increase of weight, the proportion of which it is not easy to ascertain exactly, comes from the combination of the air which is fixed in this operation.

"The whole of the phosphorus is not decomposed by the combustion. There always remains at the bottom of the crucible a small part of it that no longer takes fire. It is of a yellow iron-rust colour. This small part is nothing but a phosphorus that has lost a part of its inflammable principle. To obtain it in its former state it is only necessary to distil it with some inflammable materials.

"I shall describe on this occasion the method I have used to obtain a large quantity of the acid of phosphorus. This method has no inconvenience other than being long and tedious. However, it is sure and the dissipation of the acid is almost nil.

"I took a large dish of glazed earthenware in the centre of which I placed a small agate crucible. And I covered the dish with a large glass bell. I had previously introduced into the bell a little distilled water in order that the vapours might condense more easily. I then put a small piece of phosphorus in the agate crucible and I ignited it with the point of a knife heated in the flame of a candle. This phosphorus in burning gave a very thick white vapour which circulated in the bell but it passed outside only a very little because, a pretty considerable quantity of air being absorbed in this operation, the external air that accordingly entered the bell to replace it made the vapours flow back inside instead of making them go out.

"It needed about an hour to fix the whole of the vapours, after which I began again, but it was necessary at the end of a few experiments to remoisten the bell either with distilled water or with weak acid.

"At the end of each combustion there remains at the bottom of the crucible a small amount of phosphorus of a yellow iron-rust colour such as has been described above.

"This method of obtaining the acid of phosphorus is nearly the same as that employed to make oil of vitriol. It is observed for sulphur as for phosphorus that, if it is heated a little and if it is burnt slowly, a volatile spirit is obtained, while on the contrary by a more vigorous combustion a concentrated acid is obtained.

"A pretty large quantity of phosphoric acid can be obtained also in another way. A large flask or matrass is taken which is left open. A small piece of phosphorus is thrown in. Then the part of the flask with which the phosphorus is in immediate contact is heated with the flame of a candle. It ignites, gives a white vapour which attaches itself to the inner walls of the vessel. Very little phosphorus must be used in this experiment, considering that acid can only be obtained in proportion to the amount of air that the vessel contains. If there is more phosphorus than the air can decompose, it is sublimed without burning.

"We are tempted to believe at first sight that the external air must enter within the flask according as it is absorbed by the vapour of the acid and in this way maintain the combustion of the phosphorus, but it happens otherwise. The vapours that are formed in the flask take the place of the air. They fill the whole of the flask and prevent the access of what is outside the flask."

<sup>&</sup>lt;sup>1</sup> Translated from the original, first published by Meldrum in 1932 (Archeion, 1932, 14, 19-22). As in the previous case, the original is a rough draft; a crude, but punctuated, translation is given.

Thus, on October 20, 1772, Lavoisier, engaged in the preparation of a memoir on phosphoric acid and its salts, studied the combustion of phosphorus in unstoppered vessels and concluded that phosphorus absorbed air in burning and thereby increased in weight. Two forms of acid were produced depending on the conditions, just as occurred with sulphur. Some red phosphorus was produced; and this could be re-converted by suitable means into its usual form. So much for the facts. In his theorizing, Lavoisier supposed that the phosphorus had lost its phlogiston; and that the thick white vapours combined with air to form the acid.

The sealed note, deposited on November 1, 1772, showed what progress Lavoisier had made between these dates. It reads:

"About eight days ago I discovered that sulphur, in burning, far from losing weight, on the contrary gains it; that is to say that from a *livre* of sulphur one can obtain much more than a *livre* of vitriolic acid, making allowance for the humidity of the air; it is the same with phosphorus; this increase of weight arises from a prodigious quantity of air that is fixed during the combustion and combines with the vapours.

"This discovery, which I have established by experiments that I regard as decisive, has led me to think that what is observed in the combustion of sulphur and phosphorus may well take place in the case of all substances that gain in weight by combustion and calcination: and I am persuaded

that the increase in weight of metallic calces is due to the same cause. Experiment has completely confirmed my conjectures: I have carried out the reduction of litharge in closed vessels, with the apparatus of Hales, and I observed that, just as the calx changed into metal, a large quantity of air was liberated and that this air formed a volume a thousand times greater than the quantity of litharge employed. This discovery appearing to me one of the most interesting of those that have been made since the time of Stahl, I felt that I ought to secure my right in it, by depositing this note in the hands of the Secretary of the Academy, to remain sealed until the time when I shall make my experiments known. Paris, November I, 1772. Lavoisier." 1

Thus, Lavoisier claimed to have discovered experimentally that sulphur and phosphorus gained in weight on combustion, apart from the addition of atmospheric moisture, and that this gain was due to their combination with air in the process. He also suspected that the gain in weight of metals on calcination was due to the same cause, namely, combination with air. It is to be noted that he refers to air, not to any particular part of the air or to any particular kind of gas; and that, without examining it, he assumed it was air that was given off by the litharge.

Meldrum suggested that Lavoisier's interest in the combustion of phosphorus was first aroused by the observation of Cigna, reported in the *Observations* for May, 1772 (*Introduction*, II, 84), that phosphorus

<sup>&</sup>lt;sup>1</sup> Œuvres, II, p. 103.

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and sulphur on burning in closed vessels weakened the elasticity of the contained air (*ibid.*, p. 97). Lavoisier was a contributor to this journal: and his experiments on the diamond, carried out with Macquer and Cadet, were the subject of the memoir next but one to Cigna's.

Until 1932 only the last of these notes was available; but, now that all three can be read and studied, it is not difficult any longer to admit what Lavoisier claimed towards the end of his life, namely, that as early as 1772 he had formulated his theory of combustion; for, again quoting Meldrum, to whose interest in Lavoisier the publication of the first and second notes is due: "In support of his claim he quoted the note of the 1st November, 1772. The note bears out his claim. It is a finished composition, thought out, thought through, written with conviction. Facts that are insignificant, each taken by itself, are brought into a relation with one another that proved to have an immense value in chemistry. Lavoisier perceived that absorption of air goes on, and is accompanied by increase in weight, when phosphorus is converted into phosphoric acid, sulphur into sulphuric acid, lead into litharge. Moreover, the reverse change, litharge into lead, is accompanied by production of air. Absorption of air accompanied by increase in weight, production of air accompanied by decrease in weight, on these simple principles he proceeded to establish his doctrine on combustion and, more than that, to establish a new system of chemistry."1

The next document of importance is a memorandum drawn up by Lavoisier for his own private uses. It is written on the opening pages of a laboratory note-book, dated from February 20 to August 28, 1773. The memorandum is plainly dated February 20, 1772. It reads as follows:

"Before commencing the long series of experiments that I intend to make on the elastic fluid that is set free from substances, either by fermentation, or distillation or in every kind of chemical change, and also on the air absorbed in the combustion of a great many substances, I feel impelled to set down here some considerations in writing, in order to outline for myself the course that I ought to take.

"It is certain that there is liberated from substances, under a great many conditions, an elastic fluid; but there are in existence several doctrines as to its nature. Some, like Mr. Hales and his adherents, deemed that this fluid was the air itself, of the atmosphere, combined with substances, either by the action of vegetation and of the animal economy or by artificial means. He thought that the elastic fluid cannot differ from that which we breathe, except in its being more loaded with matters that may be harmful or healthful, according to the kind of substance from which it comes. Some scientific men, coming after Mr. Hales, observed differences so great between the air liberated from substances and that which we breathe, that they

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deemed it to be another substance, to which they have given the name of fixed air.

"A third class of scientific men were of opinion that the elastic matter which escapes from substances was different according to the substances from which it was derived, and their conclusion was that it was no other than an emanation, of the minute and ultimate parts of substances, of which one could distinguish innumerable kinds.

"A fourth class of scientific men1

"However numerous may be the experiments of Messrs. Hales, Black, Magbride, Iacquin, Cranz, Prisley,<sup>8</sup> and de Smeth, in this direction, nevertheless, they come far short of the number necessary for a complete body of doctrine. It is established that fixed air shows properties very different from those of common air. Indeed it kills the animals that breathe it; whilst the other is necessary and essential to their preservation. It combines very readily with all substances; whilst the atmospheric air, under the same conditions, combines with difficulty and perhaps does not combine at all. These differences will be exhibited to their full extent when I shall give the history of all that has been done on the air that is liberated from substances and that combines with them. The importance of the end in view prompted me to undertake all this work, which seemed to me destined to bring about a revolution in physics and in chemistry. I have felt bound to look upon all that has been done before me merely as suggestive: I have proposed

<sup>1</sup> Sic <sup>8</sup> Macbride.

to repeat it all with new safeguards, in order to link our knowledge of the air that goes into combination or that is liberated from substances, with other acquired knowledge, and to form a theory. The results of the other authors whom I have named, considered from this point of view, appeared to me like separate pieces of a great chain; these authors have joined only some links of the chain. But an immense series of experiments remains to be made in order to lead to a continuous whole. An important thing, that most authors have neglected, is to pay attention to the source of the air that is found in a great number of substances. They should have learnt from Mr. Hales that one of the main processes in animal and vegetable economy consists in fixing air, in combining it with water, fire and earth, and in producing all the compound substances that we know. Further they should have seen that the elastic fluid that arises from the union of acids, either with alkalies, or with any other substances, comes originally from the atmosphere; from this they should have been in a position to infer, either that this substance is air itself, combined with a volatile part that emanates from substances, or at least that it is a substance extracted from ordinary air. This way of considering my object made me feel the need of repeating first, and of multiplying, the experiments in which air is absorbed, in order that, knowing the origin of this substance, I could trace what becomes of it in all the various changes.

"The processes by which one can succeed in

fixing air are: vegetation, the respiration of animals, combustion, in some conditions calcination, also some chemical changes. It is by these experiments that I feel bound to begin."

This document, as stated above, though written in a laboratory note-book for 1773, is dated February 20, 1772. Berthelot accepted the latter date: Grimaux, without stating any reason, described it as of February 20, 1773.2 Since it was written in the opening pages of a note-book for 1773, Meldrum suggested that the "1772" was a mistake for "1773," the sort of mistake that is frequently made in the early months of a new year, for example, in dating cheques, and pointed out that Lavoisier elsewhere wrote January 27, 1777, instead of 1778: and he further and more importantly showed that the contents of the memorandum, considered in relation to what we know of Lavoisier's work in 1772 and 1773, show it to be far more closely connected with the experiments he carried out in 1773 than with his researches of 1772. The subject of gases was one that he had not explored when the sealed note of November 1, 1772, was deposited at the Academy; and the memorandum may well be the result of subsequent reading and reflection. Moreover, the experiments that follow in the note-book that opens with the memorandum are all dated 1773 and bear on the contents

<sup>&</sup>lt;sup>1</sup> Berthelot, pp. 46-9: the translation given here is quoted from Meldrum, pp. 8-10, an exact rendering of the original unedited, unrevised memorandum.

<sup>&</sup>lt;sup>2</sup> Grimaux, p. 104.

<sup>8</sup> Berthelot, p. 275.

of the memorandum. We shall therefore take the correct date of this document to be February 20, 1773. But, in a last word, we may quote Meldrum:

"This Memorandum conveys a great impression of Lavoisier's strength and ardour of mind and of his confidence in himself. It reveals a project, that he had formed of doing work that would bring about a revolution in physics and chemistry: he aimed at nothing less. It is known that Lavoisier, in course of time, did bring about a revolution in science and did establish a new system of knowledge. It appears that he had dreamt of doing all this alone. But it is known, also, that men, who adhered to the old ways of thought and opposed his efforts to replace them by new ways, nevertheless were of assistance to him in his labours. The Memorandum has a note of exaltation and even of inspiration. Nothing like it can be found in Hales, Black, Cavendish, Priestley, Scheele. Lavoisier applied his imagination to what he knew of past and present and he foresaw a revolution in science. The Memorandum cannot have been drawn up impromptu. Lavoisier showed in it perception of the issues involved in the subject of gases and could specify the different schools of thought. He remarked that the knowledge of gases was fragmentary, that chemists had studied the gas that is absorbed and the gas that is liberated in chemical change separately from one another: he saw that all this could be improved on and be coordinated with other knowledge so as to produce an immense effect on science. But the subject of the

gases had been neglected in France: Lavoisier hardly mentions it in his early memoirs: he had no experience of it. Hence it must needs be assumed that he spent time in reading and in reflecting on the subject before he wrote the Memorandum. Moreover, the Memorandum conveys the suggestion that Lavoisier would begin his experiments forthwith. . . . Lavoisier showed immense energy in attending to his official duties and his scientific interests. With him action followed hard upon thought. Let us suppose that his resolution, expressed in the Memorandum, to engage in work that would lead to a revolution in physics and chemistry and establish a new system of knowledge, was formed in February, 1772. Then a year of inactivity appears that cannot be accounted for: it is incompatible with Lavoisier's temperament that, having read up a subject and formed a scheme for work upon it, he should do nothing for a year. It is true that the year of inactivity was interrupted by experiments upon combustion that he made in September and October during about seven weeks. I cannot believe that Lavoisier, in February, 1772, framed a project of work that was to lead to a revolution in science, did nothing till September, worked on the subject for seven weeks and did nothing further till February, 1773."1

The project that Lavoisier had thus resolved upon on February 20, 1773, led him to carry out extensive

experiments and to prepare an account of them for publication; and, as is usual with almost all his work, this publication, which appeared in January 1774, as the Opuscules Physiques et Chymiques, was arranged in two parts, the first historical and the second experimental. Lavoisier's orderly mind invariably saw the present growing out of the past and he constantly sought to trace the connection between the two. In the Introduction to the historical part he wrote: "A great number of foreign Philosophers and Chemists are at this time employed in researches concerning the fixation of Air in bodies, and the elastic vapours which are separated in the combination as well as in the decomposition and resolution of their principles. Various Memoirs, Theses and Dissertations have appeared on this subject in England, Germany, and Holland. The French Chemists alone seem not to take any part in these important inquiries; and while the discoveries of other nations increase every year, our modern publications, the most complete, in many respects, of any that have been written in Chemistry, are almost totally silent upon this subject. These considerations induced me to think it necessary to give the public a short account of every thing, which has hitherto been done, relative to the combination of Air with bodies, and to give an accurate description of the discoveries which have been made in this subject. This I propose to do in the first part of this Treatise. . . . In the second part, an account will be given of my own experiments."1

<sup>1</sup> Opuscules, Paris, 1774, pp. 1-2: English trans. by T. Henry, Essays

In the historical part, Lavoisier referred to the work of van Helmont, Boyle, Hales, Black, Cavendish, Priestley and others. The contents of this part show that he had read widely in the subject and had made himself familiar with the works of all the more notable, and some of the minor, contributors to it. But it is the experimental part that interests us here and enables us to trace the development of Lavoisier's attack on the problems of combustion, calcination and the fixation of air in bodies. The first three chapters described experiments following those already carried out by Black and others on the action of acids on the mild alkalis and included gravimetric estimations of the amounts of "elastic fluid," as Lavoisier called it, that were contained in chalk and soda. In these experiments Lavoisier gives the reader the impression of having satisfied himself by numerous tests that Black's theory was correct: the details of the work are not of importance here, but the obvious reasoned adoption of Black's views is significant. In Chapter IV, Lavoisier described experiments in which he dissolved mercury and iron separately in nitric acid and, taking two samples of each solution, added chalk to one and slaked lime to the other, in the amounts calculated to be equivalent to the acid content. The chalk gave heavier precipitates than the lime did; and the addition of chalk produced an effervescence in each case, whereas the addition of the slaked lime did not. Hence, concluded Lavoisier, the

Physical and Chemical, London, 1776, pp. 1-2. All quotations have been taken from the English translation and checked against the French version. Henry's work is a faithful rendering of the original.

increase of weight in the precipitated as compared with the original metals appeared to be due to their union with "elastic fluid." Moreover, it was known that, if a metal dissolved in acid was precipitated, not by an earth, but by another metal, the dissolved metal was thrown down from solution in its metallic state, not as a calx, and without any increase in its weight. Here, Lavoisier saw support for his conclusions; for this result appeared to him to be due to the absence of any substance from which the metal might attract "elastic fluid." But it is clear that in these last experiments Lavoisier supposed that the "elastic fluid" he was referring to was the same in every case as that which he had been studying in chalk and soda, namely, Black's "fixed air," the substance now known as carbon dioxide. For instance, he wrote: "Experiments, sufficiently numerous, lead me to believe, that the elastic fluid, the same whose existence I have endeavoured to prove in calcareous earth and alkalis, is capable of uniting, by precipitation, with most metallic substances; that it is, in a great measure, this principle which forms the augmentation of weight in metallic calces, which deprives them of their brilliancy, which reduces them to the form of a calx, &c."1

Referring to the experiments on the precipitates of mercury and iron, Lavoisier then explained at the outset of Chapter V that, while they "may not have completely proved the possibility of the union of elastic fluid with metallic substances, they at least afforded such indications of it, as were sufficient

<sup>1</sup> Opuscules, p. 247 ; Essays, p. 289.

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to engage me to pursue the subject with particular attention. I began, from this time, to suspect, that the air of the atmosphere, or an elastic fluid contained in the air, was capable, in a great many circumstances, of being fixed and combined with metals; that to the addition of this substance, the phenomena of calcination were owing, as likewise the augmentation in weight of the metals converted into calces." These views appeared all the more probable for three reasons, which Lavoisier set out as follows: "1st. The calcination of metals cannot take place, in vessels closely stopped and exhausted of air. 2dly. It is proportionably the more readily performed, as a greater extent of surface in the metal is exposed to the air. 3dly. It is a fact known to all the metallurgists, and observed by all those who have been conversant in the operations of assaying metals, that in every reduction, there is an effervescence at the moment the metallic substance passes from the state of a calx to that of a metal; now, an effervescence is commonly no more than a separation of elastic fluid; the calx therefore contains an elastic fluid, under a fixed form, which recovers its elasticity, at the instance of reduction."2

Proceeding to test these conclusions by experiment, Lavoisier used the apparatus shown in Fig. 2, consisting of a glass receiver FGH containing air and inverted over water in the trough BCDE, the receiver containing a glass support, IK, carrying a porcelain crucible A. The height of the water in

<sup>&</sup>lt;sup>1</sup> Opuscules, p. 254; Essays, pp. 297-8.
<sup>2</sup> Opuscules, p. 255; Essays, pp. 298-9.

the receiver was suitably adjusted by means of a siphon; and a quantity of oil, to cover the surface

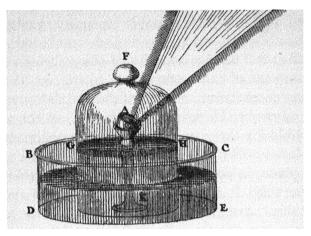


Fig. 2.

of the water in FGH and intended to prevent any of the "elastic fluid" that might be produced in the experiment from coming into contact with the water and being absorbed by it, was introduced by means of a funnel. A mixture of 2 gros of minium and 12 grains of powdered charcoal was then placed in A, the height of the water was marked with a strip of paper, and the apparatus was moved into the focus of one of Tschirnhausen's large burning lenses. When the focus of the lens fell on A, the minium was almost immediately reduced to metallic lead; and, when the apparatus was cool, the level of the water had sunk by an amount corresponding to the separation of about 14 cubic pouces¹ of "elastic fluid." But the experiment was difficult to carry

<sup>1</sup> I Paris pouce=1.066 English inch. 1 cubic pouce=1.21 cubic inch.

out: the great heat broke the receivers in some cases and might have caused the production of some "elastic fluid" from the oil. Lavoisier therefore employed another apparatus, in which a mixture of 6 onces of minium and 6 gros of powdered charcoal was heated in an iron retort in a furnace and the "elastic fluid" produced was collected over water covered by a layer of oil as before, the fluid being led from a retort along a pipe projecting above the upper surface of the water in the receiver. The volume of "elastic fluid" produced was 560 cubic pouces, or 747 times the bulk of the residue of 5 onces 7 gros 66 grains of lead from the minium. Repetitions of the experiment confirmed the result. Lavoisier realised that the "elastic fluid" produced here was identical with that obtained by the action of acids on mild alkalis, but some calculations that he made from his numerical data, from the loss in weight of the charcoal and of the minium on reduction to lead, gave him a value for the density of the " elastic fluid " that he regarded as much too high. Accordingly, he made some tests of the amounts of water produced by heating a mixture of minium and charcoal and by heating charcoal alone: but he satisfied himself that the minium was the source of the greater part of the "elastic fluid" produced. In a further experiment, in which a mixture of minium and powdered charcoal was heated in a gun-barrel, the result of the experiment was again confirmed.

"It appears to be proved from these experiments," added Lavoisier, "that it is by no means

the charcoal alone that produces the discharge of elastic fluid . . . neither is it the minium alone, since after Dr. Hales's experiments . . . it affords but a very small portion of air; the greater part of elastic fluid which is detached, arises from the union of the powdered charcoal with the minium. This last observation leads us insensibly to very important observations on the use of charcoal, and such kind of substances in general, in the reduction of metals. Do they serve, as the disciples of M. Stahl think, to restore to the metal the phlogiston which it has lost? Or rather do these substances enter into the composition of the elastic fluid? This is a point which, in my opinion, the present state of our knowledge does not permit us to decide."

The next series of experiments, described in Chapter VI, were far from satisfactory. They were intended to demonstrate that, just as there was a discharge of elastic fluid when a metallic calx was converted to metal as in the preceding experiments, so also there was an absorption of the same fluid when the metal was converted into a calx and that the calcination was nearly proportional to this absorption.<sup>2</sup> Lead was the metal chiefly used, and weighed amounts of it were calcined, by means of Tschirnhausen's great lens, in air in a receiver inverted over water, protected by a layer of oil as before, or over mercury. The lead lost in weight, but it was obvious from the deposits on the walls of the receiver that the great heat had volatilized some of

<sup>&</sup>lt;sup>1</sup> Opuscules, p. 279; Essays, p. 324. <sup>2</sup> Opuscules, p. 282; Essays, p. 327.

133 the calx. An experiment with tin was not very successful: there was a very small decrease in the air and a slight increase of weight in the tin. The absorption of the air amounted, even in the best case, only to 1/20 when excess of lead was present.1 Lavoisier further observed here that the residual air supported the combustion of a candle for about a minute and did not give a precipitate with limewater, so that it was evident "that air in which metals have been calcined, is not, by any means, in the same state as that separated by effervescence, and by metallic reductions."2 And, in another experiment, he found that moist iron filings reduced by ½ the volume of the air in which they were

Arguing from these experiments, Lavoisier concluded:

enclosed for two months.

- 1. Metals are calcined more easily in the open air than in a limited amount of air enclosed in a receiver.
- 2. There is a limit to the amount of calcination producible in a metal in a limited amount of air,

<sup>1</sup> Priestley had already obtained a much better result than this, namely an absorption of 1 to 1 (Phil. Trans., 1772, 62, 228). Lavoisier stated that he was unaware of Priestley's experiments at this time and tried to account for the difference between Priestley's result and his own by suggesting that the amount of fixable elastic fluid in the air varied with time and

place (Opuscules, p. 295, footnote; Essays, p. 341, footnote).

2 Opuscules, p. 291; Essays, p. 337. These two "airs" had already been differentiated by Daniel Rutherford in his De Aere Fixo Dicto, autherfordisco (Edin., 1772). Lavoisier mentioned Rutherford's "very well written thesis," but at this time he clearly regarded it as a summary of the work of Black and others, and Rutherford's recognition of "noxious air" (nitrogen) escaped his notice (Opuscules, p. 183, footnote; Essays, p. 217, footnote). Crum Brown's translation of Rutherford's De Aere Fixo, etc. was recently published by Dobbin (3. Chem. Education, 1935, 12, 370). The author has discussed Rutherford's work in Science Progress, 1935, 29, 650.

- 3. The volume of air decreases as calcination proceeds, the diminution corresponding nearly to the gain in weight of the metal.
- 4. These experiments together with the experiments on the reduction of minium with charcoal seem to prove that an elastic fluid combines with metals in calcination and thereby causes the well-established increase of weight observed in metals in this change, and
- 5. "Several circumstances would seem to lead to a belief, that the whole of the air which we breathe is not adapted to be fixed, and enter into combination with metallic calces; but that there exists in the atmosphere, an elastic fluid of a particular kind which is mixed with the air, and that it is at the instant when the quantity of this fluid contained under the receiver is consumed, that the calcination can no longer take place. The experiments which I shall relate in Chapter IX will give, at least, some degree of probability to this opinion."

Of these conclusions, as Meldrum<sup>2</sup> pointed out, the first is unexceptionable, while for the second and fifth Lavoisier had no satisfactory experimental evidence, but only a single experiment in which the air was diminished by  $\frac{1}{20}$  (instead of  $\frac{1}{6}$ ), and for the third he had no evidence at all; and the fourth indicates that he believed that the gas combined with lead when calcined was the same as that evolved when minium was heated with charcoal.

<sup>1</sup> Opuscules, pp. 293-4; Essays, pp. 339-40.

<sup>&</sup>lt;sup>2</sup> Meldrum, pp. 24-5.

Despite these defective conclusions, however, Lavoisier had clearly grasped the idea that calcination involved a combination of the metal with air. And he went on to say: "The experiments of which I have given an account, would appear also to lead to the two following consequences: 1st, That the calcination of metals cannot take place in vessels closely stopped, or, at least, that it can only be in proportion to the quantity of fixable air which is confined in them: 2dly, That in case the calcination could proceed in vessels closely stopped and exhausted of air, it should then be without increase of weight, and consequently with circumstances very different from those observed in calcination performed in air."1 The experiments suggested in this place were carried out later and we shall refer to them in another chapter. Meanwhile, the further experiments described in the Opuscules demand our attention.

In the next series of experiments, described in Chapter VII, Lavoisier showed that the "elastic fluid" obtained by the reduction of minium with charcoal and that obtained by the action of acid on chalk had identical properties. Sparrows, mice, and rats plunged into either perished immediately, and lighted candles and red-hot charcoal were extinguished instantly: and both fluids precipitated lime-water. Lavoisier had thus proved, for the first time in the history of chemistry, that the "elastic fluid" or gas produced in the reduction of calces with charcoal was identical with Black's "fixed air"

<sup>1</sup> Opuscules, p. 294; Essays, p. 340.

—a very important advance in chemical knowledge, although its explanation was still some years distant.

Lavoisier had however involved himself in a difficulty. In determining the quantities of the two "elastic fluids" mentioned above that were absorbed by lime-water, he passed the two gases separately through an apparatus which contained lime-water, but from which the air had not been removed: he thought that the removal of the air would have been preferable, but the apparatus was not of a form that permitted this. However, he tested the residual gas and found that it supported the respiration of animals. Accordingly, he concluded "that both these fluids are composed, 1st, of a fixable part capable of being combined with water, &c. 2dly. Of another part, much more difficult to fix; capable of supporting, in a certain degree, the lives of animals, and in its nature much resembling the air of the atmosphere; . . . That this portion of common air is rather more considerable in the elastic fluid disengaged in metallic reductions, than in that detached from chalk. . . . That it seems certain that the noxious property of this fluid resides in its fixable part, because it is less fatal to animals in proportion as it is farther deprived of this part. . . . That nothing as yet enables us to decide whether the fixable part of elastic fluid from effervescing mixtures and reductions, be a substance essentially different from air, or whether it be air itself to which something has been added, or from which something has been subtracted, and that prudence demands us to suspend our judgment,

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at present, on this subject." Thus, while he recognized the identity of the "elastic fluids" from the action of acids on chalk and from the reduction of minium with charcoal, Lavoisier could not arrive at any satisfactory explanation of the relation of this fluid to ordinary air.

Lavoisier then reported in Chapter VIII various observations on the chemical properties of the aqueous solutions of the "elastic fluid" obtained either by the action of acid on chalk or from the reduction of minium with charcoal. It was clear that the solutions had the same properties, "whether we employ elastic fluid separated from effervescing mixtures, or that from metallic reductions."

In Chapter IX we come to the important experiments on the combustion of phosphorus in a receiver inverted over water. We have already quoted the three notes of September 10, October 20, and November 1, 1772, all of which referred to the combustion of phosphorus; but we are justified in assuming that the account given in the Opuscules includes both improved and additional experiments. Here, indeed, Lavoisier showed that phosphorus, ignited with a burning lens in air confined in a receiver over water that was protected by a layer of oil, reduced the volume of the air by an amount between 1 and 1 and gave copious white vapours, which condensed on the sides of the receiver and

<sup>1</sup> Opuscules, pp. 319-20; Essays, pp. 368-9.
2 Opuscules, p. 326; Essays, p. 376. In the Opuscules, dissolutions has been mis-printed for réductions, but this is corrected in the Essays quoted above and in Œuvres, I, p. 639.

then formed drops of a clear limpid liquid; that a similar result was obtained when the experiment was carried out over mercury; that in repeated experiments the diminution of the air was proportional to the amount of phosphorus burnt; and that the greatest diminution of the air producible by the combustion of phosphorus in such experiments approached  $\frac{1}{2}$  of the whole volume of air.

"These experiments," concluded Lavoisier, "seem already to lead to a suspicion that atmospheric air, or some other elastic fluid contained in the air, is combined, during the combustion, with the vapours of the phosphorus. But there is a great difference between conjecture and proof, and it was essentially requisite that it should first be well established that a combination of any kind of substance was formed with phosphorus during its combustion. The following experiments appeared to me to be proper for furnishing that proof."

But the new experiments required another kind of apparatus of the form shown in Fig. 3 opposite. Eight grains of phosphorus were placed in the small glass cup, B, resting on the base of the wide-mouthed bottle, P, which was then tightly closed with a cork and weighed. The bottle was then uncorked and placed under the glass receiver A over mercury in a dish W, the height of the mercury being suitably adjusted to the level CG. The phosphorus was then kindled by a burning lens. White flocculi of phosphoric acid were sublimed and deposited on B and the interior of P; but at least a quarter escaped

<sup>1</sup> Opuscules, p. 332; Essays, p. 383.

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and settled on the walls of the receiver, the outside of P and the surface of the mercury. When the apparatus had cooled, Lavoisier noted that the absorption was of the order of 16 to 17 cubic pouces and that a small amount of the phosphorus was left unburnt. He lifted A up and in four seconds recorked P, being thus satisfied that no sensible replacement of the air in P could have occurred.

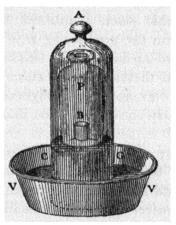


Fig. 3.

The bottle P was then cleaned on the outside and re-weighed. Its weight had increased by 6 grains, so that "instead of 8 grains of phosphorus, which I had put into the bottle, there were now 14 grains, either of concrete phosphoric acid or of phosphorus half decomposed: but it must be remembered that at least a fourth, that is to say, 3 to 4 grains, was separated during the burning, on the outside of the bottle; and consequently that 6 to 7 grains of phosphorus yield 17 to 18 grains of concrete

phosphoric acid, or in other words, that 6 to 7 grains of phosporus absorbed 10 to 12 grains of some substance contained in the air which is confined under the receiver. This experiment leaves scope enough to preclude any reasonable doubt of the result, and all the arguments which could be adduced would, at most, tend only to reduce the increase of weight to 8 to 10 grains instead of 10 to 12. The quantity of air absorbed, was, at most, seventeen cubic pouces combined with the phosphorus to form the phosphoric acid; this quantity communicated an increase in weight of from 10 to 12 grains, and therefore every cubic pouce of elastic fluid which was absorbed weighed about 3 of a grain, i.e. nearly one-fourth more than the air which we breathe."1

It therefore seemed that the part of the air absorbed by the phosphorus was the heavier part of the air; and Lavoisier now began to suspect that it might be water. To illustrate his difficulties at this period and to show how laboriously these now apparently simple problems approached their solution, we shall again quote Lavoisier's own words here. "But," he wrote, "if the matter attracted by the phosphorus, during its combustion, be the heavier part of the air, why may it not be water itself which that fluid holds dissolved, and is diffused so abundantly in the atmosphere in a kind of state of expansion? Without doubt, I reasoned with myself, water is necessary to the aliment of flame; in proportion as air contains it, it is proper to sup-

<sup>&</sup>lt;sup>1</sup> Opuscules, pp. 334-5; Essays, pp. 385-6.

port combustion; but when deprived of it, combustion can no longer take place." If this supposition was true, it should follow, added Lavoisier: "1st. That by restoring to the air confined under the receiver in which the phosphorus has been burnt, a quantity of water reduced to vapours proportionable to that which has been absorbed, the combustion, instead of ceasing, should be prolonged much farther. 2dly. That in this case there should be no farther diminution in the bulk of air in proportion as the phosphorus burns. 3dly. That by restoring to a quantity of air, in which phosphorus has been burnt, and consequently deprived of its water, and diminished about a fifth, a proportion of water reduced to vapours, an increase should be produced in its bulk equal to the diminution which it had suffered during the combustion."2

Lavoisier therefore proceeded to test this hypothesis experimentally in an apparatus consisting of two small agate cups, placed in an earthen dish floating on mercury under a glass receiver. One cup contained a known weight of phosphorus: the other contained water. In the first test, Lavoisier boiled the water with a burning lens until the steam was condensing in drops on the sides of the receiver and then ignited the phosphorus with the lens; but the fractional diminution of the air was the same as before and the only difference was that the phosphoric acid was produced in a liquid instead of a solid form. In the second test, the phosphorus

<sup>&</sup>lt;sup>1</sup> Opuscules, pp. 335-6; Essays, p. 386. <sup>2</sup> Opuscules, p. 336; Essays, p. 387.

was ignited first and then the water was boiled; the same absorption of air occurred, the only difference being that the phosphoric acid was first produced as a white deposit on the walls of the vessel. which then deliquesced, as Lavoisier correctly saw, through the moisture that was supplied by the water even in the cold; but the subsequent boiling of the water in the cup produced no change in the volume of the air. In the third test, a much larger amount of phosphorus was taken, while the other cup contained water. The water was boiled first and then the phosphorus was kindled. After combustion had ceased, the absorption was found to be very nearly the same as before, no greater amount of phosphorus had disappeared and it was not possible to re-kindle the phosphorus with the burning lens. Not one of the conclusions, drawn from the hypothesis that it was water from the air that had been absorbed and had led to the diminution of the air, was supported by experiments: and hence Lavoisier concluded: "1st. That the greater quantity of the substance absorbed by phosphorus during its burning, is something else than water. 2dly. That it is to the addition of this substance that the phosphoric acid owes its increase in weight. 3dly. That to the subtraction of it, the diminution in the bulk of air in which phosphorus has burned is to be attributed."1

As a further confirmation of this Lavoisier then converted 2 gros 10 grains of phosphorus into phosphoric acid and dissolved the acid in distilled water. The solution was weighed and found to be 3 gros

<sup>1</sup> Opuscules, p. 341; Essays, p. 392.

27½ grains heavier than an equal volume of distilled water; "from whence," wrote Lavoisier, "it evidently follows that the phosphorus had attracted, during its combustion, at least 1 gros 17 grains of some kind of substance. This substance could not be water, because water could not have augmented the specific gravity of water; it was therefore either air itself, or some other elastic fluid contained, in a certain proportion in the air which we respire. This last experiment appears so demonstrative to me, that I do not foresee any objection that can be advanced against it."1

The only further experiments recorded in the Opuscules, in the two final short Chapters X and XI, are proofs of the already established observations that in a vacuum neither phosphorus nor sulphur will burn, and neither gunpowder nor a mixture of sulphur and nitre will explode, and some tests made on the air in which phosphorus had been burnt, whereby Lavoisier showed that a lighted candle plunged into this air was instantly extinguished, although a bird put into the same air for half a minute suffered no difficulty in respiration.2 He noted that, if placed in fixed air, the bird would have perished instantly. In a final experiment, Lavoisier mixed one part of the "elastic fluid" ("fixed air" from an effervescing mixture) with

<sup>1</sup> Opuscules, p. 346; Essays, p. 398.
2 The air had been diminished by the burning of phosphorus only to the extent of 1/2 th and was therefore sufficiently respirable for this test. Long before this Boyle had shown in numerous experiments that animals placed in closed vessels of air together with inflamed substances easily survived the extinction of the flames, even when the flames were put out by gradual evacuation of the air (New experiments, touching the Relation betwixt Flame and Air London, 1572, pp. 100-111. and Air, London, 1672, pp. 109-118).

two parts of the air in which phosphorus had been burnt to see if the latter would thereby have restored to it its property of supporting flame: the mixture immediately extinguished a lighted candle. But the experiment emphasizes for us the fact that the "elastic fluid," the absorption of which Lavoisier argued to be demonstrated in combustion and calcination, was further supposed by him to be that obtainable by the action of acids on chalk, namely, "fixed air."

The Opuscules was submitted to the Academy, reported on by de Trudaine, Macquer, Le Roy and Cadet on December 7, 1773, and published with the approbation of the Academy in January, 1774. The whole production had taken less than a year to complete. The book certainly gives the reader the impression of much unsatisfactory experiment; but, if allowance be made for the fact that Lavoisier was here engaged in pioneer labours among masses of bewildering data, the completion and publication of the book within a year, a year of a life that was already amply filled with other exacting activities and responsibilities, was a creditable and, indeed, a great performance; and, with all the hesitancy that the modern reader can detect in it here and there, it continually and soundly insists that the cause of the gain in weight of metals on calcination is their combination with air or an elastic fluid contained in the air. That Lavoisier supposed this fluid to be identical with Black's "fixed air" was an error in detail rather than in principle: but it was an error that led him to vacillate in his explanations

145 from time to time. For instance, in one place he argues that it is the "elastic fluid" ("fixed air") that combines with metals to form calces and increases their weight1; in another, that this is due to the air of the atmosphere or an elastic fluid contained in it and capable of fixation and combination with metals2; in another, that it is not air, but an elastic fluid of a particular kind contained in it3; and in others, that the similar gain in weight of phosphorus on combustion is due to some substance contained in the air or to the heavier part of the air,4 or, either air or some other elastic fluid contained in it.5 Thus he could not decide whether air was a mixture or not: and we see him hesitating between the two possibilities, but holding fast in all places to the sound principle that gain in weight arises from combination with the air or with a part of it. What the Opuscules reveals is not so much Lavoisier's personal difficulties, though he had these in plenty as a beginner in pneumatic chemistry, but, as Meldrum pointed out, the slow progress of knowledge: and in 1773, with oxygen still unrecognised, that progress was bound to be slow. In the next Chapter, therefore, we shall need to consider the isolation and recognition of oxygen by

Before proceeding to this, however, it is interesting to take some notice of the entries made by Lavoisier

Joseph Priestley in 1774-5.

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<sup>1</sup> Opuscules, p. 247; Essays, p. 289.
2 Opuscules, p. 254; Essays, pp. 297-8.
3 Opuscules, pp. 293-4; Essays, p. 340.
4 Opuscules, pp. 334-5; Essays, pp. 385-6.
5 Opuscules, p. 346; Essays, p. 398. 6 Meldrum, p. 33.

in his laboratory note-books during the time that he was engaged on the experiments described in the Opuscules. Unfortunately, there is only one note-book available. This covers the period from February 20, to August 28, 1773. This is the one we have referred to previously: in its first pages Lavoisier wrote down the memorandum quoted on pp. 120-3 above. Turning to the note-book1 therefore, we find that Lavoisier was studying the calcination of lead and tin in retorts on February 22 and 23. Some days later he noted that minium acted on sal ammoniac in the same way as lime acted, namely, to give volatile alkali, that is, an alkali deprived of air. This puzzled him greatly. He noted that, according to his theory, minium was a compound of air and lead—une combinaison d'air et de plomb. Therefore, this air ought to pass unto the volatile alkali, with which it had a great affinity. But it was dissipated and escaped in the operation. "This difficulty," wrote Lavoisier, "is perplexing and I think it must lead us to conclude that the air combined with lead in minium is not the fixed air that combines so very readily with the alkalis. It is without doubt atmospheric air itself. Perhaps also the air evolved from the lead is not sufficiently charged with phlogiston to combine with the alkalis; for, according to some, fixed air is an air combined with phlogiston; but I confess that all this is very uncertain."2 Thus, late in February, 1773, Lavoisier had already suspected that the air

Berthelot, pp. 225-49.
 Berthelot, pp. 233-4. The originals are rough notes: crude translations are therefore given here for all such cases.

combined with lead in minium was not fixed air but atmospheric air; and at the same time he had not rejected phlogistic notions, but he had gone so far as to find them unsatisfactory.

On March 1 Lavoisier considered apparatus for combining fixed air with various liquids; and also during this month he noted that he must determine the weight, i.e., the density, of fixed air, whether its volume altered with heat and cold like the air of the atmosphere, whether it was elastic and compressible, and what effect the washing of it with various substances would have on its action on animals. On March 29 he carried out the experiment on the calcination of lead that we have already described above (p. 132). And he wrote in his note-book: "I noted with surprise that the lead was not calcined further. From that time I began to suspect that contact with circulating air is necessary for the formation of the metallic calx; that perhaps also the whole of the air that we breathe did not enter into the metals that are calcined, but only a part of it that is not very large in a given mass of air; perhaps also the layer of calx that covered the surface of the metal prevented the direct contact of the air and arrested the progress of the calcination—the temperature of the surrounding air not being taken, this is an experiment to repeat. The lead, being taken out, had not gained in weight: it had on the contrary lost about half a grain, which was doubtless due to the vapours that had been emitted from it."1

<sup>&</sup>lt;sup>1</sup> Berthelot, p. 236. Cf. Opuscules, pp. 284-5; Essays, p. 329.

Lavoisier had thus almost tripped over a discovery that was still to cause him much difficulty and labour; for here, on March 29, 1773, he had suspected that it was only a part, and that not the larger part, of the air that combined with metals in calcination. His published expression of this suspicion we have already quoted above (p. 134): it is the substance of the fifth conclusion derived from the experiments described in Chapter VI of the *Opuscules* on the calcination of lead in air confined over water or mercury.

On the same date apparently Lavoisier obtained a partial reduction of minium with charcoal. Some air was evolved, but he thought that the charcoal might have contributed to it. On March 30 he studied the calcination of tin, and on March 31 the reduction of lead in the burning lens, noting that the volatilized calx had probably absorbed some air.

On April 6 Lavoisier studied "alkali combined with metallic calces." He wrote: "I have many times made an objection against my system of the reduction of metals and it consists in this: lime in my view is a calcareous earth deprived of air; the metallic calces on the contrary are metals saturated with air. Yet both produce a like effect on the alkalis, they make them caustic." Here again we see Lavoisier confusing fixed air with that part of the atmospheric air that on March 29 he had suspected of being involved in the calcination of metals. He went on here to record an experiment

in which crystals of soda were boiled with minium, but did not become caustic. This contradicted the statement he had just written down, but Lavoisier made no comment, proceeding to carry out an experiment in which mercury was precipitated by a solution of soda from its solution in nitric acid. The precipitate was greenish grey in colour; and it was much darker when the alkali from the preceding experiment with minium was used instead of soda. "All this," wrote Lavoisier, "would lead me to think that the alkali that passed over the minium contains more air than the other, very far from being deprived of it and from being caustic. The objection would seem to me to be thoroughly refuted as far as fixed alkali is concerned; but it stands completely for volatile alkali-indeed, the volatile alkali that is obtained from the combination of sal ammoniac and minium is in a state of causticity. How explain this phenomenon? I confess that I still know nothing about it." Lavoisier was, however, merely assuming that air had passed from the minium to the fixed alkali; but the reaction between minium and salammoniac lent itself to no such assumption, since caustic volatile alkali was produced.

On April 21 Lavoisier noted that fulminating gold exploded noiselessly in a vacuum under the action of a burning lens<sup>2</sup>; and that in a vacuum

<sup>1</sup> Ibid., pp. 238-9.

<sup>&</sup>lt;sup>2</sup> This experiment on aurum fulminans, following on the difficulties cited by Lavoisier, recalls Black's comment on this substance, quoted on p. 74 above. Lavoisier seems to have been strongly influenced by Black in the way we have suggested in the footnote on p. 75 above: and that he should turn to experiment with this substance at this time tends to confirm that suggestion. Unfortunately Berthelot gives no details of Lavoisier's comments on the experiment.

phosphorus sublimed without igniting or turning water acid, sulphur sublimed and gunpowder did not explode.1 On May 4 he heated vitriolic acid with charcoal in the burning lens and decided to repeat the experiment on another occasion; and he noted that phosphorus under spirits of wine could not be ignited by the burning glass "for lack of contact with the air." During the next few days Lavoisier carried out experiments on the absorption of fixed air in water, on cooling it with a freezing mixture, on passing it into lime-water, etc.; and noted that fixed air that had stood over water should be tested to find out whether it became ordinary air.2

On May 10 Lavoisier experimented on the boiling of volatile alkali under the air pump. He wrote: "The volatile alkali obtained from sal ammoniac by minium is certainly a caustic alkali; I tested it with lime-water and it did not precipitate it in the least. This fact is extremely peculiar and seems to me wholly inexplicable on the principle of the fixation of air in the metallic calces." Again, he was confused through supposing that fixed air was the air that combined with metals in calcination. During the next few weeks, he carried out various experiments that were suggested by Black's work and that were subsequently included in the early chapters of the Opuscules; and on May 20, on

<sup>&</sup>lt;sup>1</sup> Reported in Chapter X of the Opuscules; see p. 143 above.

<sup>2</sup> This apparent change had been noted by several experimenters at this time: it was due to exchange of fixed air and the ordinary air already dissolved in the water.

<sup>3</sup> Berthelot, p. 240.

slaking lime in a vacuum, he wrote: "I did not observe that there was any production or absorption of air in this operation; but as the receiver luted to the air pump allowed some air to enter, this experiment is to be repeated." Towards the end of the month, he reverted to experiments with lead: and on May 23 he heated minium and charcoal in an iron retort, concluding that: "Lead on conversion to minium takes up nearly 388 times its volume of air."2 Next day, he pumped the air obtained in the previous day's experiment through bottles containing lime-water and found—he does not say how-that "this air did not completely contain fixed or combinable air—supposing fixed air of the same weight as atmospheric air"; and on May 27 he experimented again with caustic volatile alkali prepared with lime.

During June, 1773, Lavoisier carried out further experiments suggested by Black's work on the alkalis; and, from determinations of the specific gravity of lime-water before and after the action of fixed air and of the decrease in the specific gravity of fixed alkali by the action of lime, concluded that fixed air had almost the same density as atmospheric air. He then reverted to experiments on the combustion of phosphorus and the reduction of minium with charcoal.

On July 1 Lavoisier studied the air in which

<sup>&</sup>lt;sup>1</sup> *Ibid.*, p. 242.

<sup>&</sup>lt;sup>2</sup> Ibid., p. 243. Another experiment was reported in the Opuscules in which the figure was much higher, namely, 747 (Opuscules, p. 268; Essays, p. 312). Cf. p. 131 above.

<sup>8</sup> Ibid., pp. 243-4.

phosphorus had been burnt. It extinguished a candle; but it neither precipitated lime-water nor asphyxiated a bird. These experiments were described in the closing chapters of the Opuscules and have already been referred to on p. 143 above. Further, he wrote in his note-book: "Persuaded that the combustion of phosphorus absorbs the fixed air contained in the air, or rather suspecting it, I thought that by restoring fixed air to that air it could perhaps be made common air again. I made the mixture, but I introduced too much fixed air. etc. The mixture extinguished a small candle, etc."1 Here again is Lavoisier's confusing and persistent assumption that the part of the air that is active in combustion is fixed air, an assumption that may possibly have arisen through Black's description of fixed air as "dispersed thro' the atmosphere, either in the shape of an exceedingly subtile powder, or more probably in that of an elastic fluid."2

Further experiments suggested by Black's researches on the alkalis were carried out in July and August, 1773; and some of them were later on described in Chapter IV of the *Opuscules* and have been already referred to. On July 3 Lavoisier put a rat into the air obtained by the reduction of minium with charcoal, an experiment reported in Chapter VII of the *Opuscules*. On July 20 and 22 he burnt phosphorus again and determined the "increase of weight of the acid of phosphorus"; and he evidently made on July 22 the experiments to test the

<sup>&</sup>lt;sup>1</sup> Berthelot, p. 246. <sup>2</sup> Cf. p. 77 above. <sup>8</sup> Cf. p. 135 above.

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effect of water vapour on this reaction.¹ On July 22 there is an entry without details referring to the reduction of mercury precipitated by chalk; and another entry about the same time refers to a reduction of calx of lead with charcoal, and on July 31 a further reduction of "minium or calx of lead," the air produced being passed through lime-water. On August 7, Lavoisier burnt phosphorus over mercury: and at this time he also studied the proportion between the weights of lead and the weight of minium.

Thus Lavoisier's laboratory note-books for this period provide further and ample evidence of the confusion and hesitation in which he was involved through being unable to differentiate between "fixed air" and the part of the air that was active in combustion and calcination. The clue to these difficulties was provided late in 1774 when Priestley informed him, as described in the next chapter, of his isolation of the gas now known as oxygen.

<sup>&</sup>lt;sup>1</sup> Cf. pp. 140-2 above.

## CHAPTER V

## PRIESTLEY'S ISOLATION AND RECOGNITION OF OXYGEN

THE RESEARCHES that led Joseph Priestley to the isolation and recognition of oxygen or, as he called it, "dephlogisticated air" have been generally described by those who have written about them as the mere haphazard experiments of an amateur chemist working with neither plan nor vision.1 The author has however pointed out elsewhere2 that such a criticism of any part of Priestley's work is both inaccurate and unjust: and it is particularly so with regard to his experiments on "dephlogisticated air." Priestley was the first pneumatic chemist; and his discoveries proved so extensive and increased so rapidly from 1772 onwards that they became too large for publication in the Philosophical Transactions. He accordingly published them as Experiments and Observations on Different Kinds of Air (London, 3 vols., 1774, 1775 and 1777) and Experiments and Observations Relating to Various Branches of Natural

<sup>1</sup> Notable exceptions are Sir Philip Hartog in (1) "Joseph Priestley and his Place in the History of Science" (Proc. R. I., 1931, 26, 395), (2) "The Bicentenary of Joseph Priestley" (J. Chem. Soc., 1933, p. 896), and (3) Article on Joseph Priestley in Dic. Nat. Biog; and the late Professor A. N. Meldrum in "The Bicentenary of Joseph Priestley" (J. Chem. Soc., 1933, p. 902).

2 Science Progress, 1933, 28, 17.

3 For brevity, these volumes are referred to below as E. & O.

Philosophy; with a Continuation of the Observations on Air (London, 1 vol., 1779, and Birmingham, 2 vols., 1781 and 1786); and, because he believed in the immediate publication of everything relating to this new branch of chemistry, the experiments described in these six volumes were set down in the order in which they were carried out without revision or rearrangement, additions and corrections appearing in the later volumes, so that there is no apparent system in their presentation and careless readers are only too ready to conclude from hurried reference to his works that Priestley's researches were a series of disorderly and random experiments inspired by the blindest caprices of chance. If this were true, never did Fortune smile so handsomely on any investigator of Nature. It is, however, not true, although it is frequently defended by some passage or other from Priestley's works that has been torn from its context in order to justify this ill-informed criticism.

Turning to Priestley's account of the isolation of "dephlogisticated air" (E. & O., 1775, II, pp. 33-4), we find that he writes: "At the time of my former publication [E. & O., Vol. I], I was not possessed of a burning lens of any considerable force; and for want of one, I could not possibly make many of the experiments that I had projected, and which, in theory, appeared very promising. I had, indeed, a mirror of force sufficient for my purpose. But the nature of this instrument is such, that it cannot be applied, with effect, except upon substances that are capable of being suspended, or resting on a very

slender support. It cannot be directed at all upon any substance in the form of powder, nor hardly upon any thing that requires to be put into a vessel of quicksilver; which appears to me to be the most accurate method of extracting air from a great variety of substances. . . . But having afterwards procured a lens of twelve inches diameter, and twenty inches focal distance, I proceeded with great alacrity to examine, by the help of it, what kind of air a great variety of substances, natural and factitious, would yield, putting them into . . . vessels . . . which I filled with quicksilver, and kept inverted in a bason of the same. Mr. Warltire, a good chymist, and lecturer in natural philosophy, happening to be at that time in Calne, I explained my views to him, and was furnished by him with many substances, which I could not otherwise have procured."

It is evident from this that Priestley had planned a series of experiments in which various substances were to be heated by means of a burning lens in order to examine what kinds of air might be evolved from them. In obtaining this so-called "air" from bodies, he was repeating the work described by Stephen Hales<sup>1</sup> in his Vegetable Staticks (London,

<sup>1&</sup>quot; That the calces of metals contain air, of some kind or other, and that this air contributes to the additional weight of the calces, above that of the metals from which they are made, had been observed by Dr. Hales. . . . I had likewise found, that no weight is either gained or lost by the calcination of tin in a close glass vessel. . . ." (E. & O., 1774, I, p. 192). Hales had obtained "air" by heating red lead, and had concluded that, in the preparation of this substance from the metal, air was absorbed by the sulphureous particles of the fuel and that the combination thus produced was "lodged" in the metal and thereby increased its weight "about 1/20 part." Moreover, the redness of this substance indicated "the addition of plenty of sulphur in the operation" (Vegetable Staticks, London, 1727, pp. 286–7).

1727); and in projecting a study of the various "airs" that might be obtained from different bodies, he was proposing an obvious extension of his own researches, which had already led him to the discovery of "nitrous air," "diminished nitrous air," "acid air," "alkaline air" and "vitriolic acid air," that is, to the discovery of several "airs" that were chemically distinguishable from common air and from one another. There is therefore no haphazardness or lack of design in this work: it was "projected and . . . in theory, appeared very promising," as Priestley himself stated.

Priestley continues (p. 34): "With this apparatus, after a variety of other experiments, an account of which will be found in its proper place, on the 1st of August, 1774, I endeavoured to extract air from mercurius calcinatus per se; and I presently found that, by means of this lens, air was expelled from it very readily. Having got about three or four times as much as the bulk of my materials, I admitted water

¹ Priestley had obtained the burning lens in 1774 shortly after the publication of E. & O., Vol. I; and he had already made use of it in June 1774, to study the action of heat on various metals, calces and salts (E. & O., 1775, II, pp. 104-5). These experiments, to which he refers in the passage quoted above, are described in the same place (tbid., pp. 105-20): they were begun in June, 1774, and preceded the experiments that led to the isolation of "dephlogisticated air," although in Vol. II Priestley inserted his account of them after the section dealing with "dephlogisticated air." This work is clearly a part of his general study of the "air" obtained from bodies by heating them with a powerful burning lens: the experiments that led to the isolation of "dephlogisticated air" are another part of the same study, but they are evidently reported in a separate and preceding section because of their differing results, which Priestley very properly considered to be highly remarkable.

<sup>&</sup>lt;sup>2</sup> This substance was prepared by heating mercury nearly to its B.P. on a sand-bath for several months in a curiously shaped matrass with a very flat shallow body and an elongated neck terminating in a very narrow opening, the apparatus being commonly known as "Boyle's hell" (Lavoisier, *Traité Élémentaire de Chimie*, Paris, 1789, II, p. 520).

to it, and found that it was not imbibed by it. But what surprised me more than I can well express, was, that a candle burned in this air with a remarkably vigorous flame, very much like that enlarged flame with which a candle burns in nitrous air, exposed to iron or liver of sulphur; but as I had got nothing like this remarkable appearance from any kind of air besides this particular modification of nitrous air, and I knew no nitrous acid was used in the preparation of mercurius calcinatus, I was utterly at a loss how to account for it."

What Priestley was surprised at here was not the production of air, as some writers have thought, but the property of the air produced, namely, that it allowed a candle to burn in it " with a remarkably vigorous flame." Only once previously had he observed anything resembling this-in the case of "diminished nitrous air" (nitrous oxide), which he had obtained by the action of iron or liver of sulphur on "nitrous air" (nitric oxide). Accordingly, he was "utterly at a loss" how to account for this curiously vigorous burning of the flame, since he knew that no "nitrous" (nitric) acid was used in the preparation of the mercurius calcinatus. Moreover, he had already noted "that flame cannot subsist long without change of air, so that the common air is necessary to it, except in the case of substances, into the composition of which nitre enters; for these will burn in vacuo, in fixed air, and even under water, as is evident in some rockets, which are made for this purpose" (E. & O., I, p. 43).

Priestley noted too, though, as he says, he did

not give sufficient attention to it at the time, that the flame of the candle was larger and "burned with more splendor and heat" than it did in "that species of nitrous air," and added that "a piece of red-hot wood sparkled in it, exactly like paper dipped in a solution of nitre, and it consumed very fast: an experiment which I had never thought of trying with nitrous air "2 (p. 35). And he obtained some more of this air "with the very same property" from ordinary red precipitate, that is, from the precipitate obtained by adding alkali to a solution of mercury in spirit of nitre.

This last result suggested to Priestley that this remarkable property, resembling that of the modified "nitrous air," was in some way communicated by the "nitrous" (nitric) acid and that the mercurius calcinatus, which was prepared by heating mercury in air, had taken "something of nitre" from the atmosphere.

But, since his mercurius calcinatus had been bought "at a common apothecary's," he suspected that it "might, in fact, be nothing more than red precipitate." "However," he added, "mentioning this suspicion to Mr. Warltire, he furnished me with some that he had kept for a specimen of the preparation, and which, he told me, he could warrant to be genuine.

Nitrous oxide.

<sup>1</sup> Nitrous oxide.

2 Here also nitrous oxide is meant; Priestley had just written of it as a "species" or "modification" of "nitrous air." Later he referred to this gas as "diminished" and sometimes "phlogisticated nitrous air." The latter term was evidently derived from one of the methods of preparation used by Priestley, namely, the exposure of "nitrous air" (nitric oxide) to a moist mixture of iron-filings and brimstone (E. & O., I, p. 118), a method that, following Hales, he had already used to "diminish" common air (E. & O., I, p. 105). Priestley's discovery that a candle burned in this air is described in E. & O., I, pp. 215-6.

This being treated in the same manner as the former, only by a longer continuance of heat, I extracted much more air from it than from the other" (p. 36).

At this time too, Priestley obtained the same kind of air from red lead. One third of the product was soluble in water; this part was probably "fixed air," Priestley pointing out that he had previously (E. & O., 1774, I, pp. 192-3) obtained "fixed air" from red lead by heating it with a candle. In the remainder "a candle burned very strongly, and with a crackling noise" (p. 37).

But Priestley had no suspicion that his new "air" was "wholesome," that is, respirable, "so far," he says, " was I from knowing what it was that I had really found; taking it for granted, that it was nothing more than such kind of air as I had brought nitrous air2 to be by the processes above mentioned; and in this air I have observed that a candle would burn sometimes quite naturally, and sometimes with a beautiful enlarged flame, and vet remain perfectly noxious" (p. 37). The production of the new air instead of "fixed air" from red lead he ascribed to the heat of the lens being greater than that of the candle, and he noted that the part of the red lead where the focus fell turned yellow (ibid.). Moreover, the results of this experiment with red lead helped to confirm his suspicion "that the mercurius calcinatus must get the property of yielding this kind of air from the atmosphere, the process by which that preparation, and this of red lead is made, being similar " (p. 38).

<sup>&</sup>lt;sup>1</sup> Nitrous oxide.

<sup>&</sup>lt;sup>2</sup> Nitric oxide.

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Here, for the moment, Priestley's experiments ended. Later in the month (August, 1774) he set out to accompany his patron, Lord Shelburne, on a Continental tour. In October the party were in Paris, where Priestley met many of the leading French chemists of that time. His recent studies were still much in his mind. He says that the result of the experiment with Warltire's specimen " might have satisfied any moderate sceptic"; but, in Paris, he took the opportunity "to get an ounce of mercurius calcinatus prepared by Mr. Cadet, of the genuineness of which there could not possibly be any suspicion" (p. 36), and he carried it back to England for further study (p. 38). And it was on this visit to Paris in October, 1774, that Priestley frequently expressed to Lavoisier and others his "surprise at the kind of air" obtained from this substance (p. 36) and from red lead (p. 38), a communication that is of special interest to us here.

Priestley had thus isolated the gas we now know as oxygen in August, 1774, from mercurius calcinatus, red precipitate and red lead, and had observed its insolubility in water and its property of enhancing the combustion of a candle and a piece of red-hot wood; but he had no suspicion of its respirability, supposing it to be nothing more than nitrous oxide, a gas that he had already obtained in his previous studies. His surprise arose, not from the production of an "air" from mercurius calcinatus, but from the kind of air that was produced, because it resembled the "air" he had obtained from "nitrous air," although no "nitrous acid" was used in the

preparation of the *mercurius calcinatus*. And, in any case, the first chemist to isolate oxygen and to begin the study of its properties is well entitled to express his surprise.

After his return from abroad, Priestley began experiments with Cadet's mercurius calcinatus. He heated ½ oz. of it and obtained thereby one ounce measure of air that was neither re-absorbed by the solid from which it had been obtained nor dissolved by water. A candle burned in it "with a vivid flame." But what Priestley observed now (November 19, 1774) for the first time was that, whereas agitation in water for a few minutes deprived "modified nitrous air" (nitrous oxide) of its property of allowing a candle to burn in it, (owing to the solution of the nitrous oxide in the water leaving only nitric oxide), more than ten times as much such agitation produced no sensible change of this property in the new air, since "a candle still burned in it with a strong flame" and since "it did not, in the least, diminish common air, which I have observed that nitrous air, in this state, in some measure, does." Two days later the "air," which had been kept in contact with water, was subjected to violent agitation in water for five minutes, but "a candle still burned in it as well as in common air." This agitation would, he thought, have made the "phlogisticated nitrous air" fit for respiration, but not fit to support the combustion of a candle. Priestley, as will be seen in the next paragraph, was here misled by an earlier malobservation that all "airs" were made respirable

PRIESTLEY'S ISOLATION & RECOGNITION OF OXYGEN 163 by agitation in water (E. & O., 1775, II, pp. 38-9).

Accordingly, Priestley was "fully convinced . . . that there must be a very material difference between the constitution of the air from mercurius calcinatus, and that of phlogisticated nitrous air [nitrous oxide], notwithstanding their resemblance in some particulars" (pp. 39-40). He did not doubt that the former was fit for respiration after agitation in water, as, he alleged, was the case in other "airs" he had tested, but, he says: "I still did not suspect that it was respirable in the first instance; so far was I from having any idea of this air being, what it really was, much superior, in this respect, to the air of the atmosphere" (p. 40).

Thus, further experiments carried out in November, 1774, convinced Priestley that the new "air" was materially different from nitrous oxide, which he had first supposed it to be in August, 1774.

Priestley continues: "In this ignorance of the real nature of this kind of air, I continued from this time (November) to the 1st of March following.... But in the course of this month, I not only ascertained the nature of this kind of air, though very gradually, but was led by it to the complete discovery of the constitution of the air we breathe" (p. 40).

Between November 1774 and March 1775, Priestley had been carrying out experiments with "vitriolic acid air" and the various "airs" from spirit of nitre. When he wrote of his "ignorance of the real nature" of the new "air," he was clearly referring, as will be seen below, not to any ignorance of its being a new kind of "air," chemically different from other "airs," since he had, as shown above, already satisfied himself in November, 1774, that its constitution differed materially from that of "phlogisticated nitrous air" (nitrous oxide), but to his ignorance of its fitness for respiration.

Until March 1, 1775, wrote Priestley (p. 40), he had so little suspicion that the new "air" was respirable that he had not even thought of testing it with "nitrous air" (nitric oxide); but the observation that a candle burned in it after long agitation in water finally led him to carry out the test, and, he says: "Putting one measure of nitrous air to two measures of this air, I found, not only that it was diminished, but that it was diminished quite as much as common air, and that the redness of the mixture was likewise equal to that of a similar mixture of nitrous and common air" (p. 41). He now had no doubt that the new air was fit for respiration and that it had all the other properties of "genuine common air" (ibid.); but he admits that his notion that there was no air better than common air blinded him to the facts that the redness was deeper and the diminution greater in this test than they would have been with common air.

This discovery, namely, that "the test of nitrous air" showed that the air from mercurius calcinatus was fit for respiration, led Priestley to give up the hypothesis that he had formed at first "that the mercurius calcinatus had extracted spirit of nitre from the air": and he now concluded "that all the constituent parts of the air were equally, and in

their proper proportion, imbibed in the preparation of this substance, and also in the process of making red lead. For at the same time that I made the above-mentioned experiment on the air from mercurius calcinatus, I likewise observed that the air which I had extracted from red lead, after the fixed air was washed out of it, was of the same nature, being diminished by nitrous air like common air: but, at the same time, I was puzzled to find that air from the red precipitate was diminished in the same manner, though the process for making this substance is quite different from that of making the two others. But to this circumstance I happened not to give much attention" (pp. 41-2). What Priestley understood by "the constituent parts of the air" will appear later: in compiling the account of these experiments of March 1, 1775, he expressed the opinion that his rejection of the hypothesis first formed threw him "back into error," but, as will be seen below, neither of his explanations was satisfactory.

Next day (March 2, 1775), Priestley was surprised to find that a candle burned in the mixture from the previous day's test and indeed burned even better than in common air. The mixture had stood overnight and, if it had been common air, the maximum diminution would have taken place and the common air would have been rendered "perfectly noxious, and intirely unfit for respiration or inflammation." Priestley wrote that he made this experiment for reasons that he had forgotten, but he remembered that the result was entirely unexpected; and he added that he often carried out such

experiments on the slightest motives and, had he not had a lighted candle at this time, he would probably never have made the test and the whole train of experiments that followed might never have been attempted. This is one of the passages often quoted to exhibit Priestley, the random experimenter. But, what scientific investigator has not done the like? The difference in Priestley's case is that he frankly tells us that he did experiment in this way and, indeed, often. He planned the general outline of a series of experiments, as we have already seen; and, like any other investigator, he did not hesitate to try extempore experiments as his researches progressed. The repeated criticism of Priestley for making such experiments can surely only arise from a misapprehension of the realities of scientific research in the minds of those who are unfamiliar with those realities. Moreover, when we analyse Priestley's statement, we find that he states first of all that he had forgotten the reasons for which he made it: he does not say that he had no reasons, but that he had forgotten them. Further, the application of a lighted candle was a test that he customarily applied.1 Secondly, he says that the result was entirely unexpected: and well it might be, since on the previous day the "air" had been "diminished" by "nitrous air" in the standard

¹ See E. & O., I, pp. 25, 27, 44, 51, 58, 59, 64, 91, 127, 147, 155, 157, 158, 159, 160, 175, 178, 215, 216, 217, 219, 220, 236, 256, 257. These references are not intended to form an exhaustive list: and there seems little need to continue with references from Vol. II. The test was clearly one that Priestley regularly carried out in his experimental work. He has been taken too much at the letter of his own naïve word in such passages as this and others.

proportions as much as common air was "diminished," and yet now, on the next day, a candle still burned in it and, indeed, burned even better than in common air. Thirdly, he says that his motive in applying the candle was of the slightest and that, if he had not had a lighted candle by him at the moment, he might not have made the experiment and the whole train of his subsequent experiments might not have followed. There does not seem to us anything in this to convict Priestley of randomness in experimentation, but rather that he was acting under the best of scientific motives-" Try it and see." We may perhaps be excused from quoting his own words at this late stage, but we have chosen to do so in the hope that those readers who may have encountered this passage on other occasions may now give it what we regard as its correct interpretation. Priestley wrote: "I cannot, at this distance of time, recollect what it was that I had in view in making this experiment; but I know I had no expectation of the real issue of it. Having acquired a considerable degree of readiness in making experiments of this kind, a very slight and evanescent motive would be sufficient to induce me to do it. If. however, I had not happened, for some other purpose, to have had a lighted candle before me, I should probably never have made the trial; and the whole train of my future experiments relating to this kind of air might have been prevented "(p. 43).

But Priestley still thought that this property was "peculiar to air extracted from these substances, and adventitious" (p. 43): and at this time he

always spoke of his new product as substantially the same as atmospheric air. He told Dr. Price that he was perfectly satisfied that it was ordinary air on account of the result that he had obtained when testing it with "nitrous air": but, to satisfy others, he "wanted a mouse to make the proof quite complete" (p. 43).

On March 8, 1775, Priestley obtained a fullgrown mouse which he put into a glass vessel containing two ounce-measures of the "air" from mercurius calcinatus. Such a mouse would be expected to live for about a quarter of an hour in an equal amount of common air. In this air, however, the mouse lived "a full half hour." It was taken out apparently dead, but revived on being held to the fire and appeared to be none the worse for its experience. By this, Priestley was satisfied that the new "air" was "at least, as good as common air," but he says that he did not conclude that it was "any better" than common air, because "though one mouse would live only a quarter of an hour in a given quantity of air, I knew it was not impossible but that another mouse might have lived in it half an hour; so little accuracy is there in this method of ascertaining the goodness of air: and indeed I have never had recourse to it for my own satisfaction, since the discovery of that most ready, accurate, and elegant test that nitrous air furnishes. But in this case I had a view to publishing the most generallysatisfactory account of my experiments that the nature of the thing would admit of" (p. 44).

However, reflection on the result of the

experiment with the mouse led Priestley to suspect that this "air" was indeed better than common air and next day, March 9, 1775, he applied the "nitrous air" test to a portion of the residue of the air that the mouse had breathed. If it was common air, then it would be "very nearly, if not altogether, as noxious as possible" and would be unaffected by the addition of "nitrous air." But it proved to be better than common air; for, when mixed with "nitrous air" in the usual proportion of 2 to 1, it was diminished in the proportion of 4½ to 3½, a diminution of in a short time, whereas in a long time common air was never reduced more than 1 of its bulk by any proportion of "nitrous air" nor more than \( \frac{1}{4}\) " by any phlogistic process whatever" (p. 45).

Next day, March 10, 1775, Priestley added another measure of "nitrous air" to this same mixture and was astonished to find that it was thereby diminished almost to a half of its original bulk. A third measure produced no further diminution, but "left it one measure less than it was even after the mouse had been taken out of it" (p. 46). Priestley was now "fully satisfied that this air, even after the mouse had breathed it half an hour, was much better than common air" (p. 46). He had 11 ounce-measures of it left and he put the mouse into this residue. The mouse gave no signs of shock, as it would have done if the air had not been "very wholesome"; it remained "perfectly at its ease another full half hour" and was taken out at the end of that time "quite lively and vigorous." The air was measured next day (March 11, 1775) and was found to have been reduced from 1½ to ¾ of an ounce-measure, while the test with "nitrous air" showed that it was still nearly as good as common air. Moreover, the vigour of the mouse on its withdrawal showed that the air "could not have been rendered very noxious."

Priestley then procured another mouse and put it into less than two measures of a mixture of "air" from mercurius calcinatus and "air" from red precipitate, which he had found to be " of the same quality." This mouse lived for three-quarters of an hour, but the apparatus was not in a warm place and Priestlev suspected that the mouse died of cold. But it had lived three times as long as it probably would have lived in the same bulk of common air and, as Priestley did not expect much accuracy in these tests with mice, he made no more of them. He was now "fully satisfied" of the "superior goodness" of his new "air" and proceeded to measure its degree of purity by means of "nitrous air." He added one measure of "nitrous air" to two measures of the new "air," following his standard practice for testing the "goodness" of common air. The diminution was greater than would have occurred with common air. A second measure of "nitrous air" reduced the new "air" to 2 of its original bulk, and a third measure to ½. "Suspecting," wrote Priestley, "that the diminution could not proceed much farther, I then added only half a measure of nitrous air, by which it was diminished still more; but not much, and another half measure

made it more than half of its original quantity; so that, in this case, two measures of this air took more than two measures of nitrous air, and yet remained less than half of what it was. Five measures brought it pretty exactly to its original dimensions "(pp. 47-8).

The "air" from red precipitate was diminished in the same proportion as that from mercurius calcinatus, and 5 measures of "nitrous air" added to 2 measures of this " air " gave no increase in bulk. "Now," added Priestley, "as common air takes about one half of its bulk of nitrous air, before it begins to receive any addition to its dimensions from more nitrous air, and this air took more than four half-measures before it ceased to be diminished by more nitrous air, and even five half-measures made no addition to its original dimensions, I conclude that it was between four and five times as good as common air. It will be seen that I have since procured air better than this, even between five and six times as good as the best common air that I have ever met with " (p. 48).

Priestley was "now fully satisfied with respect to the nature of this new species of air." It could take more phlogiston from "nitrous air" than common air did and must therefore have originally contained less phlogiston than the latter. "Philosophically speaking," it was "much dephlogisticated." At this point Priestley carried out a long series of experiments on the different "preparations of lead, made by heat in the open air" and on some other substances (the calces of lead, various calces and earths moistened with spirit of nitre, and saltpetre) to see what kind of air they yielded, applying the test of "nitrous air" to the products (pp. 49–90).

Later in this volume (p. 87), Priestley drew attention to the fact that he had previously (E. & O., I, p. 155) extracted from saltpetre an "air" with properties that at the time appeared to him "very extraordinary." He had heated saltpetre in a gunbarrel and obtained an "air" in which a candle burned "with a very strong flame and with a crackling noise" and in which a candle still burned after the "air" had stood a whole year over water. At the time he had thought that this "air" was "phlogisticated nitrous air" (nitrous oxide), but he was now satisfied that it must have been "dephlogisticated air." He now found that saltpetre heated in a glass vessel gave "very pure dephlogisticated air."

Reference to these earlier experiments shows that Priestley had obtained "dephlogisticated air" at some date before November, 1771, but had not recognized it as a new "air." He found that a candle burned in it "just as in common air," while in another sample "a candle not only burned, but the flame was increased, and something was heard like a hissing, similar to the decrepitation of nitre in an open fire" (E. & O., I, p. 156). He thought that, in this latter sample, there were some particles of nitre present because the "air" was freshly made when the experiment was carried out and there had not been sufficient time for their deposition. In an

<sup>&</sup>lt;sup>1</sup> That it was standing over water is not stated until Vol. II, p. 87.

unsatisfactory test,¹ carried out on November 6, 1772, Priestley found that a sample of this air that had been standing over water for a year had become thoroughly noxious. "It made no effervescence with nitrous air, and a mouse died the moment it was put into it"; but, when the "air" was washed with rain-water for ten minutes," it was restored to its former perfectly wholesome state. It effervesced with nitrous air as much as the best common air ever does, and even a candle burned in it very

well " (E. & O., I, pp. 156-7).

To these passages, there is a curious footnote (E. & O., I, p. 155) referring to the "air" from saltpetre. It reads: "Experiments, of which an account will be given in the second part of this work, make it probable, that though a candle burned even more well in this air, an animal would not have lived in it. At the time of this first publication, however, I had no idea of this being possible in nature." By the "second part of this work," Priestley is referring, not to Vol. II of the E. & O., but to Part II of Vol. I, that is, to work carried out before the publication of Vol. II; and where he speaks of "the time of the first publication," he is referring to the previous publication of this work in the Philosophical Transactions (1772, 62, 147).2 Further, the "experiments, of which an account will be given in the second part of the work" are worthy of notice here for more than one reason. Proceeding 1 Satisfactory to Priestley, on account of the incorrect observation referred

which it refers being carried out later.

to on pp. 162-3 above.

2 Vol. I was published in 1774: the *Preface* is dated "Feb. 1774." The footnote was, of course, not inserted in the *Phil. Trans.*, the experiment to

from the well-known fact (established by Lane)1 that an aqueous solution of "fixed air" dissolved iron, Priestley "had the curiosity to try whether fixed air alone would do it; and as nitrous air is of an acid nature, as well as fixed air, I, at the same time, exposed a large surface of iron to both the kinds; first filling two eight ounce phials with nails, and then with quicksilver, and after that displacing the quicksilver in one of the phials by fixed air, and in the other by nitrous air; then inverting them, and leaving them with their mouths immersed in basons of quicksilver. In these circumstances the two phials stood about two months, when no sensible change at all was produced in the fixed air, or in the iron which had been exposed to it, but a most remarkable, and most unexpected change was made in the nitrous air; and in pursuing the experiment, it was transformed into a species of air, with properties which, at the time of my first publication on this subject, I should not have hesitated to pronounce impossible, viz. air in which a candle burns quite naturally and freely, and which is yet in the highest degree noxious to animals, insomuch that they die the moment they are put into it; whereas, in general, animals live with little sensible inconvenience in air in which candles have burned out. Such, however, is nitrous air, after it has been long exposed to a large surface of iron. It is not less extraordinary, that a still longer continuance of nitrous air in these circumstances (but how long depends upon too many, and too minute circumstances to be

<sup>1</sup> Phil. Trans., 1769, 59, 216.

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ascertained with exactness) makes it not only to admit a candle to burn in it, but enables it to burn with an enlarged flame, by another flame (extending everywhere to an equal distance from that of the candle, and often plainly distinguishable from it) adhering to it. Sometimes I have perceived the flame of the candle, in these circumstances, to be twice as large as it is naturally, and sometimes not less than five or six times larger; and yet without any thing like an explosion, as in the firing of the weakest inflammable air. Nor is the farther progress in the transmutation of nitrous air, in these circumstances, less remarkable. For when it has been brought to the state last-mentioned, the agitation of it in fresh water almost instantly takes off that peculiar kind of inflammability, so that it extinguishes a candle, retaining its noxious quality. It also retains its power of diminishing common air in a very great degree."1

While it is worth observing here that this passage gives an excellent example of Priestley the investigator at his work, having that "curiosity to try" that is the spearhead of most, if not all, scientific advance, it is more relevant to us at this moment to note that he had here converted nitric oxide into nitrous oxide and had thereby obtained a gas that possessed what were to him the almost contradictory properties of supporting the combustion of a candle and of being noxious to animal life. This was, it appears to us, the observation that ultimately led to Priestley's delay from November, 1774, to

March, 1775, in recognizing the respirability of oxygen after he had isolated it and begun to study its properties in August, 1774. Moreover, earlier in his work, he had assumed that combustion and respiration were processes that had the same effects on air, the "consumption" of air in either being "of the same nature, and in the same degree" (E. & O., I, p. 47). But, when he found that animals lived nearly as long in air in which candles had burnt out as they lived in common air, an observation, which, as he noted, had been previously made by Boyle<sup>1</sup> and others, he abandoned this view with, as will be seen, unfortunate results: and he had concluded, it is interesting to note in passing, that it was probable "that burned air is air so far loaded with phlogiston, as to be able to extinguish the candle, which it may do long before it is fully saturated " (E. & O., I, p. 117).

To the section of Vol. II of the Experiments and Observations dealing with the isolation of the gas we now know as oxygen Priestley gave the title: Of Dephlogisticated Air, and of the Constitution of the Atmosphere. Here, almost at the beginning, he says that: "There are, I believe, very few maxims in philosophy that havelaid firmer hold upon the mind, than that air, meaning atmospherical air (free from various foreign matters, which were always supposed to be dissolved, and intermixed with it) is a simple elementary substance, indestructible, and unalterable, at least as much so as water is supposed to be. In the course of my inquiries, I was, however,

<sup>&</sup>lt;sup>1</sup>Cf. p. 143, footnote 2, above.

PRIESTLEY'S ISOLATION & RECOGNITION OF OXYGEN 177 soon satisfied that atmospherical air is not an unalterable thing; for that the phlogiston with which it becomes loaded from bodies burning in it, and animals breathing it, and various other chemical processes, so far alters and depraves it, as to render it altogether unfit for inflammation, respiration, and other purposes to which it is subservient; and I had discovered that agitation in water, the process of vegetation, and probably other natural processes, by taking out the superfluous phlogiston, restore it to its original purity. But I own I had no idea of the possibility of going any farther in this way, and thereby procuring air purer than the best common air. I might, indeed, have naturally imagined that such would be air that should contain less phlogiston than the air of the atmosphere; but I had no idea that such a composition was possible" (pp. 30-1). And, he adds: "Several of the known phenomena of the nitrous acid [nitric acid] might have led me to think, that this was more proper for the constitution of the atmosphere than the marine acid: but my thoughts had got into a different train, and nothing but a series of observations, which I shall now distinctly relate, compelled me to adopt another hypothesis, and brought me. in a way of which I had then no idea, to the solution of the great problem, which my reader will perceive I have had in view ever since my discovery that the atmospherical air is alterable, and therefore that it is not an elementary substance, but a composition, viz. what this composition is, or what is the thing that we breathe, and how is it to be

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made from its constituent principles" (pp. 32-3).

Accordingly, when he found that he obtained the air that he here begins to speak of as "dephlogisticated air" from various calces and earths moistened with "nitrous acid" (nitric acid), he wrote: "I was the more confirmed in my idea of spirit of nitre and earth constituting respirable air, by finding, that when any of these matters, on which I had tried the experiment, had been treated in the manner above mentioned, and they had thereby yielded all the air that could be extracted from them by this process; yet when they had been moistened with fresh spirit of nitre, and were treated in the same manner as before, they would yield as much dephlogisticated air as at the first. This may be repeated till all the earthy matter be exhausted" (pp. 55-6).

Later, he noted further: "The hypothesis maintained in this section, viz. that atmospherical air consists of the nitrous acid and earth, suits exceedingly well with the facts relating to the production of nitre; for it is never generated but in the open air, and by exposing to it such kinds of earth as are known to have an affinity with the nitrous acid; so that by their union common nitre may be formed. Hitherto it has been supposed by chymists, that this nitrous acid, by which common nitre is formed, exists in the atmosphere as an extraneous substance, like water, and a variety of other substances, which float in it, in the form of effluvia; but since there is no place in which nitre may not be made, it may, I think, with more probability be

supposed, according to my hypothesis, that nitre is formed by a real decomposition of the air itself, the bases that are presented to it having, in such circumstances, a nearer affinity with the spirit of nitre than that kind of earth with which it is united in the atmosphere" (p. 60). As for the "fixed air" that appeared in combustion Priestley had already supposed "that flame disposes the common air to deposit the fixed air it contains; for if any limewater be exposed to it, it immediately becomes turbid. This is the case, when wax candles, tallow candles, chips of wood, spirit of wine, ether, and every other substance which I have yet tried, except brimstone, is burned in a close glass vessel, standing in lime-water. This precipitation of fixed air (if this be the case) may be owing to something emitted from the burning bodies, which has a stronger affinity with the other constituent parts of the atmosphere" (E. & O., I, p. 44). In subsequent experiments on respiration he concluded that "the compound mass of the air" was disposed "to part with some constituent part belonging to it (which appears to be the fixed air that enters into its constitution) " (*ibid.*, pp. 78–9).

Priestley then carried out some experiments to determine the density of "dephlogisticated air" and found it to be "a little heavier than common air" (E. & O., II, pp. 91-4) and others which show that "dephlogisticated air" mixed so readily and completely with "phlogisticated air, or air injured by respiration, putrefaction, &c.," that "the purity of the mixture may be accurately known from the

quantity and quality of the two kinds of air before mixture" (p. 97). "Thus," he wrote, "if one measure of perfectly noxious air be put to one measure of air that is exactly twice as good as common air, the mixture will be precisely of the standard of common air." And there was no change in volume on mixture—" I observed also, in making this experiment, that after mixing one measure of each of these kinds of air, they made exactly two measures." But Priestley did not carry out many experiments on the mixing of "dephlogisticated air" with other kinds of air "because the analogy which it bears to common air is so great, that I think any person may know before-hand, what the result of such experiments would be." He pointed out, too, that this "pure air" might be used to purify the air of rooms "in which much company should be confined " (p. 98); and he noted that a mixture of "little more than one-third of highly dephlogisticated air, and the rest inflammable air " exploded with considerable noise and violence and with the production of much heat. He thought that the new air might be used to increase the explosive force of gunpowder and to "augment the force of fire to a prodigious degree." The latter experiment he tried by forcing this air from a bladder through a glass tube on to a piece of lighted wood, and he mentioned it to Michell who thought that platina

¹ The total bulk of the mixture is 2 measures—I of nitrogen and I of air that is twice as good as common air, i.e., air consisting of \{\frac{1}{2}\} nitrogen and \{\frac{1}{2}\} oxygen. The mixture will consist of \{\frac{1}{2}\} measures of nitrogen and \{\frac{1}{2}\} measures of oxygen, i.e., 80 per cent nitrogen and 20 per cent oxygen, "the standard of common air."

might be melted by using such a blast. Here also he supposed that the new "air" might be useful in the treatment of respiratory diseases, a therapeutic procedure that has become a commonplace in modern times: "From the greater strength and vivacity of the flame of a candle, in this pure air, it may be conjectured, that it might be peculiarly salutary to the lungs in certain morbid cases, when the common air would not be sufficient to carry off the phlogistic putrid effluvium fast enough. But, perhaps, we may also infer from these experiments, that though pure dephlogisticated air might be very useful as a medicine, it might not be so proper for us in the usual healthy state of the body: for, as a candle burns out much faster in dephlogisticated than in common air, so we might, as may be said, live out too fast, and the animal powers be too soon exhausted in this pure kind of air. A moralist, at least, may say, that the air which nature has provided for us is as good as we deserve" (p. 101). He tried the experiment on himself: "My reader will not wonder, that, after having ascertained the superior goodness of dephlogisticated air by mice living in it, and the other tests above mentioned, I should have the curiosity to taste it myself. I have gratified that curiosity, by breathing it, drawing it through a glass-syphon, and, by this means, I reduced a large jar full of it to the standard of common air. The feeling of it to my lungs was not sensibly different from that of common air; but I fancied that my breast felt peculiarly light and easy for some time afterwards. Who can tell but that, in time, this pure air may become a fashionable article in luxury. Hitherto only two mice and myself have had the privilege of breathing it " (p. 102).

Thus Priestley had isolated on Monday, August 1, 1774, a new "air," which had properties resembling those of "phlogisticated nitrous air" and which was distinct from other "airs" that he knew in that it was at once insoluble in water and yet a vigorous supporter of combustion.

But it was not until November 21, 1774, that he concluded that it was materially different from "phlogisticated nitrous air." Then on March 1, 1775, he discovered that it was respirable and therefore concluded that it was common air: on March 8, he suspected that it was "better" than common air and a week later he named it "dephlogisticated air" or air that had been deprived of its phlogiston, announcing his discovery in a letter to Sir John Pringle, P.R.S., written on March 15, 1775 (Phil. Trans., 1775, 65, 384), the relevant passage in which reads: "At the time of my last publication I had not a large burning lens; and as the focus of the mirror cannot be thrown upon anything in the form of a powder, or that requires a solid support, my experiments with the solar rays were exceedingly incomplete. I have now procured one of twelve inches in diameter; and the use of it has more than answered my highest expectations. The manner in which I have used it has been to throw the focus upon the several substances I wished to examine, either in vacuo, or, when confined by quicksilver, in PRIESTLEY'S ISOLATION & RECOGNITION OF OXYGEN 182 vessels filled with that fluid, and standing with their mouths immersed in it. I presently found that different substances yield very different kinds of air by this treatment. . . . But the most remarkable of all the kinds of air that I have procured by this process is, one that is five or six times better than common air, for the purpose of respiration, inflammation, and, I believe, every other use of common atmospherical air. As I think I have sufficiently proved, that the fitness of air for respiration depends upon its capacity to receive the phlogiston exhaled from the lungs, this species may not improperly be called dephlogisticated air. This species of air I first produced from mercurius calcinatus per se, then from the red precipitate of mercury, and now from red lead. The two former of the substances yield it pure; but the red lead I have generally met with yields a greater proportion of fixed air along with it. Another quantity, however, gave this air and hardly any thing else. On what this difference depends I cannot tell; but hope to be able to investigate. That this air is of that exalted nature, I first found by means of nitrous air, which I constantly apply as a test of the fitness of any kind of air for respiration, and which I believe to be a most accurate and infallible test for that purpose. Applying this test, I found, to my great surprise, that a quantity of this air required about five times as much nitrous air to saturate it, as common air requires. Common air is diminished about one-fifth, by a mixture of one-half nitrous air; but one quantity of this air was diminished one-half, and another two-thirds, by the addition of twice as much nitrous air; and three times the quantity, left it little more than it was at the first. A candle burned in this air with an amazing strength of flame; and a bit of red-hot wood crackled and burned with a prodigious rapidity, exhibiting an appearance something like that of iron glowing with a white heat, and throwing out sparks in all directions. But to complete the proof of the superior quality of this air, I introduced a mouse into it; and in a quantity in which, had it been in common air, it would have died in about a quarter of an hour, it lived, at two different times, a whole hour, and was taken out quite vigorous; and the remaining air appeared to be still, by the test of nitrous air, as good as common air.... My conjectures concerning the cause of these appearances are as yet too crude to lay before the Society. My present ideas . . . are, that, together with other observations which I shall lay before the publick, they afford some foundation for supposing that the nitrous acid is the basis of common air. and that nitre is formed by a decomposition of the atmosphere. But it is possible I may think otherwise to-morrow. It is happy, when with a fertility of invention sufficient to raise hypotheses, a person is not apt to acquire too great attachment to them. By this means they lead to the discovery of new facts, and from a sufficient number of these the true theory of nature will easily result."1

This letter was written in London on March 15,

<sup>1</sup> Phil. Trans., 1775, 65, 386-9.

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1775: it was read before the Royal Society on March 23, 1775,1 which is therefore the date of the first public announcement of Priestlev's isolation and recognition of "dephlogisticated air" oxygen, since Volume II of the Experiments and Observations was not published until much later in the year, as is evident from the date "Nov. 1775" printed on the dedication page.

On April 1, 1775, Priestley wrote from Calne, Wiltshire,2 to Dr. Price that "by the heat of the flame of a candle. . . . I get the pure air I discovered in London in great plenty, from a variety of cheap materials." This letter was read to the Royal Society on April 6, 1775.8

On May 24, 1775, Priestley wrote from London a further letter to Sir John Pringle in which he stated that he had now obtained "dephlogisticated air" from various earths moistened with nitric acid and further that: "Upon the whole, I think it may safely be concluded, that the purest air is that which contains the least phlogiston: that air is impure (by which I mean that it is unfit for respiration, and for the purpose of supporting flame) in proportion as it contains more of that principle. . . . "4 This letter was read to the Society on May 25, 1775.5

Thus it is evident that, while Priestley had first obtained his "dephlogisticated air" in 1771, he made no significant experiments until he obtained it from mercurius calcinatus on August 1, 1774; that

<sup>&</sup>lt;sup>1</sup> Phil. Trans. (1775, 65, 384) gives date May 25, but Hartog (Nature, 1933, 132, 25) has pointed out that the correct date is March 23.

<sup>2</sup> From the country house of his patron, Lord Shelburne.

<sup>3</sup> Hartog, loc. cit.

<sup>4</sup> Phil. Trans, 1775, 65, 392.

<sup>5</sup> Hartog, loc. cit.

he did not conclude that it was materially different from "phlogisticated nitrous air" until November 21, 1774; that he discovered its respirability on March 1, 1775, and therefore concluded that it was common air; that on March 8, 1775, he suspected that it was "better" than common air; and that on March 15, 1775, he announced to Pringle his discovery that it was "five or six times better than common air, for the purpose of respiration, inflammation, and, I believe, every other use of common atmospherical air" and named it "dephlogisticated air."

There has been some doubt as to where Priestley first isolated his "dephlogisticated air" and discovered its characteristic properties.1 The historical importance of his work justifies some reference to this matter. The experiments of August 1, 1774, from the reference to Warltire being in Calne at that time, appear to have been carried out at Calne. Yet Priestley, in a letter to Price quoted above, refers to "the pure air I discovered in London." In this letter, however, he is clearly referring to his discovery of the respirability of the new air. As the matter now stands, it is evident that the experiments of August, 1774, were carried out at Calne, since the dedication page of Priestley's Examination of Dr. Reid's Inquiry into the Human Mind, etc. (London, 1774), is inscribed "Calne, August 10, 1774 "2; and that Priestley discovered the respirability of the new air in March, 1775,

<sup>&</sup>lt;sup>1</sup> See Caven, Nature, 1933, 132, 25; and Hartog, ibid., pp. 25-6.

<sup>2</sup> The author owes this reference to the kindness of Sir Philip Hartog, to whom it was communicated by the late Professor Meldrum.

Empiricism (London, 1775) Priestley mentions his being in London at "some time in January"

<sup>2</sup> Rutt gives letters written from Calne on May 31 and June 4, 1774 (Life and Correspondence of Joseph Priestley, London, 1831, I, pp. 231 and

<sup>&</sup>lt;sup>1</sup> These letters with other letters from Priestley to Lindsey are now preserved in Dr. Williams's Library, Gordon Square, W.C.1. The author expresses his thanks to the Trustees of the Library for kindly allowing him to consult these letters and to publish information taken from them; and to the Trustees' Librarian, Mr. Stephen K. Jones, B.A., for his assistance in tracing the facts sought.

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3</sup> He left England for the Continent some time in August and was already in Lille on August 26, 1774 (Rutt, ibid., p. 237).

(p. 23) and on February 6 and 7 (pp. 23-4), in Calne on March 30, and again in London on May 23, on which date he showed his experiments at Shelburne House (p. 4) — the day before he wrote the second letter to Pringle that we have noted above on p. 185. Together with the dates and places given in the three letters published in the Philosophical Transactions, these facts support the conclusion that Priestley was at Calne on August 1, 1774, and that he made his discovery of the respirability of oxygen on March 1, 1775, in Shelburne House, now Lansdowne House, Mayfair, perhaps a curious locality for the scientific researches of an 18th-century Unitarian divine who had unpopular democratic notions in his head and was presently to be elected an honorary citoyen of the French Republic.

We have already noted Priestley's statement that he informed Lavoisier of his experiments of August 1774, when they met in Paris in October of that year.¹ In the next chapter we shall return to consider the use to which Lavoisier turned this information at a time when he was baffled with the problems of combustion and calcination.

<sup>&</sup>lt;sup>1</sup> Priestley was in Paris for several weeks. He wrote from there on October 6 and 21, 1774 (Rutt, op. cit., pp. 242, 251); and he did not return to London until early in November, as already noted above.

## CHAPTER VI

## LAVOISIER'S MEMOIRS, 1774-5, AND THEIR REVISION: THE NEW THEORY, 1777-8

LAVOISIER continued his experiments on calcination in 1774. In the Opuscules he had already suggested that, if his theory was correct, the calcination of metals in sealed vessels should take place without increase of weight. Of his experiments between August 1773 and March 1774, we have no record, since his laboratory note-book for that period, as indicated previously, is missing: and the note-book for the next period contains no reference to experiments in sealed vessels. However, on April 14, 1774, Lavoisier submitted to the Academy a memoir on this work which was read at the rentrée publique of November 12, 1774, and published in the December issue of the Observations (1774, 4, 446). Later it appeared in the Mémoires for 1774 (p. 351), published in 1778, and in a form that had obviously been revised in the light of the advances that had been made between November 12, 1774, when it was first read, and May 10, 1777, when the revised version was submitted to the Academy. The contents of the two memoirs differ in notable ways.

<sup>1</sup> Opuscules, p. 294; Essays, p. 340.

In the original memoir, as published in the Observations, Lavoisier referred to the experiments on the calcination of metals by a burning lens described in the Opuscules, in which there was a diminution of the air confined over water or mercury and an increase in the weight of the metals nearly proportional to the diminution of the air, this proportionality, however, as noted above, being a mere supposition: and added that he thought it was possible to conclude thence that a part of the air, or of some matter contained in the air, combined with the metals in their calcination and thereby increased their weight, this theory being confirmed by his observation that an evolution of an elastic fluid, a kind of air, identifiable with fixed air, occurred in the reduction of calces with charcoal. He then pointed out that Boyle had studied the calcination of metals in sealed vessels, observed an increase in the weight of the metals calcined and ascribed that increase to the addition of fireparticles that had passed through the pores of the glass.1 His own explanation differed from that suggested by Boyle. He therefore argued that, if Boyle was correct, the weight of the sealed vessel and its contents after heating and before opening would be greater than it was before it was heated; but, if the increase in the weight of the metal was due to fixation of air or of some matter contained in the sealed vessel, there would be no increase in the total weight, but the vessel would be partly emptied of its air and the increase of weight would

<sup>&</sup>lt;sup>1</sup> Cf. p. 63 above.

occur only when air was re-admitted to take the place of that which was missing.1 Accordingly, he put lead in small pieces and in quantities of 8 onces into each of several glass retorts of various sizes; and he treated equal weights of tin in the same way. The retorts were sealed, weighed on a sensitive balance and then heated on a charcoal furnace, very carefully and slowly to avoid fracture by the expansion of the air consequent on heating. In this way the metals were maintained "in complete fusion" for two hours.2 He noted that calcination appeared to cease after an hour and that more calx was formed in the large retorts than in the small ones. When the retorts had cooled and before they were opened, they were carefully re-weighed and showed not the least difference in their weight: but, at the moment when the point of the seal was broken, a slow whistling noise was heard as the external air went in, this noise being plus rapide, plus fort & plus long in the large than in the small retorts. Further, when the retorts were re-weighed after this admission of air, they all showed an increase in weight, an increase in fairly exact proportion to their capacities and, in every case, exactly equal to the gain in weight detected in the

<sup>&</sup>lt;sup>1</sup> Father Cherubin of Orléans had already pointed out in 1679 that Boyle ought to have weighed his sealed vessels before opening them and contended that Boyle's results showed that it was air and not fire that produced the increase in weight (Dissertation sur l'Imperméabilité du Verre, etc., Paris, 1679; quoted in Gobet's edn. of Jean Rey's Essays, Paris, 1777, pp. 213-16).

<sup>2</sup> Some of Lavoisier's phrases closely resemble those of Boyle: times and amounts, too, are identical. And he warns his readers in a footnote that the

<sup>&</sup>lt;sup>2</sup> Some of Lavoisier's phrases closely resemble those of Boyle: times and amounts, too, are identical. And he warns his readers in a footnote that the experiments are dangerous owing to explosions. Boyle had reported that one of his vessels "suddenly broke in a great multitude of pieces, and with a noise like the Report of a Gun; but (thanks be to God) it did no harm neither to me nor others that were very near it."

metal when weighed separately. Therefore, concluded Lavoisier, it evidently follows from these experiments "that the increase in the weight of metals calcined in sealed vessels comes neither from the matter of fire nor from any matter outside the retort, but that it is from the air contained in the vessel that the metal takes the substance that increases its weight and converts it into calx; and that what deceived Boyle was this, that in all his experiments he had neglected to weigh the vessel before opening it and he had attributed to the matter of fire an increase of weight that actually came from that part of the external air that entered the vessel."

In the final paragraph, Lavoisier explained that experimental details would have to be reserved for the ordinary meetings of the Academy, a rentrée publique evidently imposing limits to communications read on such occasions; and he would also have to omit any reference to the experiments that he made on the air in which metals had been calcined, air which was deprived of its fixable part-"I could almost say of the acid part that it contains"-air that had been in some manner decomposed, although his experiments had suggested a means of analysing the fluid that constituted the atmosphere and of examining its constituent principles. His results might not be entirely satisfactory, but he thought he could assert that even the purest air, air free from all moisture and matter foreign to its composition, was far from being a simple substance or element, as was commonly supposed, and must be, on the contrary, classified among the mixtures and perhaps even among the compounds.

This memoir led to a number of incidents that must have surprised Lavoisier. News of his experiments had got abroad; for on November 12, the very day of the reading, Beccaria wrote to him from Turin to point out that fifteen years previously he had studied the calcination of tin in a sealed vessel and had found that the calcination was limited, proceeding only in proportion to the capacity of the apparatus, that the total weight remained constant, that the various flasks that he had attached to the vessel during the different experiments all showed a decrease of weight after the calcination and that Cigna had referred to his work with tin and lead in the second volume of the memoirs of the Turin Academy. Lavoisier arranged with Rozier to publish the extract from Beccaria's letter relating to the experiments, together with the relevant extract from Cigna's memoir, and prefaced these with a letter and a note.1 In the letter Lavoisier asked that these communications should be published with his own memoir, on the following page if possible, so that it might be made clear that he had no wish to appropriate the discoveries of others: and he admitted that Beccaria had anticipated him in these experiments, expressed his pleasure at finding confirmation by such a distinguished physicist of a theory which he thought he had been the first to develop and which, however, appeared as solidly established as any fact could be in physics and chemistry. The note asserted that the memoir had been drawn up several months earlier and initialled by the Secretary of the Academy on April 14 of that year, that Lavoisier did not know of Boyle's experiments at that time or of Priestley's experiments in sealed vessels, that he had not heard of Beccaria's work until the receipt of his letter and that he wished to give the latter the justice that was due to him.

Lavoisier was evidently disturbed over this matter. It seems as if he was not given to reading original memoirs. But his position was secure here; for the theory that he was expounding on the gain in weight of metals on calcination was already contained in its germinal form in the work of Black and of Hales, but nothing of any significance had hitherto been made of it, and Lavoisier was taking it up with the intention of developing it alongside the study of the gases because it seemed to him "destined to bring about a revolution in physics and chemistry." But his enthusiastic belief that he was the first to suggest that air played a part in calcination received a severe set-back.

And there was more to follow. Bayen was studying the calx of mercury at the same time as Lavoisier was working on the *Opuscules*. He published four papers on his experiments, two<sup>2</sup> between the publication of the *Opuscules* and the reading of

<sup>&</sup>lt;sup>1</sup> E. & O., 1774, I, p. 192. <sup>2</sup> Observations, 1774, 3, 127 and 278.

Lavoisier's memoir and two1 in 1775. In the first paper, published in February, 1774, he had found that the calx of mercury could be converted into the metal by merely heating it without the addition of charcoal, that is, without supplying phlogiston, and that it lost weight in the process2: and he thought his results bore on the work reported by Lavoisier in the Opuscules on the gain in weight of metals on calcination. In his second paper he found that the elastic fluid obtained by heating calx of mercury with charcoal was soluble in water and had other properties of fixed air<sup>3</sup>; that, when he heated the calx without charcoal, a gas was evolved, which had the same properties4; and, again on heating the calx without charcoal, he obtained a gas insoluble in water, an apparent anomaly that he ascribed to the impurity of the sample of oxide used. Bayen was evidently much interested in the problem of calcination and was closely following Lavoisier's work. In the course of his reading, some time after the publication of his second paper, Bayen found in the collection of chemical works owned by de Villiers an imperfect copy of Jean Rey's Essays sur la Recherche de la cause pour laquelle l'Estain et le Plomb augmentent de poids quand on les calcine (Bazas, 1630), in which the author had explained the increase of weight of metals on calcination as due

<sup>5</sup> Ibid., pp. 289-92.

<sup>&</sup>lt;sup>1</sup> Ibid., 1775, 5, 147 and 6, 487. These two papers were mainly concerned with showing that the calces of mercury used by Bayen which had been obtained by precipitation were true calces, a point of dispute at that time.

<sup>&</sup>lt;sup>3</sup> Ibid., pp. 284-5.

<sup>4</sup> Ibid., pp. 287-8. He may not have cleared his apparatus of the fixed air from the previous experiments.

to the calces taking up air that had been rendered denser in the heating. Bayen then found a perfect copy of this rare book in the Bibliothèque du Roi: and he published in Rozier's Journal for January 1775 a letter drawing attention to the fact that Rev<sup>2</sup> had already propounded in the seventeenth century the theory that was now being advanced by contemporary chemists and inviting Rozier to let the chemists of all countries know that "it was a Frenchman who by the power of his genius had first divined the cause of the gain in weight shown by certain metals when exposed to the action of fire and converted into calces; and that this cause was precisely the same as that the truth of which had just been proved by the experiments described by M. Lavoisier at the last public seance of the Academy of Sciences."

It may be noted in passing that the neglect of Rev's work up to this time is very curious, especially as it was known to Mersenne. Its re-discovery aroused great interest in France, and two years later Rey's book was reprinted by Gobet (Paris, 1777). From the indifferent details given by Gobet, we have been able to trace some references to Rey's book previous to 1775. It is mentioned twice, as the seventh and tenth items, in the right-hand column of the thirty-third page of the section entitled Philosophi, Medici, Anatomistæ. . . . Scriptores in

<sup>&</sup>lt;sup>1</sup> Ibid., 1775, 5, 47.
<sup>2</sup> Little is known about Rey except that he practised medicine in Périgord, corresponded with Mersenne, and died about 1645.
<sup>3</sup> Rey's theory was crude; he supposed that the thickened air adhered to the calx, not that air combined with the metals to give calces.

Trichet's unpaginated Catalogus Librorum Bibliothecæ Raphaelis Tricheti du Fresne (Paris, 1662). It occurs again as a livre rare in a list of Auteurs connus seulement depuis l'impression de ce Catalogue inserted at the beginning of the third volume of Lenglet-Dufresnoy's Histoire de la Philosophie hermétique (Paris, 1742). And Spielmann recommended it to his students (Institutiones Chemia, 2nd edn., Strasbourg, 1766: French trans. by L. C. Cadet, Instituts de Chymie, Paris, 1770, II, p. 120). These facts, especially the last, make it very curious that Rev's work was unknown to the French chemists in 1774. Spielmann was well known and his book was much valued: he had entertained Lavoisier at Strasbourg, and during his visit Lavoisier had bought a large number of books on chemistry.1

Lavoisier thus found himself anticipated once more, but in the revival of Rey's work, the essential improvement made by Lavoisier in the theory seems for the moment to have escaped even the notice of Lavoisier himself. Others may have, indeed did, anticipate him in the data of these experiments and in crude theorizings that the increase in weight of metals on calcination was due to air, but he was the first to realize that this theory, applied alongside a study of the gases, was capable of working great changes in the science of chemistry.

We have seen that Lavoisier recognized the work of Beccaria: he did likewise with Rey when preparing his *Mémoires de Chimie*, which were published posthumously.<sup>8</sup> But he ignored Bayen's work in the

<sup>&</sup>lt;sup>1</sup> Cf. p. 29 above.

<sup>2</sup> Œuvres, II, p. 100.

memoir read at the Academy on November 12, 1774, and in the revised version published in the Mémoires of the Academy for 1774,1 where he ignored Rey as well, although he mentioned Beccaria again in this place. It is curious that there is no reference to Bayen, because the reduction of the calx of mercury without the addition of charcoal was a matter of some interest, apart from Bayen's work. For instance, on November 19, 1774, together with Sage and Brisson, Lavoisier had presented a report to the Academy establishing the contention of Cadet, announced before the Academy on September 3, 1774, that the precipitate of mercury per se2 was reducible to mercury by mere heating without the addition of phlogiston. The report<sup>3</sup> stated that this was recognized by chemists, with the exception of Baumé, who held that phlogiston must be supplied to effect the reduction4; and that it was found in experiments, at which Macquer, Le Roy and Bossut were present, that samples of this substance obtained from Cadet and Baumé were both reduced to mercury on heating without addition of other matter.

Moreover, as we have noted in the previous chapter, Lavoisier had been informed by Priestley in October, 1774 of the experiments carried out on this substance and on red lead in August, 1774, in

<sup>&</sup>lt;sup>1</sup> Published in 1778.

<sup>&</sup>lt;sup>2</sup> Red oxide of mercury, obtained by heating mercury in air for long periods, also called mercurous calcinatus per se, the substance Priestley had used in August, 1774, in experiments the results of which he communicated to Lavoisier in Paris in October, 1774.

<sup>8</sup> Œuvres, IV, p. 188.

<sup>4</sup> Chymie Expérimentale et Raisonné, Paris, 1773, II, p. 390.

which Priestley had found that an air with surprising properties was evolved by heating these substances with a burning lens. The effect of Priestley's communication will be discussed presently. Meanwhile, we may note that at the end of the memoir read on November 12, 1774, Lavoisier spoke of the "fixable part" of the air as being its "acid part" and of air being a mixture and possibly a compound. He had previously treated air as a mixture, deriving, we have suggested, this idea from Black; but we have not met the suggestion about the "acid part" before this. Priestley, as we have seen, had supposed that the mercurius calcinatus had extracted spirit of nitre from the air, and in his customary way he may have expressed this view to the chemists he met in Paris in October, 1774.1

It is here that we begin to see the effect on Lavoisier's studies of the stimulus imparted by Priestley's communication to him in October, 1774, of the results of the experiments carried out at Calne in August of that year; for at the rentrée publique of April 26, 1775, Lavoisier read to the Academy the most famous of all his memoirs, On the Nature of the Principle that combines with the Metals in Calcination and that increases their Weight. The memoir was first published in the May issue of Rozier's Journal<sup>3</sup>; and subsequently appeared, after being revised and

<sup>&</sup>lt;sup>1</sup> Bergman had suggested that "fixed air" be called "aerial acid" in 1774, but it is doubtful whether this was known in France at that time. Priestley did not know of Bergman's work; for although he had a copy of Bergman's paper he could not read Swedish (E. & O., II, pp. vi-vii). Lavoisier had, however, in October, 1774, written in his laboratory notebook, without stating reasons, that fixed air seemed to be an acide en vapeurs like air marin and air nitreux (Berthelot, p. 259).

<sup>2</sup> Observations, 1775, 5, 429.

read again on August 8, 1778, in the volume of the Mémoires for 1775 which was published in 1778.1 We shall deal with the earlier memoir here and the revised version later.

The earlier memoir carried on its first page a footnote to the title stating that the relevant experiments had been carried out over a year previously. Those on precipité per se, i.e. calx of mercury, had been tried in the burning glass in November, 1774, and afterwards carefully carried out at Montigny, coniointly with de Trudaine on February 28 and March 1 and 2, 1775, and repeated on March 31, 1775, in the presence of others. In the opening paragraph Lavoisier stated that he had intended to discuss before the Academy the question of whether there existed different kinds of air or merely modifications of atmospheric air; the occasion however demanded that he should limit himself to showing that the principle that united with the metals during their calcination, increased their weight and converted them into calces, was neither one of the constituent parts of air nor a particular acid diffused through the atmosphere, but that it was the air itself in its entirety, unaltered and undecomposed, so that if, after having been united in this combination, it was set free, it was evolved purer and more respirable, if such an expression was allowable, than the air of the atmosphere and more fit to support the inflammation and combustion of bodies. He had thus abandoned the theory that it was

<sup>&</sup>lt;sup>1</sup> Mém. Acad. R. Sci., 1775, p. 520. <sup>2</sup> In the original there is an obvious misprint here of est for et.

fixed air that combined with metals in calcination.

The description of the experiments on which this conclusion was based opened with the statement that most calces were reduced to the metallic state only in contact with carbonaceous matter, containing what was called phlogiston; that the charcoal when added in proper amount disappeared completely in the process, whence it followed that the air evolved in these reductions was not a simple substance, but in some way the result of a combination of the elastic fluid set free from the metal and of that evolved from the charcoal; and therefore, since the fluid appeared as fixed air, it was not iustifiable to conclude that it existed in that state in the calx before its combination with the charcoal. These circumstances therefore suggested the advisability of experimenting with calces that were reducible without the addition of other matter.

Lavoisier then proceeded to explain how he had reduced the calces of iron in glass receivers over mercury by means of the large burning glasses already mentioned, but that owing to admixture with common air the results were not satisfactory, that he therefore turned to the calx of mercury, precipitate per se, that he had carried out repeated experiments with this substance, of which he would describe only one. To satisfy himself that the substance was a true calx, he first reduced it with charcoal in a small retort of negligible capacity and obtained from it an air that by every test proved to be fixed air, since it was soluble in water, asphyxiated animals, extinguished lighted candles, precipitated

lime-water and combined with alkali. Lavoisier then placed 1 once of the calx in a retort, of capacity 2 cubic pouces, with a long narrow neck bent round so that its open end passed under a glass bell filled with and inverted over water. When the retort was heated, the air from the calx was slowly evolved and a total volume of 78 cubic pouces was collected. The weight of mercury recovered, mostly from the neck of the retort and from a vessel placed under the water below the open end, weighed 7 gros 18 grains, and thus, if the weight lost was entirely due to the air evolved, I cubic pouce of the air must have weighed a little less than & of a grain, which was not much different from the weight of the same volume of common air. The air obtained was insoluble in water, did not precipitate lime-water, neither combined with alkalis nor rendered them mild, could be used to calcine metals, was diminished like common air by "nitrous air" (nitric oxide), and had none of the properties of fixed air. It did not asphyxiate animals or extinguish lighted candles and other combustibles; the flame of a candle was enlarged by it in a very remarkable way, burning with greater light and brilliance than in common air. All these observations, said Lavoisier, proved that this air was not merely common air, but that it was more respirable, more "combustible," and consequently purer than the air that we live in; and thus, the principle that combined with metals in calcination and increased their weight was the purest part of the air that we

<sup>&</sup>lt;sup>1</sup> An inversion of meaning here.

live and breathe in, which passed from a state of expansibility into a state of solidity in this process, whereas, if charcoal was used in these reductions, this air was obtained in the state of fixed air, a result that must be due to the charcoal, and, if all calces could be reduced without charcoal, they would very probably give only common air. Common air purified by calcination is obviously meant: and "the purest part of the air" evidently means purified air, not a particular constituent of the air. All these observations on the air from calces, added Lavoisier, were applicable to the air obtained from nitre in the explosion of gunpowder; since the greatest part of that air was fixed air, and since charcoal was essential to the explosion, what occurred here was a conversion of common air into fixed air and therefore the air combined in nitre was the common air of the atmosphere deprived of its expansibility. Moreover, since common air became fixed air by combining with charcoal, it seemed natural to conclude that fixed air was a compound of common air and phlogiston,1 but Lavoisier asked that judgment should be suspended on this until he had carried out his intention of proving its doubtfulness.

Lavoisier's laboratory note-books for this period provide little information about these experiments. He was studying "inflammable air," nitric acid and nitre, in the early months of 1774. On October 28, he experimented at Montigny, repeating,

<sup>&</sup>lt;sup>1</sup> Lavoisier ascribed this view erroneously to Priestley, who promptly denied it (E. & O., II, p. 313). It was Rutherford's theory.

according to Berthelot, Priestley's experiments1; on November 22, he worked at Paris on the calcination of lead and on air in which metals had been calcined. On November 29, he wrote in his notebook that the nature of charcoal was unknown: "We know very well that on combustion it is evidently converted into fixed air, but we do not know whether it yields this fixed air itself, what is disengaged during the combustion and the proportion of the residue to the original weight of the charcoal. It would be very interesting to burn charcoal in a closed vessel; if the charcoal is composed of a sensible quantity of its weight of phlogiston, this must pass out and escape through the vessels and a decrease of weight ought to occur after the combustion. This experiment could be carried out in a hermetically sealed matrass."2 In March 1775, he repeated Priestley's experiments with the calx of mercury; he thought at first that the air evolved was fixed air, but it did not precipitate lime-water and, further, it enhanced the burning of a flame; and tests with "nitrous air" led him to conclude that it was common air with "a little of the nature of inflammable air." He reduced the calx of mercury with charcoal and obtained fixed air. The air from the calx of mercury did not asphyxiate a bird that was put into it.4 No date is given for this last observation, but its place indicates that it was not made before March 31,

<sup>&</sup>lt;sup>1</sup> Berthelot, p. 259, footnote 2. <sup>2</sup> Ibid., pp. 67-8 and 260-1. <sup>3</sup> Ibid., pp. 264-5. A further observation of the enhanced brilliance of a flame is noted on p. 267. <sup>4</sup> Ibid., p. 266.

1775, at a time when Priestley had already completely established the respirability of this "air" on much wider grounds. Thus Lavoisier's experimental study of the "air" from the calx of mercury was limited at this time, March, 1775, to the bare observations that it did not precipitate lime-water, enlarged the flame of a candle, did not asphyxiate a bird and reacted with nitrous air. However, after the appearance of the second volume of Priestley's Experiments and Observations, etc., late in 1775, in which Priestley pointed this out (op. cit., p. 320) and complained of the inaccurate accounts of his own work given by Lavoisier in the Opuscules (ibid., pp. 306-16), Lavoisier must have realized that Priestley had carried out much more extensive experiments, the experiments described in Chapter V above, and could profit from these detailed researches. That he did so is evident from his laboratory note-books, as will be shown presently.

Before passing on to consider the later versions of the two memoirs by Lavoisier that we have been discussing above, we may note at this point that the ideas on the combustion of the diamond expressed in the memoir of 1772, referred to in Chapter III above and not published until 1776, appear to owe something to the experimental advances of 1775.

The memoirs read by Lavoisier on November 12, 1774, and April 26, 1775, subsequently appeared in revised form in the *Mémoires* of the Academy for 1774 and 1775, both of which volumes were

<sup>&</sup>lt;sup>1</sup> See pp. 108-10 above.

published in 1778. The first memoir was returned to the Academy on May 10, 1777, and the second was read after revision on August 8, 1778. These later versions show how greatly Lavoisier's ideas had progressed in the interval between the original reading and the final publication.

In the first memoir, returned to the Academy two and a half years after its first reading, the introductory matter was much the same, but Lavoisier reported that the greater number of the sealed vessels were broken and that explosions made the experiments so dangerous that he had had to protect his face with a sheet-iron mask fitted with thick glass eye-pieces. Thus there were only two conclusive experiments with tin and "hardly one for lead." Yet in the original reading on November 12, 1774, he had spoken of a number of experiments with tin and lead and had at that time evidently anticipated the results of other experiments that were neither carried out then nor completed satisfactorily in the meantime. The same sensitive balance was used as in the experiments with water described in Chapter III. Each retort was heated before sealing its capillary end so as to expel a part of the air and reduce the risk of explosion. It was then re-weighed and heated on the furnace until calcination appeared to have ceased. After cooling, it was re-weighed and then carefully opened by "leading a crack," as we now say, round the middle of the bulb. The whole apparatus was now weighed again; and the tin, the calx, and the retort were then weighed separately. The results of the two experiments with tin, the second in a much larger retort than the first, may be set out as follows:

		I			II	
	Onces	Gros	Grains	Onces (	Gros	Grains
Weight of tin	8	0	0.00	8	0	0.00
Weight of retort	5	2	2.50	12	6	51.75
Weight of tin + open retort	13	2	2.50	20	6	51.75
Weight of tin + sealed retort,						
before heating	13	1	68.87	20	6	16⋅88
Weight of air expelled			5∙63			34.87
Weight of tin + sealed retort,						
after heating	13	I	68∙6o	20	6	18∙50
Difference in total						
weight, (negligible)			0.27			1.62
Weight of retort and contents	3		_			
after opening	13	2	5.63	20	6	61.81
Increase in total weight	;		3.13			10.06
Weight of tin remaining .	7	6	37:37	5	1	7.25
Weight of calx		1	37.75	2	7	2.75
Total weight of tin and calx	8	0	3.12	8	0	10.00
Increase in weight of tin	i		3.12			10.00
Weight of fragments of retor	t 5	2	2.50	12	6	51.62

In Experiment I the capacity of the retort corresponded to about 21 grains of air. Since 5\frac{2}{3} grains had been expelled before heating, the proportion of the air absorbed in the calcination was a little over 3 grains in 15\frac{1}{3} grains, or about \frac{1}{3}: in Experiment II a similar calculation gave a poor result, namely, \frac{1}{3} to \frac{1}{6}. As for lead, there was only one result and that was so unsatisfactory that Lavoisier preferred to defer publishing details of it.

A comparison with the details given in 1774

<sup>&</sup>lt;sup>1</sup> Lavoisier, quoting from the laboratory note-book that is now missing, gave the date of the first experiment as February 14, 1774.

reminds us that Lavoisier had then reported that the sealed retorts, weighed before and after heating "with the most scrupulous attention," showed not the slightest difference in weight and that the increase in weight found after they were opened was "always exactly equal" to the increase in weight of the metal. He was clearly here anticipating results of an accuracy that he failed to attain, indeed of an impossible accuracy, since he was unaware of the film of moisture that must have existed originally on the outside of the retorts, was removed in the heating and re-formed when the retorts were cooled; for, when he found a decrease of I grain on first weighing the sealed retort in Experiment II before it was properly cooled after the calcination, he put it aside until next day and then found it weighed 1.62 grains more than it weighed before the calcination, which difference he thought to be due to the difference in the volume of the retort at the two temperatures.

The experiments with tin, however, confirmed the conclusions announced in 1774. The weights of the sealed retorts were constant, but for negligible differences, before and after heating; and the increases in the total weights on opening the retorts corresponded very closely to the increases in the weight of the tin. While they lacked that remarkable accuracy announced in 1774, they were sufficiently good to prove the correctness of Lavoisier's theory, although, as he admitted, they left much to be desired. Experiment II led him now to make the modified statement that it was not certain that the

amount of metal calcined was exactly proportional to the capacity of the vessel. But the close correspondence between the increase in weight of the tin and the total gain in weight on admission of air to the retorts led him to conclude that the part of the air that combined with the metal in calcination had a density nearly equal to that of atmospheric air. Here Lavoisier began to propound his newer theory. Other considerations, he explained, led him to think that the part of the air that combined with the metals was a little heavier, while the air remaining after calcination was a little lighter, than common air; and that the density of common air was a mean between the densities of these two " airs." Further, although the experiments ought to be carried out in greater number and variety, they were so difficult and exacting that, as Lavoisier himself wrote, he had not yet had the courage to extend these researches. However, they had opened, he said, a new way. A part of the air was proved to be capable of combining with metals to form calces, while another part did not combine; and this led to the suspicion that air was not a simple substance, but was composed of very different substances. Other experiments, which he was carrying out on mercury and its calx, did, he thought, enable him to announce, without anticipating their results, that the whole of the air was not respirable, that it was the "salubrious part" that combined with metals in calcination and that the residue was a kind of mofette, incapable of supporting respiration or of maintaining combustion. The air of the atmosphere

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consisted of two very different "elastic fluids." But there was no reference to the experiments that in 1774 he said he had carried out on the air in which metals had been calcined, although the ideas now expressed on the composition of air were much more precise than those suggested at that time.

The problem was now at last approaching solution. Turning to the revised version of the memoir of 1775, read to the Academy, as we have already noted, on August 8, 1778, we find the complete solution set out as an inference from what has come to be regarded as Lavoisier's most classic experiment. The experimental details and quantities are exactly as reported in April, 1775, but on this occasion the apparatus was exhibited before the Academy. But the theoretical conclusions are altered throughout the revised memoir so that the "common air," purified and more respirable and "combustible," of 1775 now becomes "the most salubrious and the purest part of the air" and "eminently respirable air" in 1778; and, further, "fixed air" is a compound of charcoal and the "eminently respirable part of the air," because charcoal, when heated with calx of mercury, completely disappears and only mercury and fixed air remain. The problem was now solved: the substance that combined with metals in calcination and increased their weight was "eminently respirable air," "the most salubrious and the purest part of

<sup>&</sup>lt;sup>1</sup> He thought at this time that the *mofette* or mephitic part was very composite.

the air." Lavoisier's laboratory note-books show that between the first reading and the final submission of both these memoirs he had been repeating Priestley's experiments, the experiments described in the second volume of Priestley's Experiments & Observations etc. (1775) and discussed in Chapter V above. On February 13, 1776, he prepared l'air déphlogistiqué de M. Prisley from mercury precipitate per se and tested it with a candle and with "nitrous air." On April 7 he studied the air left after the calcination of mercury, the calcination being continued for more than 10 days, and found that it did not precipitate lime-water: his experiments were interrupted by a removal, but some time afterwards he mixed 5 parts of this air with I part of "dephlogisticated air" and found that a candle burned in the mixture nearly as in common air, calcined mercury in a retort fixed to a receiver over mercury (no details given), and noted that respiration involved partly an absorption and partly a vitiation of air.8 On October 13, he examined the air in which metals had been calcined and also Priestley's "dephlogisticated air"; he went on during this month to study the burning of candles in this air, with which he also made numerous experiments on respiration, not mere rapid tests as to whether or no it asphyxiated animals, but experiments to determine how long animals could live in it; and he found that the burning of candles and the respiration of animals in

<sup>1</sup> Sic. <sup>2</sup> Berthelot, p. 270. <sup>8</sup> Ibid., pp. 271-2. This is the only entry in the laboratory note-books referring to the most famous of all Lavoisier's experiments.

Priestley's air led to the production of "fixed air."1 In April, 1777, he studied combustion in common air and "dephlogisticated air" and wrote down in his note-book that a burning candle converted only "the pure part" of common air into "fixed air." Priestley had given Lavoisier the clue he needed: and Lavoisier had now shown that air consisted of two "elastic fluids," one respirable and the other mephitic. Lavoisier's debt to Priestley is very evident here; for it was undoubtedly through these repetitions of Priestley's work that he was led to revise the memoirs of 1774-5 with the consequences that we have seen above. Grimaux's assertion,3 founded on the revised version of the memoir of 1775, that Lavoisier took up Priestley's work in November, 1774, and "immediately surpassed his illustrious rival" is a misreading of the facts.

Thus Lavoisier had formulated his new theory of combustion and calcination and had demonstrated the composition of the air by May 10, 1777, when the memoir of November 12, 1774, was returned to the Academy in revised form; and he had set this out in greater detail and explicitness on August 8, 1778, when reading to the Academy the revised memoir of April 26, 1775. It was at these later dates, namely, the years 1777 and 1778, and not in the years 1774 and 1775, that the modern theory of combustion and calcination was established and the composition of the air demonstrated by Lavoisier. The earlier dates so frequently given are

<sup>&</sup>lt;sup>1</sup> Ibid., pp. 290-1. <sup>2</sup> Ibid., pp. 291-2. <sup>3</sup> Grimaux, pp. 107-8.

historically incorrect and their repeated appearance is due to the mere accident that the *Mémoires* of the Academy were published in arrears, the volume for a particular year appearing in some cases as many as four years after the presentation of the memoirs contained in it, so that contributors were enabled to amend and revise their work in the light of the greater knowledge accumulated in the meantime. These circumstances have allowed considerable error to creep into the history of science, through discoveries and theories being thus antedated, or at least antedated by those who are unable to afford the time and trouble necessary for the examination and comparison of numerous lengthy documents.<sup>1</sup>

We shall now turn to consider in the next chapter how Lavoisier gradually developed and extended the theory formulated in 1777-8.

<sup>&</sup>lt;sup>1</sup> For a further example, Lavoisier's attempt at a quantitative estimation of the "fixed air" produced from a given weight of charcoal appears in the *Mémoires* for 1781 (p. 448). As this volume was not published until 1784, Lavoisier was able to correct his figures for the amount of water formed from the hydrogen in the charcoal by using data for the composition of water—a composition unsuspected in 1781—determined much later.

#### CHAPTER VII

# DEVELOPMENT OF THE NEW THEORY, 1778-86

Following his formulation of a new theory of combustion and calcination, Lavoisier then proceeded to develop his ideas in a long series of memoirs. On April 20, 1776, he submitted to the Academy a memoir on the existence of air in nitric acid. This was re-submitted in December, 1777, and appeared in the *Mémoires* for 1776, published in 1770.1 In this he showed, despite experimental difficulties that prevented satisfactory quantitative results, that nitric acid contained "the purest part of the air," which he said entered into the composition of all acids and was essential to their acidity. He returned also to the combustion of phosphorus in a memoir, submitted to the Academy on March 21 and read on April 16, 1777, which appeared in the Mémoires for 1777, published in 1780.2 In this he explained that he could now present more satisfactory results and explanations than those given in the Opuscules. He showed that phosphorus ignited with a burning lens in air over mercury combined with the "eminently respirable" part of the air to give phosphoric acid and increased in

<sup>1</sup> Mem. Acad. R. Sci., 1776, p. 671.

weight in proportion to the amount of this air absorbed; that the residual air or mofette on the addition of "eminently respirable air" from lead or mercury calx recovered the properties of common air and could be used again for the combustion of more phosphorus; and that common air consisted of about one-quarter of "eminently respirable air" and three-quarters of "a mephitic air" or mofette of slightly lower density than the air of the atmosphere. And he added that the same was true of sulphur, which combined in the same way with "eminently respirable air" to produce vitriolic acid, although experimental difficulties prevented him from obtaining satisfactory quantitative results.

At the rentrée publique of May 3, 1777, Lavoisier read a memoir, subsequently included in the Mémoires for 1777,1 on the respiration of animals. He described how he had calcined 4 onces of mercury in 50 cubic pouces of common air for twelve days, red particles of calx being slowly formed on the surface of the mercury during this time and the air being finally reduced in volume between 8 and 9 cubic pouces. Calx weighing 45 grains was obtained. The residual air was mephitic: it extinguished candles, asphyxiated animals and was undiminished by "nitrous air" (nitric oxide). The calx obtained was heated in a small retort and gave about 8 to 9 cubic pouces of air, collected over water. This, as was known, was the respirable part of the air. Lavoisier then added this product to the residue from the first experiment and obtained an

air that, on testing with candles, animals and "nitrous air," proved to be common air. Thence he proceeded to show by means of a sparrow that it was the "eminently respirable air" that was used up in respiration, in which the "mephitic part" of common air was inactive, and that the "eminently respirable air" was converted into "aerial acid," i.e. "fixed air." The experiments on respiration were few, and it seems that Lavoisier was here concerned to apply his theory to observations that had already been made by others. Much the same is true of his memoir on the combustion of candles. included in the Mémoires for 1777,1 in which he showed that burning candles converted the respirable part of the air into "aerial acid," which substance he showed was produced likewise when candles were burnt in "eminently respirable air" obtained from the calx of mercury. In this place, too, he clearly distinguished four "airs," namely, common air, "eminently respirable air," the atmospheric mofette and "fixed air" and explained their relation to one another. But, throughout this and the previous memoir, Lavoisier was diffidently beginning his attack on the phlogiston theory by showing that his own theory provided more satisfactory explanations of these phenomena.

Again in the same year Lavoisier presented another memoir, in which he showed that vitriolic acid (sulphur trioxide) was composed of "eminently respirable air" combined with volatile vitriolic acid (sulphur dioxide), but experimental

<sup>&</sup>lt;sup>1</sup> *Ibid.*, p. 195.

difficulties prevented him from obtaining satisfactory quantitative results. And he applied his theory to explain the burning of Homberg's pyrophorus<sup>1</sup> and the conversion of pyrites (sulphides) exposed to the air into vitriols (sulphates) by combination with "eminently respirable air." He also put forward the suggestion that all "airs" or "elastic fluids" were compounds of evaporable liquids of volatile solids with the matter of fire or material heat.3 Further, in a memoir4 on combustion in general, he criticized the phlogiston theory and showed that the hypothesis that he had been advancing involved less contradiction and was more in agreement with the facts. In another memoir, submitted on September 5, 1777, and read on November 23, 1779,5 Lavoisier contended that all acids were formed by the combination of non-metallic substances with "eminently respirable air," which he accordingly now re-named principe acidifiant or principe oxygine (oxus, acid: gennao, I beget), the "acidifying principle" or the "oxygen principle." This, we may note, marks the first appearance of the term oxygen in chemistry. Moreover, by principle Lavoisier understood, as is clear from the introductory paragraph of this memoir, a principle of bodies, that is, an element, a substance that chemical analysis could not resolve into any simpler substance. "Eminently respirable air" was therefore the acidifying principle or element. He referred to it also as "vital air" in a

Ibid., p. 363.
 Ibid., p. 398.
 Ibid., p. 420.
 Mém. Acad. R. Sci., 1778, p. 535. Published in 1781.

later memoir, read on November 18, 1780, on the compounds of phosphoric acid.1

Thus Lavoisier, from 1778 onwards, was persistently attempting to undermine the older theory by showing that his hypothesis, as he was often careful to call it, gave explanations of chemical phenomena that were more in accordance with the facts and involved less contradiction.2 The revolution in chemistry that he had foreseen was slowly being achieved. And there seems little doubt that the experiments described in Scheele's Chemische Abhandlungen von der Luft und dem Feuer, which was published in 1777, gave him a greater confidence in his ideas; for they were explained much more satisfactorily by his theory than by Scheele's phlogistic notions. We do not know when Lavoisier first read this book. He reported<sup>3</sup> to the Academy on August 8, 1781, on Baron Dietrich's French version of Scheele's work, and he discussed it critically in the light of his own ideas in 1781.1 He had sent Scheele in 1774 a book which was almost certainly the Opuscules,4 and Scheele had written to thank him and suggest that Lavoisier should heat

<sup>&</sup>lt;sup>1</sup> Mém. Acad. R. Sci., 1780, p. 343. Published in 1784.

<sup>2</sup> The cold reception that Lavoisier's theory met with at this time is well instanced in the following extract from a letter written by Macquer to Guyton de Morveau on January 4, 1778: "M. Lavoisier m'effrayoit depuis longtemps par une grande découverte qu'il réservoit in petto et qui n'alloit pas moins qu'à renverser de fond en comble toute la théorie du phlogistique ou feu combiné; son air de confiance me faisoit mourir de peur. Où en aurions-nous été avec notre vieille chimie, s'il avoit fallier attait d'éffecte à Pour moi à vous que que qu'en qu'en propis la vous qu'en qu'en qu'en qu'en propis le vous qu'en qu'en qu'en qu'en propis le vous qu'en qu'en qu'en propis le vous qu'en qu'en propis le vous qu'en qu'en qu'en propis le vous qu'en qu'en qu'en propis le vous qu'en qu'en propis le vous qu'en qu'en l'auvois fait avoit fait qu'en l'auvois en l'auvois fait avoit fait qu'en l'auvois fait avoit fait qu'en l'auvois fait avoit fait qu'en l'auvois en l'auvois fait avoit fait qu'en l'auvois fait qu'en l rebâtir un édifice tout différent? Pour moi, je vous avoue que j'aurois abandonné la partie. M. Lavoisier vient de mettre sa découverte au jour, je vous avoue que, depuis ce temps, j'ai un grand poids de moins sur l'estomac'' (Grimaux, p. 122).

3 Œures, IV, p. 377.

4 Mém. Acad. R. Sci., 1781, p. 396.

<sup>&</sup>lt;sup>5</sup> Cf. Grimaux, p. 103.

with a burning lens the precipitate obtained by adding alkali to the solution of silver in nitric acid, collect the air evolved over lime-water and carry out tests to see whether candles could burn and animals live in it.1 Possibly Scheele may have returned the courtesy by sending Lavoisier a copy of his newly published treatise in 1777. We have no information about this, but the opinions that Lavoisier expressed on Scheele's book in 1781 speak eloquently of his appreciation, not only of Scheele's magnificent experimental skill, but also of the support that Scheele's experiments afforded for the new theory. From Scheele and from Priestley, Lavoisier was prepared to reap the rich harvests of facts produced by those princes among experimenters and reap them for the benefit of his theory. On some occasions he omitted to give his fellowlabourers their due, an action for which he has been much criticized; yet on others he recognized his indebtedness to them. But he insistently and properly claimed that the new theory was his own, and he was certainly entitled to offer better explanations of facts that other discoverers could not explain. Priestley and Scheele were far greater experimenters than Lavoisier: Lavoisier as a thinker outran them both.

It was, however, not until 1783 that Lavoisier launched his attack on the phlogiston theory in a memoir entitled Réflexions sur le Phlogistiques and

<sup>&</sup>lt;sup>1</sup> See a letter to Lavoisier from Scheele, translated by Dobbin, Collected Papers of C. W. Scheele, London, 1931, pp. 350-1.

<sup>2</sup> Mém. Acad. R. Sci., 1783, p. 505. Published in 1786. Berthelot (p. 136, footnote) wrongly suggests that this memoir was read in 1777.

described by him as a sequel to the theory of combustion and calcination published in 1777, his quotation of this date clearly indicating that he made no claim to have propounded this theory at the earlier date that is still so frequently given. This memoir, revealing Lavoisier's powers at their greatest, is one of the most notable documents in the history of chemistry and of far greater historical significance than Boyle's Sceptical Chymist; for the revolution that Lavoisier had aimed at in 1773 was now, ten years later, approaching its climax and he was here at last able to engage the phlogiston theory in the open, to rout its defenders with a destructive fire of chemical facts and to seize for his own theory the position that it now properly deserved.

The Réflexions opens with a statement that in earlier memoirs Lavoisier had explained the facts of combustion and calcination in terms of the principe oxygine and that, this principle once admitted, the chief difficulties of chemistry seemed to vanish and chemical phenomena were explicable in a remarkably simple way. Therefore, if the facts of chemistry were satisfactorily explicable without the help of phlogiston, on that ground only it was very probable that phlogiston did not exist. That entities ought not to be multiplied unnecessarily was, moreover, a principle of good logic. Lavoisier added that he might be content with this negative proof and with showing that chemical phenomena were explained better without phlogiston than with it, but it was now time to express himself more distinctly and explicitly about a theory that he

regarded as an error fatal to chemistry and a considerable hindrance to its progress.

The phlogiston theory, continued Lavoisier, was formulated at a time when the main phenomena of combustion were unknown; and nothing was more natural than to suppose that combustible bodies were such because they contained an inflammable principle. But since the founders of the theory, besides recognizing calcination as a true combustion, had supposed that phlogiston was an earthy principle, or at least a principle that contained the earthy element, and since it therefore appeared that phlogiston possessed weight, the theory could not account for the fact established long ago by Boyle that metals increased in weight on calcination nor for the subsequent extension of this observation to include such combustibles as sulphur, phosphorus, etc. Since, in terms of the theory, these bodies lost phlogiston, they should decrease in weight. Baumé had contended that the metals on calcination, while losing phlogiston, which he took to consist of the matter of fire combined with an earthy principle, took up in its place free fire, or at least free fire combined with less earth than what it had been combined with in phlogiston, and ascribed their gain in weight to addition of this free fire. This suggested that the element of fire was very heavy; for, in calcination, it not only increased the weights of metals considerably, for example, iron by more than a third, but also made up for the loss of their phlogiston, a substance that also had weight. Experiments with the most sensitive instruments showed however that the matter of fire had no appreciable weight and that metals, far from absorbing free fire in calcination, on the contrary evolved it in measurable amounts. And, further, a vapourized fluid, such as water at its temperature of condensation, which was known to contain a great amount of the matter of heat almost in a free state, ought, if Baumé was correct, to be able to reverse the effects of calcination and combustion and restore to their original state bodies that had been subjected to these processes, whereas nothing of this kind took place. Therefore, it was not to combination with free or almost free fire that such reconversions were due. Finally, according to Baumé's view, when metals were calcined in sealed vessels, an increase in weight should occur. It was however established that if these vessels were weighed both before and after calcination, without their seals being broken, no difference of weight was detectable even with sensitive balances.

Lavoisier then referred to his note of November 1772,<sup>1</sup> and to the experiments of 1773<sup>2</sup> in which he showed, firstly, that minium on reduction with charcoal gave out a large quantity of "elastic fluid," and, secondly, that this fluid was in every way similar to that obtained from chalk, calcareous earths, alkalis, fermentation, etc. In 1773 also, he pointed out, in the calcination of metals by the

<sup>&</sup>lt;sup>1</sup> See pp. 117-18 above. The note of 1772 refers to litharge, not minium, and to air which, as we have seen above, Lavoisier long supposed it to be, not "fixed air." He wrote here as if all this was known to him in November, 1772.

See p. 135 above.

burning lens in limited quantities of air, he had observed that in the calcination the volume of air decreased with the extent of calcination and that the increase of weight of the metal was exactly<sup>1</sup> equal to the amount of air that had disappeared. It was impossible not to conclude from these facts that the increase of weight was due to the fixation of a portion of the air<sup>2</sup> which combined with the metal proportionately to its calcination. These results, he said, were set out in his Opuscules. Yet, although his experiments proved his theory, the usual doubt began to be cast on the facts and, he added: "Those, who seek to persuade the public that everything that is new is not true or that everything that is true is not new, succeeded in finding the first germ of this discovery in a much older author. Without examining here the authenticity of that work, a new edition of which was then hurriedly produced, I observed with some satisfaction that the impartial public had concluded that a vague and random assertion, based on no experiments and unknown to all savants, did not prevent me from being recognized as the discoverer of the cause of the increase in weight of metallic calces."3

<sup>2</sup> Originally air itself: see pp. 117-18 above. The clear idea that it was only a part of the air dates from 1777-8 (see pp. 208-11 above).

<sup>1</sup> Here he wrote fort exactement.

<sup>&</sup>lt;sup>3</sup> Lavoisier is referring to Rey and to Gobet's edition of Rey's Essays, published at Paris in 1777. His criticisms of the excessive lauding of Rey's work are justified: Rey's ideas were—what Lavoisier said they were. Later Lavoisier spoke well of Rey; and it seems that at one time he suspected the authenticity of the Essays and thought that the work had been published at a much later date than that indicated on the title page (Eurres, II, p. 100). At one time there was also some reference to Mayow's having anticipated Lavoisier. Lavoisier wrote to Magellan in London to ask him to obtain a copy of Mayow's Tractatus Quinque, etc. (Oxford, 1674), but

To clinch his claim to the discovery, Lavoisier added: "Not only did I show at that time that increase of weight was one of the factors attending all metallic calcinations, but I also proved that the same law held in combustion; that sulphur, phosphorus, all combustibles in general, increased in weight on burning and that this increase was due to the combination, the fixation of air."

These facts contradicted the phlogiston theory and, accordingly, Macquer had attempted to reconcile the new facts with the old doctrine. In the second edition of his Dictionnaire de Chimie (Paris, 1778), Macquer had set out his compromise, namely, that phlogiston was the pure matter of light.<sup>2</sup> This, said Lavoisier, was no defence of the phlogiston theory: it was an outrage on it. It was keeping the name and rejecting the substance: it was Macquer's theory, not the phlogiston theory. Macquer's "phlogiston" was phlogiston without its earthy element. It combined with solids, increased their weight, made them opaque and gave them their characteristic colours and smells: it did not combine with air or with water. This did not. as Lavoisier pointed out, account for the gain in

Magellan could not get one (Grimaux, p. 107, footnote 1). However, when Beddoes published his Chemical Experiments and Opinions, etc. (Oxford, 1790), an absurd laudation of Mayow (see Patterson, Isis, 1931, 15, 47 and 504), Delamétherie apparently had no difficulty in finding a copy of the Tractatus Quinque in one of the libraries of Paris—he says merely à la bibliothèque publique—and published an account of it in Rozier's Journal (Observations, 1790, 37, 154), together with a reproduction of the well-known six figures of Plate V.

<sup>&</sup>lt;sup>1</sup> Lavoisier was hardly entitled to claim all this: he had, as we have seen, shown it to be true in some cases only and this at later dates.

<sup>&</sup>lt;sup>2</sup> For example, in the article *Phlogistique* (Dictionnaire, etc., Paris, 1778, II, p. 198f.).

weight of metals on calcination. Indeed, on this theory, they ought to lose weight. But Macquer had carried his compromise further by contending that the metals on calcination and combustibles on burning not only lost their phlogiston or matter of light, but also concurrently combined with the purest part of the air and thereby acquired a proportionate increase of weight. Thus, calcination and combustion involved a combination with air and a simultaneous separation of phlogiston, that is, the matter of light. The direct reducibility of the calces of gold, silver and mercury without the addition of charcoal was also explained, since Macquer's "phlogiston" could penetrate the pores of the vessels in which these substances were heated. While these phenomena could be explained on Macquer's system, there were others that could not. For instance, if phlogiston was the pure matter of light, all calces, not only those of gold, silver and mercury, ought to be convertible to metals with the burning lens just as with charcoal. Yet they were merely vitrified by the burning lens; and, therefore, the matter that existed in charcoal was not the same as that constituting the solar rays and phlogiston was not the pure matter of light. Macquer had tried to avoid this objection by stating that calces could not be converted into metals by the burning lens because the air that was present led to their being recalcined. Lavoisier disposed of this by pointing out that replacement of this air by atmospheric mofette made no difference to the result: the matter of light could not act here in the same way as charcoal, and

therefore phlogiston was not the matter of light. Chemists in general had, indeed, not accepted Macquer's views completely. Phlogiston had been preserved in name only; and contradictory properties had been unwittingly united in one vague and ill-defined term. For example, it was known that when pure charcoal was burned in "vital air" in a sealed vessel, the total weight remained constant, but the density of the air inside was increased and its increase of weight was equal to the weight of charcoal used; and most chemists, adopting the phlogiston theory, would be forced to recognize (1) that matter of heat and light had been released and had escaped through the vessel, whence, since the weight remained constant, it followed that the matter of heat and light did not possess any sensible weight, and (2) that "fixed air" was formed, and, since the weight of this was equal to the combined weights of "vital air" and charcoal, it followed independently of any system that charcoal contained a matter which had weight and which could not escape through glass vessels and which, therefore, was not the matter of heat and light. Thus the followers of Stahl gave the name "phlogiston" to two very different substances, to matter that lacked weight and passed through the pores of vessels and to matter that had weight and combined with "vital air" to form "fixed air." Here, therefore, were two distinct substances that the followers of Stahl confounded; one had no weight, the other had; one was the matter of light, the other was not; and it was by assuming these properties

sometimes for one and sometimes for the other of these substances that they had succeeded in explaining everything.

In the same way those chemists who followed Stahl had unconsciously admitted two kinds of phlogiston in the reduction of calces. The calces of gold, silver and mercury were converted into their metals on the application of heat only and left on one hand, the metal, and on the other, the "vital air" that had been combined with it, the total weight of metal and air being equal to the weight of the calx. To explain this, they had to say, with Macquer, that the matter of light had traversed the pores of the vessels and combined with the metal: and, since the weight of air and metal did not exceed the weight of the calx, it followed that their phlogiston had no weight. But with other calces the addition of some carbonaceous substance was necessary for their reduction; and here the products were "fixed air" and metal, but the total weight was increased by the weight of the charcoal used. Here, therefore, was a phlogiston that had weight; and the followers of Stahl were compelled to give the name of phlogiston to two different substances, namely, to the matter of light or the element of fire which had no weight, and to a carbonaceous matter which had weight.

Moreover, the reduction of calces provided another argument against these chemists. The substance that combined with a metal to form a calx was undoubtedly "vital air," the principe oxygine. But in reductions with charcoal, this substance was

released in the form of "fixed air." Since the weight of the charcoal and the "vital air" used corresponded to the weight of the "fixed air," had the phlogiston of the charcoal therefore combined with the "vital air" to form "fixed air"? But, if the whole weight of the charcoal had entered the "fixed air," it had not combined with the metal or at least what had combined with the metal did not possess weight. Thus again they had to admit two different substances with the same name. Beyond these difficulties common to the various forms of the phlogiston theory, Macquer's theory presented its own special difficulty: for, if phlogiston was the pure matter of heat and light, metals must contain more of the matter of heat than calces contained, whereas measurements of specific heat proved the contrary. Thus it followed that either phlogiston was not the matter of heat and light, or else that the metals contained less phlogiston than their calces. One or the other of these consequences must be admitted: and either destroyed Macquer's system and, indeed, the doctrine of phlogiston in general. Here Lavoisier delivered his most decisive blow: "The partisans of Stahl's doctrine," he wrote, "are continually in similar difficulties. If they are asked what happens when mercury is calcined in vital air, the English philosophers reply that the phlogiston, in proportion as it is released from the metal, combines with the air in which the experiment is performed and changes it into fixed air or phlogisticated air. But this is again absolutely contrary to the facts. When the experiment is

performed in perfectly pure vital air, the air can be absorbed to the last bubble and, if the experiment is interrupted before the absorption is completed, the part of the vital air that remains is entirely unaltered; it contains exactly the same amount of mephitic air as that originally contained in the total amount of air taken. The phlogiston in this experiment has therefore not combined with the air, as the English philosophers maintain, and then it must be admitted, with M. Macquer, that it has escaped in the form of free fire, of the matter of light, through the pores of the vessels. But, if phlogiston can thus pass freely through the pores of vessels, if in the calcination of metals in vital air it has the property of penetrating glass, if it has this same property in the revivification of the calces of gold, silver and mercury, why does it not have it with regard to the other metallic calces? Thus the partisans of Stahl's doctrine, after having been forced to say that phlogiston sometimes has weight and sometimes has not, are further compelled to admit that, even in its free state, it sometimes penetrates through the pores of the closest vessels and sometimes it does not, properties that are incompatible in the same entity and which prove more and more that the same name has been given to two very different things."

Again, phlogiston was alleged to be the principle of colours. Yet the calces of lead and iron deepened in colour on progressive calcination, that is, in what was considered to be a process involving an increasing loss of phlogiston: and the calx of mercury was

red, while, as for white calces, they surely contained all the colours. Again, paper and linen became black when burnt, that is, when they were alleged to have lost their phlogiston. If phlogiston was the principle of colour, burnt paper must contain more phlogiston than white, that is unburnt, paper did. It was the same with taste and causticity, of both of which phlogiston was said to be the principle. If this were true, the metals should be highly caustic and the calces tasteless, whereas the reverse was the case.

"All these reflections," added Lavoisier, "confirm what I have advanced, what I intended to prove, what I am going to repeat again, that chemists have made phlogiston a vague principle which is not strictly defined and which consequently fits all the explanations required of it; sometimes the principle has weight, sometimes it has not; sometimes it is free fire, sometimes it is fire combined with earth; sometimes it passes through the pores of vessels, sometimes these are impenetrable to it; it explains at once causticity and non-causticity, transparency and opacity, colour and the absence of colour. It is a veritable Proteus that changes its form every instant."

"It is time," he continued, "to lead chemistry back to a stricter way of thinking, to strip the facts, with which this science is daily enriched, of the additions of rationality and prejudice, to distinguish what is fact and observation from what is system and hypothesis, and, in short, to mark out, as it were, the limit that chemical knowledge has reached

DEVELOPMENT OF THE NEW THEORY 231 so that those who come after us may set out from

that point and confidently go forward to the advancement of the science."

Before giving any further account of his own theory, which had already been expounded on numerous occasions from 1777 onwards, Lavoisier then proceeded to describe his views on the nature of heat. The phenomena of heat suggested the existence of a fluid matter of heat, a fluid that was admittedly hypothetical, but this, he contended, was the only hypothesis involved in his theory and it was one recognized by those who accepted the phlogiston theory. Experiments showed that vital air contained more of this matter of heat than did any other known elastic fluid. In combustion this heat was set free. For example, in the combustion of phosphorus the vital air passed from an aeriform state to a concrete state and thus abandoned the matter of heat with which it had been combined and which had given it its aeriform state. This heat was then set free. The general phenomena of combustion might therefore be set out as follows:

1. There was true combustion, evolution of flame and light, only in so far as the combustible body was surrounded by and in contact with vital air. Combustion could not take place in any other kind of air or in a vacuum: burning bodies plunged into either of these were extinguished just as if they had been plunged into water.

<sup>&</sup>lt;sup>1</sup> The methods then in use for measuring the specific heats of gases were very inaccurate. Cf. McKie and Heathcote, The Discovery of Specific and Latent Heats, London, 1935, p. 43, footnote 4.

- 2. In all combustion there was absorption of the air in which the combustion occurred; and, if this air was vital air, it could be completely absorbed.
- 3. In every combustion there was an increase in weight of the body burnt and this increase was exactly equal to the weight of the air that had been absorbed.
- 4. In every combustion there was an evolution of heat and light.

"My only object in this memoir," concluded Lavoisier, "is to extend the theory of combustion that I announced in 1777; to show that Stahl's phlogiston is imaginary and its existence in the metals, sulphur, phosphorus and all combustible bodies a baseless supposition, and that all the phenomena of combustion and calcination are explained in a much simpler and much easier way without phlogiston than with it. I do not expect that my ideas will be adopted at once; the human mind inclines to one way of thinking and those who have looked at nature from a certain point of view during a part of their lives adopt new ideas only with difficulty; it is for time, therefore, to confirm or reject the opinions that I have advanced. Meanwhile I see with much satisfaction that young men, who are beginning to study the science without prejudice, and geometers and physicists, who bring fresh minds to bear on chemical facts, no longer believe in phlogiston in the sense that Stahl gave to it and consider the whole of this doctrine as a

scaffolding that is more a hindrance than a use for extending the fabric of chemical science."

The Réflexions appeared in the volume of the Mémoires for 1783, which was published in 1786. It is curious that Lavoisier allowed this memoir to appear in print with only the slightest reference to the discovery that water was a compound of "vital air" and "inflammable air," because the relevant facts were already known to him in June, 1783, even before Cavendish announced his results publicly in January, 1784, that is, nearly three years before the publication of the Réflexions. It may be that in 1786 Lavoisier found such acceptance for his theory that the revision of memoirs was no longer a matter of importance.2 The discovery of the composition of water however involved Lavoisier in some curious incidents that we shall need to take notice of here.

In 1781 in experiments on the explosion of "inflammable air" with common air in closed glass vessels by means of the electric spark, Priestley noted that the inside of the glass vessels "became dewy" after the explosion. His friend Warltire had long hoped to be able to apply experiments on the

written the Discours in 1774.

Laplace had given Lavoisier his support for some time and Berthollet had accepted the theory publicly at the Academy on April 6, 1785. Guyton de Morveau followed in 1786 and Fourcroy in 1787, Monge, Meusnier and Chaptal later.

<sup>&</sup>lt;sup>1</sup> Almost ten years earlier, in March, 1774, a lengthy and anonymous attack on the phlogiston theory under the title Discours sur le Phlogistique had appeared in Rozier's Journal (Observations, 1774, 3, 183–98). Berthelot (p. 54) thought that this ought to be ascribed to Lavoisier. Comparison of the text of this criticism with that of the Reflexions and a realization of the differences in the views of chemists in general in 1774 and in 1783 amply confirm Berthelot's suggestion. No other hand than Lavoisier's could have written the Discours in 1774.

<sup>\*</sup> Experiments and Observations, etc., Birmingham, 1781, II, pp, 395-8.

combustion of these two airs to decide whether or no heat had weight by weighing the vessels before and after the explosion, but had thought it too dangerous. Priestley advised him to try it in small amounts in copper vessels. This proved quite safe. Meanwhile, the intrepid experimenter Priestley had been exploding the mixture in glass vessels and had noticed the appearance of dew. Warltire confirmed this observation and noted also that a "smoke" had always escaped from the copper vessels when air was blown through them after the explosions. This marked the beginning of the researches that led to the discovery of the composition of water. Priestley, writing of the particular experiment on which Warltire had commented, described this as "a mere random experiment, made to entertain a few philosophical friends, who had formed themselves into a private society, of which they had done me the honour to make me a member." This society was, in all probability, the famous Lunar Society of Birmingham, which was so-called because its monthly meetings were held at the full moon to reduce the inconvenience of returning home in the dark and which numbered among its members Watt, Boulton, Wedgwood, Erasmus Darwin, Keir, Withering, Samuel Galton, Richard Lovell Edgeworth and Thomas Day, the author of Sandford and Merton: and Birmingham has thus some claim to be considered the scene of the first observation that led to the discovery of the composition of water. As for Priestley's unfortunate phrase about the "random experiment," it has in

the eyes of most historical writers branded him as a scientific leper ever since, as if he had never carried out this experiment before and as if its performance on this occasion was the outcome of some sportive fancy. Yet, all that his statement amounts to is this, that, among the many experiments that might have been performed before the society, this one was chosen at random and that when Warltire first observed the formation of moisture on this occasion he remarked that it supported a theory that he had long held, namely, that common air deposited its moisture when it was phlogisticated. That Priestley had observed the effect before this is clear, since Warltire reported to him: "I have fired air in glass vessels since I saw you venture to do it, and have observed, as you did, that though the glass was clean and dry before, yet after firing the air, it became dewy."2

However, Warltire's experiment always indicated a variable loss of weight<sup>3</sup> and Priestley communicated the information to Cavendish, who took up the problem of the alleged loss of weight and decided to investigate also the nature of the "dew." In repetitions of these experiments Cavendish found that there was generally no loss of weight, and he discovered "that when inflammable air and common air are exploded in a proper

<sup>&</sup>lt;sup>1</sup> He had already repeatedly exploded these gases by the electric spark either over water or over mercury or in Nairne's pistol (ibid., pp. 124-7); and he had previously exploded mixtures of "inflammable air" and "dephlogisticated air" with the flame of a candle (E. & O., II, pp. 98-9).

<sup>2</sup> Experiments and Observations, etc., 1781, II, pp. 396-7.

<sup>3</sup> It was the loss of weight that aroused their interest: this bore on the important question of whether heat had weight.

<sup>4</sup> Phil. Trans., 1784, 74, 119.

proportion, almost all the inflammable air, and near one-fifth of the common air, lose their elasticity, and are condensed into dew . . . that this dew is plain water, and consequently that almost all the inflammable air, and about one-fifth of the common air, are turned into pure water." These results were confirmed in further experiments on the explosion of "inflammable air" with "dephlogisticated air." This classic work is so well known that it is not necessary to describe its details in this place. We shall take note, however, that Cavendish's explanation of the result was not that the two "airs" combined to produce water, but was set out in terms of the phlogiston theory to which he adhered. It may be stated briefly thus: "dephlogisticated air" is water deprived of its phlogiston and "inflammable air" is phlogisticated water, and by the phlogisticating action of the electric spark the mixture is condensed into water. Thus the water came from both "airs," in each of which it was originally contained.

Cavendish's memoir was read to the Royal Society on January 15, 1784. The experiments, it was stated, had been performed in the summer of 1781. This fact established, we shall now turn to consider Lavoisier's work on the problem and the consequences of Cavendish's discovery on Lavoisier's new chemical theory. It is well known that the events we are now approaching gave rise to much bitter controversy, to deal critically with the literature relating to which would demand a whole book in itself, but here it will suffice to set down the facts

as they appear in the original documents up to the time of Lavoisier's first publication of his own

studies on the composition of water.

In March 1774, Lavoisier had written in his laboratory note-book: "I was convinced that the burning of inflammable air was nothing but a fixation of a part of the atmospheric air, a decomposition of air, and that the liberation of the matter of fire could very well arise from the inflammable principle which was separated from the fixed part of the air. In this case, in every burning of the air there ought to be an increase of weight. To satisfy myself, I put some suitably diluted vitriolic acid in a matrass: I weighed the matrass in this state and I added to it a known quantity of iron-filings. The dissolution took place with commotion and heat. I brought a candle near and an explosion occurred. But throughout the dissolution, there was a diminution of weight: a feeble flame continued to burn above the neck of the matrass until the dissolution was complete and this did not prevent a continual and progressive diminution amounting to a total of 30 grains." This curious but interesting attempt to study the combustion of "inflammable air" shows Lavoisier still thinking in terms of phlogiston, but intent on applying the principle that combustion involves increase of weight in the combustible. There is however no indication that he had even the most remote suspicion as to what happened in this combustion.

On April 8, 1775, Lavoisier exploded with a Berthelot, pp. 256-7.

candle a mixture of "inflammable air" and common air in vessels over water and noted: "It appeared that the inflammable air burns only in proportion to the common air that is introduced into it. By introducing the candle several times, it burnt completely; then, what remained?" The experiment, Berthelot<sup>2</sup> pointed out, was carried out over water and the product, since it also was water, passed unrecognized. But Lavoisier's query as to what remained possibly refers to the residual gas, nitrogen, left after the combustion was completed. There are however no further entries in the notebook at that time to enable us to decide to what he was referring. In 1777 in work carried out with Bucquet, Lavoisier burnt "inflammable air" over lime-water to test Bucquet's opinion that "fixed air" was produced, but no precipitate was formed: and in the winter of 1781-2 with Gengembre. Lavoisier burnt a jet of "vital air" in an atmosphere of "inflammable air," but failed to detect any sign of the acidity that he expected from the combustion of "vital air," the "acidifying principle." Meanwhile, Macquer had noted that a colourless liquid, which appeared to be pure water, was formed on a piece of porcelain held in the flame of "inflammable air."4

<sup>&</sup>lt;sup>1</sup> Ibid., pp. 267-8.

<sup>2</sup> Ibid., p. 252.

<sup>3</sup> Lavoisier gave these details in the printed version of the memoir read at the rentrée publique of November 12, 1783, and included in the Mémoires for 1781, [p. 468], published in 1784. In the original reading he omitted reference to Macquer and Gengembre.

<sup>\*</sup>Dictionnaire de Chimie, 2nd edn., Paris, 1778, II, p. 583. Lavoisier subsequently gave the date of Macquer's work as 1776 or 1777 and stated that he was not then aware of it (Mém. Acad. R. Sci., 1781, pp. 469-70: published in 1784).

In June 1783, Blagden, 1 Cavendish's assistant. was in Paris and gave Lavoisier an account of the experiments that Cavendish had carried out in 1781. On June 24, 1783, Lavoisier in the presence of Blagden and others attempted a hurried and improvised verification of Cavendish's work. The record in the laboratory note-book reads: "In the presence of MM. Blagden, of [illegible], de Laplace, Vandermonde, de Fourcroy, Meusnier, Legendre, dephlogisticated air and inflammable air obtained from iron by vitriolic acid were combined in a bell-glass. . . . The amount of water can be estimated at 3 gros; I once I gros 12 grains of water ought to have been produced. Thus it must be supposed that two-thirds of the air was lost or that there was a loss of weight."2

Next day, June 25, 1783, the following report was submitted to the Academy and recorded in the Registers: "MM. Lavoisier and de Laplace announced that they had lately repeated in the presence of several members of the Academy the combustion of combustible air with dephlogisticated air; they worked with about sixty pints of these airs and the combustion was carried out in a closed vessel: the result was water in a very pure state."

On November 12, 1783, at the rentrée publique of the Academy, Lavoisier read a memoir entitled "On the Nature of Water and on Experiments that appear to prove that this Substance is not properly speaking an Element, but can be decomposed and

<sup>&</sup>lt;sup>1</sup> Charles Blagden. Knighted in 1792. Secretary of the Royal Society, 1784-97.
<sup>2</sup> Berthelot, p. 293.

<sup>&</sup>lt;sup>8</sup> Ibid., pp. 115-16.

recombined." This was printed at once in the December issue of Rozier's Journal1: and it appeared later in considerably extended form in the Mémoires, not for 1783, but for 1781, the volume for 1781 being still unpublished in 1783 and not appearing until 1784. A reading of the memoir, as published in Rozier's Journal, leaves no doubt that Lavoisier was fully aware of the outstanding importance of Cavendish's discoveries. It states that Lavoisier and Bucquet's experiments of 1777 had been confirmed (!) by Cavendish, who had also noted that "a sensible amount of moisture was produced." It proceeds: "As the verification of this fact was of extreme importance for chemical theory, M. Lavoisier & M. de la Place proposed to verify it by an experiment on a large scale; and in order that their work might be fully authenticated, they arranged for several members of the Academy to be present." A description of the experiment followed. About 30 pints of "inflammable air" were slowly burnt with 15 to 18 pints of "dephlogisticated air" in a kind of lamp with two very narrow jets, one for each of the gases and proportioned so as to supply each gas in the amount necessary for the combustion. The lamp was fixed under a glass receiver over mercury. Water condensed in the lower part of the apparatus and nearly all of it was collected by means of a funnel. It was pure as distilled water and it weighed nearly 5 gros, "which corresponded nearly to the combined weight of the two airs." The apparatus was

<sup>1</sup> Observations, 1783, 23, 452.

shown to the Academy, stated the memoir, "in July or August last." Later on, reference was made to the fact that shortly afterwards Monge had reported the results of other experiments on the production of water by the combustion of these two "airs" in a totally different apparatus and that a letter recently written from London by Blagden to Berthollet showed that Cavendish had lately repeated the same experiment in different ways with the same result. It was therefore difficult to deny the conclusion that the constituents of water were "inflammable air" and "dephlogisticated air," less the quantity of fire liberated in the combustion. But Lavoisier, ran the memoir, felt that proofs should be multiplied and the decomposition of water attempted in order to confirm these syntheses. He accordingly placed some pure ironfilings with a small amount of water in air over mercury. The iron rusted and a quantity of "inflammable air" was liberated proportional to the quantity of "dephlogisticated air" that had been absorbed by the iron, judging by the increase of weight found in the iron after it had been dried. Thus water had been decomposed into "dephlogisticated air," which united with the iron to form a calx, and "inflammable air" which was separated. Taken along with the synthetic experiments, this result showed that water was not an element. Lavoisier proposed to develop the work further.

The results reported in this memoir are insignificant, except perhaps the experiment—not a very satisfactory one—on the decomposition of water by

iron, beside the researches of Cavendish, whose paper was read shortly afterwards, on January 15, 1784, before the Royal Society of London. When Cavendish's paper was published, a paragraph was interpolated by Blagden, to the effect that he had informed Lavoisier of Cavendish's experiments in 1783 "as well as of the conclusion drawn from them, that dephlogisticated air is only water deprived of phlogiston; but at that time so far was M. Lavoisier from thinking any such opinion warranted, that, till he was prevailed upon to repeat the experiment himself, he found some difficulty in believing that nearly the whole of the two airs could be converted into water."

In the later version of this memoir which, although read before the Academy at the rentrée publique of November 12, 1783, was included in the Mémoires for 1781, published in 1784, there is the same indifferent notice of Cavendish's work. Internal evidence proves that the memoir was re-edited at least as late as May, 1784, since it mentions Blagden as Secretary of the Royal Society, an office to which he was not appointed until that date. The memoir includes the experiment carried out on June 24, 1783, but with the statement that the amount of water was a little less than 5 gros, the same quantity as that given in the Observations but not as entered in the laboratory record on June 24, 1783, namely, 3 gros. No amounts were quoted for the "airs" used because, it stated, the gases were fed into the apparatus by leather

<sup>1</sup> Phil. Trans., 1784, 74, 134-5.

tubes and it was known that these were not impermeable to air. However, since it was a principle none the less sound in physics than in geometry, that the whole is equal to its parts, Lavoisier felt that he was entitled to conclude that, since nothing but pure water was obtained in the experiment, the weight of this water was equal to that of the two "airs" from which it had been produced. It would be remembered that the Observations gave approximately the volumes of the "airs" used. Evidently, the data given here belong to some later repetition, not to the original experiment of June 24, 1783. Yet Lavoisier says that this was the experiment at which Blagden was present, but as shown above (p. 239) only 3 gros of water were obtained and there was a loss of two-thirds of the total weight of the "airs" on that occasion. The memoir described also later experiments made conjointly with Meusnier and the important confirmatory experiment in which water was decomposed by passing it drop by drop along a red-hot gun-barrel or along a red-hot copper tube containing iron, the principe oxygine from the water combining with the iron to form a calx and the principe inflammable aqueux being liberated and collected over water. There was no mention of this experiment with the gun-barrel in November, 1783.1 Moreover, the first reference to such an experiment in Lavoisier's laboratory note-books is dated March 1784, repetitions of the experiment being made on March 22 and 29.2 A faithful and

<sup>&</sup>lt;sup>1</sup> Cf. p. 241 above.

complete translation by Pelletier of Cavendish's paper as published in the *Philosophical Transactions* with the well-known additions made after its reading, the date of which is given as January 15, 1783, instead of 1784, appeared in two parts in the issues of Rozier's Journal for December 1784 and January 1785. On December 30, 1784, Lavoisier's laboratory note-books indicate that he then prepared for a further experiment on the composition and decomposition of water and that the experiments were carried out on February 27 and 28, 1785.<sup>2</sup>

Enough has been said to show that Lavoisier's claim to the discovery of the composition of water is inadmissible and that his attempts to verify Cavendish's work were indifferent. It remains however to point out that it was Lavoisier who correctly explained the facts discovered by Cavendish. We may note also that Meusnier and Lavoisier jointly carried out further experiments on the decomposition of water, that a memoir on this work was read at the Academy on April 21, 1784, and included in the volume of the Mémoires for 1781, which was not published until, as we have already seen, at least after May, 1784.

Lavoisier on several occasions made similar claims to the discovery of oxygen, stating that this gas had been discovered nearly at the same time by Priestley, Scheele and himself. This claim has never been admitted, even by Lavoisier's own countrymen; but it is fully recognized everywhere

<sup>1</sup> Observations, 1784, 25, 417 and 1785, 26, 38. 2 Berthelot, pp. 299-300.

that he discovered the composition of the air and the rôle of oxygen in combustion, calcination and respiration and that he gave the correct interpretation of Cavendish's experiments on the synthesis of water from its elements and confirmed the results analytically.

Before concluding this chapter we shall take note of another research carried out by Lavoisier and bearing on his theory. In the winter of 1782-3, Lavoisier in collaboration with Laplace constructed an ice-calorimeter for the measurement of specific and latent heats, some twenty years after Black's original discoveries in this field1; and the results of their work appeared in the volume of the Mémoires for 1780, which was published in 1784.2 Lavoisier had, as we have already seen, been much influenced by Black's work on "fixed air": he was no less influenced by Black's discovery of latent heat, his theory that aeriform fluids were compounds of evaporable liquids or volatile solids with the matter of heats being an obvious derivation from Black's idea that heat combined with ice to form water and with water to form steam. It was this combined heat in "vital air," for example, that he considered to be evolved in combustion, as in the burning of phosphorus. But in the last section of the memoir we are here referring to, Lavoisier and Laplace extended this idea to the

<sup>&</sup>lt;sup>1</sup> Grimaux (p. 118) eulogistically described Lavoisier and Laplace as the founders of calorimetry, whereas the quantitative science of heat had been founded twenty years earlier by Black. See McKie and Heathcote, ob. cit.

<sup>&</sup>lt;sup>2</sup> Mém. Acad. R. Sci., 1780, p. 355.

<sup>&</sup>lt;sup>8</sup> Cf. p. 217 above.

process of respiration. They measured the amount of "fixed air" produced in a known time by a guinea-pig breathing in "pure air" (oxygen) confined in a receiver over mercury; and they then determined the quantities of ice melted in the calorimeter, firstly, when the guinea-pig was put inside this apparatus for the same length of time, and, secondly, when the same amount of "fixed air" was produced directly from the combustion of charcoal. From the rough agreement between the quantities of ice melted, and therefore between the quantities of heat produced, they concluded that respiration was a slow combustion and that the constant temperature of the animal organism was maintained by means of the "matter of heat" that was liberated in the process of respiration by the combination of the "pure air" of the atmos-phere with the base of "fixed air" in the blood.

#### CHAPTER VIII

# THE NOMENCLATURE CHIMIQUE (1787) AND THE TRAITE ELEMEN-TAIRE DE CHIMIE (1789)

FOR SOME TIME, while Lavoisier had been extending his theory, there had been a movement, led principally by Guyton de Morveau, towards an improved chemical nomenclature: and from 1782 onwards Lavoisier, Berthollet and Fourcroy collaborated with de Morveau in devising a new system. The first results of their joint efforts appeared in a memoir read by Lavoisier at the rentrée publique of the Academy on April 18, 1787, on the necessity of reforming and perfecting the nomenclature of chemistry. Lavoisier's position among the chemists of his country and, with it, the recognition now being given to his theory are evident from the fact that, of the four chemists who had been engaged on this work, it was he who was chosen to read the first of the memoirs and to explain the principles on which the proposed changes in nomenclature were based. Lavoisier began by observing that de Morveau, recognizing that it was necessary, not only to create a new language, but also to secure agreement for its adoption, had called on the chemists of the Academy for advice and assistance. The problem had been complicated by the recent rapid advances in chemistry and by the rise of new theories. The detailed proposals would be given by de Morveau in a later memoir. For the present it would suffice to explain the principles that had guided the reformers. Languages, said Lavoisier, were, as the Abbé de Condillac had maintained, analytical methods by which man advanced from the known to the unknown and the art of reasoning was the art of analysing. The perfection of a language was therefore of the greatest importance in science. In the infancy of chemistry, suppositions rather than conclusions were drawn; the prejudices into which these had degenerated through centuries, and which had been accepted as fundamental truths even by the greatest minds, were now being corrected by experiments; and experiments, especially if they were carried out in the way that mathematicians proceeded to the solution of their problems, provided the only means of preventing reason from leading science astray. But the study and teaching of chemistry now required the introduction of such a method; and its introduction demanded a reformed nomenclature, since the logic of the sciences was necessarily connected with their language.

"If after having considered languages as analytical methods," proceeded Lavoisier, "we consider them simply as a collection of representative signs, they present us with some observations of another

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kind. In this second point of view, we shall have three objects to consider in every physical science, namely, the series of facts that constitute the science, the ideas that recall the facts, and the words that express them. The word should give birth to the idea and the idea should portray the fact: these are three impressions of the same seal. And, since it is the words that preserve the ideas and transmit them, it follows that it is impossible to improve the science without perfecting its language and that, however true the facts may be, however correct the ideas that they give rise to, they will still transmit only false impressions if we have no exact expressions to convey them. The perfection of chemical nomenclature, considered in this respect, consists in rendering the ideas and the facts in their exact truth, without suppressing anything they present and above all without adding anything to them: it must be nothing but a faithful mirror; for, we cannot repeat it too often, it is neither Nature nor the facts she presents, but our own reason that deceives us."1

These were principles, continued Lavoisier, that had not hitherto been applied to the language of chemistry: the terms in current use had come down from times when even the method of studying the science was not known. Many of the terms had been introduced by the alchemists and had been deliberately enigmatized by them. "It was thus," he said, "that the oil, the mercury and even the

<sup>&</sup>lt;sup>1</sup> Méthode de Nomenclature Chimique, by de Morveau, Lavoisier, Berthollet and Fourcroy, Paris, 1787, pp. 13-14.

water of the philosophers were neither oil nor mercury nor water in the sense that we attach to them. Homo galettus, the armed man, signified a cucurbit equipped with its head, the death's head was the head of an alembic, the pelican indicated a distilling vessel and the caput mortuum or terra damnata meant the residue from a distillation. Another class of philosophers who have not less disfigured the language of chemistry are the systematic chemists. They have expunged from the number of facts those that did not square with their ideas, they have in a manner misrepresented those that they did not wish to preserve and they have accompanied them with a solemnity of argument by which the facts themselves are lost sight of, so that the science in their hands is no more than a fabric of their imagination."1

These impediments to the progress of chemistry must be removed, added Lavoisier, and, although a new nomenclature even with the greatest care in its construction must be far from perfect, yet, if it is based on sound principles and if it is a method of naming rather than a nomenclature, it will adapt itself to future discovery, indicate the name and place of new substances that may be discovered and need only some local and particular changes. Discussion of the constituent principles of bodies would, he went on, be out of place at this time and therefore the reformers had here contented themselves with regarding as simple all substances that could not be decomposed into simpler substances, all

<sup>&</sup>lt;sup>1</sup> *Ibid.*, pp. 15-16.

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such as were the last result of chemical analysis1: these substances, now apparently simple, would doubtless be decomposed in the future "and we are probably approaching this epoch with regard to siliceous earth and the fixed alkalis," but imagination must not anticipate the facts. These simple substances, without doubt improperly called so, were the first to be named and, as most of them already had well-known names, these were retained except where they involved false ideas, in which case new names were given, usually derived from Greek and expressing the most general and characteristic properties of the substances designated.

With regard to bodies composed of two simple substances, their number was so great that classification was necessary. The acids formed one of these classes. They were composed of two simple substances, one of which was common to them all and gave rise to their acidity and the other of which was different for each acid; and thus the first gave a name to the class or genus, while the specific name was derived from the second component principle. But in most acids the two constituent principles, the acidifying principle and the acidified principle, existed in different proportions that were each points of equilibrium or saturation, as in vitriolic acid and sulphurous acid. These two states of the same acid were indicated by varying the termination of the specific name. The calces of

<sup>&</sup>lt;sup>1</sup> The first re-appearance of the definition given by Boyle in 1661 in the Sceptical Chymist. Cf. p. 59 above.

<sup>2</sup> Lavoisier's sound judgment is justified here: for in 1807 Davy decomposed these substances and isolated potassium and sodium, and a few years later Gay-Lussac and Thénard isolated silicon.

metals formed another class that could be treated in the same way.

Bodies composed of three simple substances presented greater difficulty: the nature of their constituent principles necessitated more complex names. "We have had to consider," said Lavoisier, "in the bodies that form this class, such as, for example, the neutral salts, firstly, the acidifying principle that is common to all, secondly, the acidifiable principle that constitutes their particular acid and, thirdly, the saline, earthy or metallic base that determines the particular kind of salt. We have taken the name of each class of salt from that of the acidifiable principle common to all the members of that class and we have then distinguished each kind by the name of its particular saline, earthy or metallic base."1 Changes in the terminations of the names would satisfactorily cover cases where salts were composed of varied proportions of the same three constituents. In this way, Lavoisier claimed, the reformers had succeeded so well that by a single word it was at once evident what was the combustible substance in any compound, whether this substance was combined with the acidifying principle and in what proportion, in what state the acid was, to what base it was united, if it was exactly saturated, and whether it was the acid or the base that was in excess.

"It can be imagined," concluded Lavoisier, that we could not have achieved these different objects without often doing violence to established

<sup>&</sup>lt;sup>1</sup> Méthode, pp. 21-2.

custom and without adopting denominations that may at first seem harsh and barbarous; but we have observed that the ear readily becomes accustomed to new words, especially when they are connected in a general and rational system. Besides, the names that are actually in use, such as powder of Algaroth, sal alembroth, pompholix, phagedenic water, turbith mineral, æthiops, colcothar and many others are neither less harsh nor less extraordinary; it needs much practice and a great memory to remember the substances that they express and especially to recognize to what kind of combination they belong. The names of oil of tartar per deliquum, oil of vitriol, butters of arsenic and antimony, stowers of zinc, etc. are still more ridiculous, because they give rise to false ideas, because, properly speaking, there does not exist in the mineral kingdom, and especially in the metallic, either butter or oil or flowers and, lastly, because the substances designated by these deceitful names are mostly violent poisons."1

The details of the new system were given by de Morveau in a memoir read before the Academy on May 2, 1787. The substances that chemists had not so far been able to decompose were divided into five classes, (1) those that approached nearest to a state of simplicity, (2) the acidifiable bases or radical principles of the acids, (3) the metals, (4) the earths and (5) the alkalis. The first class comprised light, matter of heat, dephlogisticated or vital air, inflammable gas and phlogisticated air. The matter of heat was re-named caloric,

<sup>&</sup>lt;sup>1</sup> *Ibid.*, pp. 23-5.

dephlogisticated or vital air became oxygen (oxus, acid; and gennao, I beget), inflammable gas became hydrogen (hudor, water; and gennao, I beget) and phlogisticated air azote (a, no; and zoe, life). The second class included charcoal, sulphur and phosphorus, which were known acidifiable bases or radicals of acids, as well as the muriatic radical, the boracic radical and the other radicals that had not been isolated but which by analogy with the first three members of this class were presumed to exist. The two acids of sulphur became sulphuric and sulphurous acids, their salts sulphates and sulphites. The term sulphuret (modern, sulphide) was also introduced. The pure principle of charcoal was named carbon and "fixed air" became carbonic acid and its salts carbonates. Similarly, the terms phosphates, nitrates, muriates, acetates, borates, etc. were introduced. In the third class, the metals retained their names but their compounds with oxygen were re-named oxides. The earths of the fourth class became silice (modern, silica), alumine (modern, alumina), barytes (modern, baryta) and magnesia alba became magnesia, while lime was retained. In the fifth class, "pure vegetable fixed alkali" and "pure mineral fixed alkali" were re-named potash and soda respectively.

Fourcroy prepared a table of the new terms, arranging substances in the classes named above and including their compounds, the older names being set alongside the new ones proposed and explanations of the system being given. An

<sup>&</sup>lt;sup>1</sup> Ibid., pp. 75-100.

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names followed1 and a dictionary of the new nomen-

clature was added.2

The historical significance of the new system of nomenclature is obvious; for it was based on the new theory of chemistry formulated by Lavoisier. As to its merits as a scientific instrument, it is enough to say here that with slight modification it has stood the test of a century and a half and still remains the basis of the language of modern chemistry. It is however interesting to turn to the report<sup>3</sup> on the new system presented to the Academy on June 13, 1787, by Baumé, Cadet, Darcet and Sage, who, without entering into a detailed discussion, made certain comments on the work. "This new theory, this table," they wrote, "is the work of four men, who are justly renowned in science and who have been long engaged upon it; doubtless they have not drawn it up without careful comparison of the bases of the ancient theory and those of the modern theory." The new theory rested on imposing experiments and it had been set up by the continued labours of men of genuis; but the old theory, the theory of phlogiston, had united the opinions of all chemists. The new views therefore required careful study, time, experiments and the calm reflection of chemists, because received ideas were not to be rejected in a day. The old theory was admittedly incomplete, but the new theory was not without its difficulties. It was not easy to renounce the principles in which chemists had been

<sup>1</sup> Ibid., pp. 101-43. 2 Ibid., pp. 144-237. 8 Ibid., pp. 238-52.

educated and it was still more difficult to admit suddenly that a large number of substances, indicated by every chemical analogy to be more or less compounded, must henceforth be regarded as simple, without taking into account the number that must be simplified every day, "as if we were again at the origin of things and at the first moments of the Creation."1 The new theory had its advantages in agreeing better with certain facts and it owed these advantages to the precision and exact calculation afforded by the improvement of apparatus; but it seemed that, in the experiments on the composition and decomposition of water, too little attention had been paid to the matter of heat because its weight was unknown. However, the work must be judged impartially; and it was thought that the new system, the theory and the nomenclature, should be left to the test of time. The table of the new nomenclature and the relevant memoirs should be printed and published under the privilege of the Academy in such a way that it might not be inferred whether the Academy approved or rejected the system.

This damning with faint praise of the new nomenclature together with the vague criticism of the new theory seems to have irritated Lavoisier; for, in a report<sup>2</sup> submitted to the Academy by Lavoisier, Berthollet and de Fourcroy on June 27, 1787, on the new chemical symbols suggested in two memoirs<sup>2</sup> by Hassenfratz and Adet, a report in which the hand of Lavoisier and the spirit

<sup>&</sup>lt;sup>1</sup> Ibid., p. 248. <sup>2</sup> Ibid., pp. 288-312. <sup>3</sup> Ibid., pp. 253-87.



PORTRAIT OF LAVOISIER

After an engraving by Mlle. Brossard-Beaulieu

"TRAITÉ ÉLÉMENTAIRE DE CHIMIE" 257 of the Reflexions of 1783 are both very evident, the new theory was again expounded at some length and the authors of the report on the new nomenclature described as the partisans of a theory which they referred to as ancient, whereas it had already been changed in such a way that there remained almost nothing of its original form; it was explained that, while the reformers would have been honoured if the Academy had adopted their system, they were content if the Academy would continue to treat with friendliness a theory formed in their midst, costing nearly twenty years' labour and already accepted by several distinguished chemists and in favour of which a much greater number now appeared ready to decide; and it was recommended that the work of Hassenfratz and Adet, which was adapted to the new nomenclature, merited the approbation of the Academy and should be published under its privilege.

That Lavoisier could turn on his critics in this way shows that in 1787 his theory had already won considerable acceptance among the chemists of the Academy. What was needed now was a complete and detailed exposition of the new system of chemistry. This Lavoisier gave two years later in his *Traité Elémentaire de Chimie*<sup>1</sup> (Paris, 1789),

<sup>&</sup>lt;sup>1</sup> The Traité was published under the privilege of the Academy, the report of Darcet and Berthollet being received by the Academy on February 4, 1789. Two editions were published at Paris in 1789, both in two volumes, the only apparent difference between the two editions being that the two volumes of the first edition are paginated continuously and those of the second edition separately. Two other editions followed in 1793 and 1801. A copy did not reach Edinburgh until the middle of September, 1790, and it was rapidly translated into English by Robert Kerr as Elements of Chemistry for the beginning of the University session at the end of October. Four other

a treatise which laid the foundations of modern chemistry as securely as Newton's Principia had a century earlier laid the foundations of modern mechanics. In the Preface Lavoisier explained that, when he began to write, he had intended merely to extend and explain the memoir that he read to the Academy in 1787 on the necessity of reforming the nomenclature of chemistry, but the work gradually developed, without his being able to prevent it, into a treatise on chemistry, because the nomenclature of the science and the science itself could not be separated. Repeating what he had said previously on the value of fact and experiment in the memoir referred to, Lavoisier added: "Convinced of these truths, I have imposed upon myself the law of never advancing but from the known to the unknown, of deducing no consequence that does not immediately derive from experiments and observations."1

The Traité, wrote Lavoisier, would contain nothing about the constituent and elementary parts of bodies. The tendency to reduce all bodies in nature to three or four elements was an ancient prejudice and a mere hypothesis. All that could be said about the number and nature of elements was confined to discussions that were purely metaphysical, to indeterminate problems capable of an infinite number of solutions, not one of which in all probability was consistent with nature.

editions appeared at Edinburgh, the last in 1802. German, Italian and Dutch translations were also published. The first French edition was already such a rare book in 1864 that Dumas, editing the Œuvres de Lavoisier for their official publication under the Minister of Education, had to make use of a copy of the second edition.

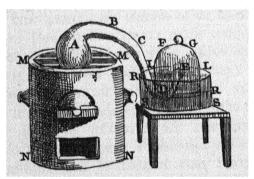
1 Trait, etc., p. xi.

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"I shall therefore content myself with saving," he added, "that, if by the term elements, we mean to express the simple and indivisible molecules that compose bodies, it is probable that we know nothing about them: but if, on the contrary, we express by the term elements or principles of bodies the idea of the last point reached by analysis, all substances that we have not yet been able to decompose by any means are elements to us: not that we can assert that these bodies that we consider as simple are not themselves composed of two or even a greater number of principles, but, since these principles are not separated, or rather since we have no means of separating them, they are to us as simple substances, and we must not suppose them compounded until experiment and observation have proved them to be so."1

The Traité was divided into three parts. The first part dealt with the formation and decomposition of aeriform fluids, the combustion of simple substances and the formation of acids, in other words, with Lavoisier's new system of chemistry. The second part treated of the combination of acids with bases and with the formation of salts; and the third part was devoted to a detailed description of the methods and apparatus of chemistry. With the exception of the table of chemical elements, which is given at the beginning of the second part and which we shall refer to below, it is the first part only that concerns us here. The first chapter of this part dealt generally with

the subject of heat and with the idea that elastic aeriform fluids, to which Lavoisier now gave the name of gases, were formed by the combination of caloric with the base of the gas. The second chapter extended these ideas to account for the formation and composition of the atmosphere; and the third chapter dealt with the analysis of atmospheric air and its division into two elastic fluids, the one fit and the other unfit for respiration. Here Lavoisier described for the first time with full details and in complete form the most famous of all his experiments, the apparatus for which is shown in Fig. 4 below. His description of the experiment ran as follows:



F10. 4

"I took a matrass A of capacity about 36 cubic pouces with a very long neck BCDE of internal diameter about 6 to 7 lines. I bent the neck as shown, in such a way that it could be placed on a furnace MMNN, while the extremity E of its neck could be inserted under the bell-jar FG, placed in a trough of mercury RRSS. I put 4 onces

of pure mercury into the matrass and then, by suction with a siphon which I introduced under the bell-jar FG, I raised the mercury to LL: I carefully marked this height with a strip of gummed paper and I noted accurately the readings of the barometer and thermometer.

"The apparatus being thus prepared, I lighted a fire in the furnace MMNN, which I kept up almost continually for twelve days, in such a way that the mercury was heated nearly to the degree necessary to make it boil.

"Nothing remarkable occurred during the first day: the mercury, though not boiling, was in a state of continual evaporation; it covered the inside of the vessels with small drops, which, at first very minute, afterwards increased in size and, when they had acquired a certain volume, fell back into the bottom of the vessel and were re-united with the rest of the mercury. On the second day, I began to see swimming on the surface of the mercury small red particles, which gradually increased in number and size during four or five days, after which they ceased to increase and remained completely unchanged. At the end of twelve days, seeing that the calcination of the mercury made no further progress, I extinguished the fire and allowed the vessels to cool. The volume of air contained in the matrass, in its neck and in the bell-jar, reduced to a pressure of 28 pouces and 10° [R.] of the thermometer, was about 50 cubic pouces at the beginning of the experiment. When the experiment was completed, the volume of the air, reduced to the same pressure and temperature, was only about 42 to 43 cubic *pouces*; a decrease in volume of about one-sixth had therefore occurred. On the other hand, having carefully collected the red particles that had been formed and having separated them as much as possible from the liquid mercury with which they were mixed, I found that their weight was 45 grains.

"I was obliged to repeat this calcination of mercury in closed vessels several times, because it is difficult in one and the same experiment to recover all the air experimented with and all the red particles or calx of mercury produced. It will often happen that I shall thus combine in one statement the result of two or three experiments of the same kind.

"The air that remained after the experiment, and which had been reduced to five-sixths of its volume by the calcination of the mercury, was no longer fit either for respiration or for combustion; for animals that were put into it perished in a few seconds and candles were extinguished immediately, as if they had been plunged into water.

"On the other hand, I took the 45 grains of red matter that had been formed during the experiment; I put them into a very small glass retort, to which was fitted an apparatus suitable for receiving the liquid or aeriform products that might be separated: having lighted the fire in the furnace, I observed that, in proportion as the red matter grew hot, the intensity of its colour increased. When the retort was almost glowing hot, the red matter began

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gradually to decrease in bulk and in a few minutes it disappeared completely; at the same time 41½ grains of liquid mercury were condensed in the small receiver and 7 to 8 cubic *pouces* of an elastic fluid, much fitter than the air of the atmosphere for supporting both combustion and the respiration of animals, passed over into the bell-jar.

"A part of this air being put into a glass tube of about 1 pouce diameter, a candle was plunged into it and burned in it with a dazzling splendour; charcoal, instead of being consumed quietly as in ordinary air, burned in it with a flame and a kind of decrepitation, like phosphorus, and with a brilliancy of light that the eyes could hardly bear. This air, which was discovered nearly at the same time by Mr. Priestley, Mr. Scheele and myself, was named by the first dephlogisticated air, and by the second, fire air. At first I gave it the name of eminently respirable air, for which has since been substituted that of vital air. We shall presently see what we ought to think of these denominations.

"In reflecting upon the circumstances of this experiment, we see that the mercury during its calcination absorbs the salubrious and respirable part of the air or, to speak more strictly, the base of this respirable part; that the part of the air that remains is a kind of mofette, incapable of supporting combustion and respiration; and therefore that the air of the atmosphere is composed of two elastic fluids of a differing and, if I may say so, opposite nature.

"A proof of this important truth is that, by

recombining the two elastic fluids that we have thus obtained separately, namely, the 42 cubic pouces of mofette, or irrespirable air, and the 8 cubic pouces of respirable air, we reproduce an air similar in every respect to the air of the atmosphere and fit very nearly to the same degree for combustion, the calcination of metals and the respiration of animals."

The experimental details given here correspond with those quoted in the memoir read to the Academy on May 3, 1777, on the respiration of animals,<sup>2</sup> but they are here set out in complete form and for the first time a diagram of the apparatus is included. After describing some similar experiments with iron, in which he claimed that the gain in weight of the iron was exactly equal to the loss in weight of the air, Lavoisier pointed out that the experiment with mercury determined the constituent parts of atmospheric air both by analysis and synthesis.

Chapter IV of the Traité was devoted to a consideration of the nomenclature of the components of atmospheric air. Here Lavoisier gave his approval to Macquer's re-introduction of the term gas, originally used by van Helmont, as a generic name for the various aeriform elastic fluids other than common air. And here again he explained, as he had already done in the Méthode de Nomenclature Chimique, the significance of the terms oxygen and azote as applied to the two constituents of common air, which was composed of oxygen gas, formed by the

<sup>&</sup>lt;sup>1</sup> *Ibid.*, pp. 35-9.

<sup>&</sup>lt;sup>2</sup> Cf. pp. 215-6 above.

union of caloric with the base oxygen, and azotic gas, formed by the union of caloric with the base azote. The fifth Chapter dealt with the decomposition of oxygen gas by sulphur, phosphorus and carbon and with the formation of acids in general. Experiments on the combustion of phosphorus in oxygen were described, evidently a development of the earlier work that we have already noted. With regard to sulphur, Lavoisier stated that he could not give accurate quantitative results; for, although he had proved that sulphur on burning increased in weight and that the increase of weight was equal to the weight of the oxygen absorbed, the relative proportions of sulphur and oxygen entering into the combination were uncertain.2 Much of what this chapter contained had been included in the memoirs already published. The sixth Chapter dealt with the nomenclature of the acids as given in the Méthode de Nomenclature Chimique; and the seventh Chapter explained the reasons for the introduction of the term oxide in place of calx. In Chapter VIII Lavoisier described, without touching on their disputed history, various experiments demonstrating the composition of water. Chapter IX gave the results of experiments carried out in the icecalorimeter on the quantities of caloric given out in various combustions. Chapter X dealt with the names given to compounds of combustible substances with one another, such as the compounds of

<sup>&</sup>lt;sup>1</sup> Cf. pp. 214-5 above. <sup>2</sup> Lavoisier laboratory note-books show that on May 18, 1785, he burnt sulphur and found that the combining proportions were 38.98 of sulphur to 61.02 of "vital air" (Berthelot, p. 301).

sulphur with the metals and of hydrogen with carbon, sulphur and phosphorus. Chapter XI dealt with the various vegetable and animal acids and Chapter XII was concerned with the decomposition of vegetable and animal substances by the action of heat: the bases of the vegetable acids were recognized as consisting either of hydrogen and carbon, or of hydrogen, carbon and phosphorus, while animal acids were more compound, their bases generally consisting of combinations of carbon, phosphorus, hydrogen and azote. This phenomenon of a double base in one acid resembled. according to Lavoisier, what had been observed in the compound acid, nitro-muriatic acid (aqua regia), and was continually met with in the animal and vegetable kingdoms.

In Chapter XIII Lavoisier described his researches on vinous fermentation. He pointed out that the fermentation of diluted fruit juices or other saccharine matter gave rise to the evolution of carbonic acid gas and the formation of a vinous liquor, from which the inflammable liquid formerly known as spirit of wine and now re-named alcohol was obtained by distillation. "This operation," wrote Lavoisier, "is one of the most striking and extraordinary of all those that chemistry presents to us; and we must examine whence proceed the disengaged carbonic acid gas and the inflammable spirit produced, and how a sweet body, a vegetable oxide, can thus be converted into two such different substances, whereof one is combustible and the other is highly incombustible. It will be seen that to

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solve these two questions, we must first know the analysis of the fermentable body and the products of the fermentation; for nothing is created in the operations either of art or of nature, and it can be taken as an axiom that in every operation an equal quantity of matter exists both before and after the operation, that the quality and quantity of the principles remain the same and that only changes and modifications occur. The whole art of making experiments in chemistry is founded on this: we must always suppose an exact equality between the principles of the bodies examined and those obtained by analysis. Thus, since must of grapes gives carbonic acid gas and alcohol, I can say that must of grapes = carbonic acid + alcohol. Thence it follows that we can ascertain in two ways what happens in the vinous fermentation, firstly, by determining the nature and the principles of the fermentable body and, secondly, by carefully observing the products of the fermentation, and it is evident that the knowledge that we can acquire about one of these leads to accurate conclusions about the nature of the other."1 Analysis showed that 100 parts of sugar were composed of nearly 8 parts of hydrogen, 64 parts of oxygen and 28 parts of carbon. Accordingly Lavoisier took 100 livres of sugar and added 400 livres of water and 10 livres of yeast from beer. The weights of the materials undergoing fermentation were set out as follows:

								Livres	Onces	Gros	Grains
Water								400	0	0	0
Sugar								100	0	0	0
Yeast fr	om	beer	made	into	∫Wat	er		7	3	6	44
a pas	te c	ompo	sed of	•	l Dry	yea	st	2	12	1	28
					Total	L		510	0	0	0

The compositions of these were then set out as follows:

		Livres	Onces	Gros	Grains
407 livres 3 onces 6 gros 44 grains of water composed $\{ H \\ O \}$	ydrogen	61	1 2	2	71.40
of O	xygen	346	2	3	44·6o
$\begin{array}{cccc} \text{100 livres of sugar composed} & H \\ \text{of} & \cdot & \cdot & \cdot & \cdot \\ C & \cdot & \cdot & \cdot & \cdot \end{array}$	ydrogen	8	o	О	o
O tures of sugar composed of	xygen	64	0	O	0
or (C	arbon .	28	O	О	О
O linner 10 orest 1 ores 28	arbon .		12	4	59.00
	zote .			5	2.94
posed of H	lydrogen Xygen		4	5	9.30
posed of (O	xygen	I	10	2	28.76
То	tal .	510	О	0	0

The amounts of the constituent principles were then recapitulated thus:

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The mixture was allowed to ferment for several days until fermentation had almost ceased. 35 livres 5 onces 4 gros 19 grains of dry carbonic acid were collected and the weight of water carried off by the carbonic acid was 13 livres 14 onces 5 gros. The weight of the residual liquor was 460 livres 11 onces 6 gros 53 grains. The analysis of the products was set out as follows:

Livres Onces Gros Grains		Livres	Onces (	Gros (	Frains
35 5 4 19 of oxygen carbonic acid carbon carbon.		25	7	1	34
composed of carbon .		9	7 14	2	57
composed of					
408 15 5 14 of [oxygen .		347	10	o	59
408 15 5 14 of oxygen . water composed of hydrogen .		61	5	4	27
• • •					
oxygen combin	ned		_		_
57 II I 58 of hydrogen com with oxygen of with oxygen hydrogen com with carbon carbon	n .	31	6	I	64
57 II I 58 of hydrogen com	bined	_	8	_	_
of hydrogen composed	hined	5	o	5	3
with carbon		4	o	5	0
carbon .		ıĜ	11	5	63
2 8 o o of (hydrogen .			2	4	o
dry acetic acid oxygen .		1	11	4	o
$\begin{array}{cccc} \textbf{2} & \textbf{8} & \textbf{o} & \textbf{o} & \textbf{of} \\ \textbf{dry acetic acid} & & \textbf{oxygen} & \textbf{.} \\ \textbf{composed of} & & \textbf{carbon} & \textbf{.} \end{array}$			10	0	O
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			5	I	67
residue of sugar oxygen .	•	2	9	7	27
composed of (carbon .	•	I	2	2	53
			2 13 6		
1 6 o 50 of hydrogen .	•		2	2	41
dry yeast composed oxygen	•		13	1 2	14 30
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	•		Ü	2	37
	•				
510 0 0 0 To	tal	510	0	0	o
description of the second of t					

A recapitulation of the results was then given as follows:

Livres Onces Gros Grains		Livres	Onces	Gros (	Grains
	∫in water	347	10	0	59
	in carbonic acid .	25	7	1	34
409 10 0 54	j in alcohol	31	6	I	64
of oxygen	in acetic acid .	I	11	4	0
	in residue of sugar	2	9	7	27
	(in yeast		13	1	14
	(in carbonic acid .	9	14	2	57
	in alcohol	16	11	5	63
28 12 5 59	√ in acetic acid .		10	o	Ō
of carbon	in residue of sugar	1	2	2	53
	lin yeast		6	2	30
	(in water	61	5 8	4	27
	in water of alcohol.	5	8	5	3
71 8 6 66	bon in alcohol .		0		0
of hydrogen	in acetic acid	4	2	5	0
or nydrogen	in residue of sugar		5	4 1	67
	in yeast		2	2	41
2 37 0	of azote		2	2	_
2 3/0	лагон				37
510 0 0 0	Total	510	0	0	0

At the end of these tables relating to an imposing experiment apparently carried out on a reacting system with a total weight of 510 livres, Lavoisier explained that he had experimented with only a few livres of sugar and then calculated the figures for the larger quantity; and hence the weights even to a grain had been inserted in the tables precisely as he had calculated them, although such accuracy could not be attained in practice. But what he was insisting on was that a chemical reaction could be formulated as an algebraic equation and that a kind of balance-sheet of quantities could be drawn

up for the reaction, since "nothing is created in the operations either of art or nature." Here for the first time in the history of chemistry we meet with a clear assertion that the Law of Conservation of Mass or the Law of the Indestructibility of Matter applies to chemical change, a principle that had already been implied in the researches of Black and Cavendish.

Chapter XIV of the *Traité* dealt with putrefactive fermentation briefly, little being known at that time about the composition of animal substances; and Chapter XV gave a short account of the acetous fermentation. Chapters XVI and XVII dealt with the formation of neutral salts.

Part II of the Traité included extensive tables of the compounds of the acids with various bases. But its most interesting and revolutionary feature was the table of simple substances or list of chemical elements given in the first section and reproduced overleaf. Discussing this table, Lavoisier wrote: "Chemistry in subjecting to experiments the various bodies in nature aims at decomposing them so as to be able to examine separately the different substances that enter into their composition. In our time this science has made very rapid progress, as will be easily seen by consulting the various writers on chemical systems. Oil and salt were formerly regarded as elements of bodies, whereas, experiment and observation having brought new knowledge, it has since been shown that the salts are not simple but composed of an acid and a base, and that their neutrality results from this combination. Modern

discoveries have further extended by several stages the limits of analysis1: they have enlightened us on the formation of acids and have shown us that acids are formed by the combination of an acidifying principle common to all, namely, oxygen, and of a radical that is particular to each acid. . . . Chemistry advances towards its goal and its perfection by dividing, subdividing and re-subdividing; and we do not know what the limit of its achievements may be. Therefore we cannot assert that what we at present suppose simple is so in fact: all that we can say is that such a substance is the actual limit that chemical analysis has attained and that it is not further subdivisible in the present state of our knowledge. We may suppose that the earths will soon cease to be counted among the number of simple substances; they are the only bodies in the whole of this class that have no tendency to combine with oxygen and I am much inclined to think that this indifference towards oxygen, if I might be allowed to use this expression, results from their being already saturated with it. The earths, according to this view, should be compound's substances, perhaps metallic oxides oxygenated up to a certain point. At the most this is only a mere conjecture. I hope that the reader will take care not to confound what I relate as truths of fact and experience with what is yet only hypothetical. I have omitted the

<sup>&</sup>lt;sup>1</sup> A footnote inserted at this point directs the reader's attention to Lavoisier's memoirs of 1776 and 1778 (Mém. Acad. R. Sci., 1776, p. 671 and 1778, p. 535. Cf. pp. 214 and 217 above).

<sup>2</sup> Simples in original, but composées seems intended. Lavoisier's conjecture proved correct. Davy decomposed lime, baryta and magnesia in 1808 and Wöhler isolated aluminium in 1827. Cf. p. 251 above, footnote 2.

	Noms nouveaux.	Noms anciens correspondans.
	Lumière	Lumière.
Substances sim-	Calorique	Chaleur. Principe de la chaleur. Fluide igné. Feu.
ples qui appar- tiennent aux trois règnes & qu'on peut regar- der comme les	Oxygène	Matière du feu & de la chaleur. Air déphlogifiqué. Air empiréal. Air vital. Bafe de l'air vital.
élémens d <del>es</del> corps.	Azote	Gaz phlogistiqué, Mosete, Base de la mosete, Gaz instammable,
(	Soufre	Base du gaz instammable. Soufre.
Subflances fim- ples non métalli- ques oxidables & acidifiables.	Phosphore Carbone Radical muriatique. Radical fluorique.	Phosphore. Charbon pur. Inconnu. Inconnu.
·	Radical boracique (Antimoine Argent	Incohnu. Antimoine. Argent. Arfenic.
	Bismuth	Bismuth. Cobolt. Cuivre.
Subflances fim- ples métalliques oxidables & aci-	Fer Manganèse Mercure.	Etain. Fer. Manganèle. Mercure.
difiables.	Molybdène Nickel	Molybdène. Nickel. Or.
	Platine Plomb Tungstène Zinc	Platine. Plomb. Tungstene. Zinc.
Substances sim-	Chaux Magnéfie Baryte	Terre calcaire, chaux: Magnésie, base du sel d'Epsom. Barote, terre pesante:
ples salifiables terreuses.	Alumine	Argile', terre'de l'alun, base de l'alun. Terresiliceuse, terre vitrissable.

.....

"TRAITÉ ÉLÉMENTAIRE DE CHIMIE" 273 fixed alkalis, potash and soda, from this table because these substances are evidently compound, although we do not know as yet the nature of the principles that enter into their composition."<sup>1</sup>

The publication of the Traité with its list of thirtythree elements marks the foundation of modern chemistry. We may have rejected Lavoisier's notion that oxygen is an essential constituent of acids and advanced far from his primitive conception of the thermodynamics of chemical change, but the theory set out in his great treatise is in all its essentials still the fundamental theory of chemistry. What Boyle had glimpsed only distantly in 1661 had now come to pass: and the four elements and the three principles and all the word-spinning that went with them were now swept clean away. To achieve the revolution that he had aimed at in 1773 Lavoisier had to build on the work of Black, Priestley and Cavendish, but the revolution proved far greater and struck much deeper than he had foreseen. And, although he built and improved on the work of others, Lavoisier is in every sense entitled to be named as the "Father of Modern Chemistry."

The Traité found rapid acceptance. In a letter written to Chaptal in 1791 Lavoisier could say: "Vous voir adopter les principes que j'ai annoncés le premier est pour moi une véritable jouissance. Votre conquête, celle de M. de Morveau et d'un petit nombre de chimistes épars en Europe est tout ce que j'ambitionnois, et le succès passe mes espérances, car je reçois de toutes parts des lettres

qui m'annoncent de nouveaux prosélytes, et je ne vois plus que les personnes âgées qui n'ont plus le courage de recommencer de nouvelles études ou qui ne peuvent plier leur imagination à un nouvel ordre de choses, qui tiennent encore à la doctrine du phlogistique. Toute la jeunesse adopte la nouvelle théorie et j'en conclus que la révolution est faite en chimie."

<sup>&</sup>lt;sup>1</sup> Grimaux, p. 126.

#### CHAPTER IX

#### LAST YEARS, 1789-94

BUT THE TIDE of another revolution was now rising fast about Lavoisier. The financial difficulties of France had become hopelessly insoluble and her people were groaning under oppressive taxes and social inequalities. The collapse of Calonne's magic methods of raising loans brought France to bankruptcy and in May 1789 the King was compelled to summon the States-General which had not met since 1614. By July the States-General had become the National Assembly; and an ominous National Guard was organized. Constitutional monarchy breathed awhile, but the Commune of Paris gained the real control of France. The National Assembly gave way to the Legislative Assembly and finally, in 1792, to the National Convention which declared France a republic. The King was guillotined in January 1793: a similar fate awaited the officials of the Ferme.

Lavoisier had long been a national figure. In 1785 the King had appointed him Director of the Academy of Sciences, and Lavoisier had then been able to carry out various long-needed reforms in its constitution. But, after the publication of the *Traité* in 1789, the early events of the revolution had kept

him from his laboratory and prevented him from continuing the researches on respiration that he had begun in collaboration with Séguin. Indeed, other and more insistent duties had already taken up much of his time. When local assemblies were set up in the last years of the monarchy to carry out the administration that was previously in the hands of provincial governors, Lavoisier was, in 1787, elected a representative for Romorantin in the Orléans Assembly; he was appointed secretary and took a very prominent part in the deliberations of the Assembly. He sat for the third estate, although his father had purchased the hereditary title of écuyer. He was much concerned about the inequalities in taxation between the various classes and the neglect of agriculture: and he prepared a scheme of voluntary contributory insurance among the poorer classes against ill-health and old age, but the intervention of a company of speculative financiers upset his plans. He did considerable work on the Committee for Agriculture, of which he was secretary, and he was a member of the Royal Society of Agriculture, which succeeded the Committee in 1788; and since 1778 he had carried out agricultural experiments on his estate at Fréchines, introducing new crops and better breeds of stock. Later he prepared an extensive work under the title De la Richesse territoriale du Royaume de France (Paris 1791), for which he received on March 15, 1791, the thanks of the National Assembly, who ordered that the work should be printed.

Lavoisier was elected to the States-General of

1789 as député suppléant for the nobility of Blois, who were also represented by the Vicomte de Beauharnais. He also became a representative for Culture-St.-Catherine to the Commune of Paris, a body that included many other Academicians. He was a prominent member of the 89 Club, founded to propagate the cause of constitutional monarchy; but constitutional monarchists became suspected of aristocratic leanings and finally accused of incivisme and, while the Jacobin Club grew, the 89 Club sank into decay and its members into political disrepute. Hassenfratz, indeed, was able to retain his membership of the Jacobin Club only by proving that he had been expelled from the 89. Lavoisier's scientific work, especially his work with Guettard, when engaged on which he had seen for himself the miserable poverty of the French peasants and had realized the unexploited agricultural wealth of the soil on which they struggled for a bare existence, and his association with Turgot led him to adopt the economic theories of the physiocrats who held that the soil of a country was its only source of wealth and therefore the only object of a just taxation, that a nation should be governed by those who were naturally fitted for the task and that trade and industry must be free and property secure. On many occasions Lavoisier gave publicity to his broad political views; but in the dismal passions of his time he was remembered for his connection with the Ferme.

In 1791 Lavoisier was venomously attacked by Marat in the issue of the latter's newspaper, L'Ami

du Peuple, for January 27. Marat was an old enemy: he had been ambitious for scientific distinction and had produced in 1780 his Recherches physiques sur le Feu, a work devoid of merit. The Journal de Paris incorrectly announced that the work had been approved by the Academy. Lavoisier had denied this report with some scorn.1 Marat's attack read: "I denounce to you the coryphæus of charlatans, the sieur Lavoisier, son of a land-grabber, pupil of the Geneva stock-jobber, farmer-general, controller of gunpowder and saltpetre, governor of the discount bank, secretary to the King, member of the Academy of Sciences. Would you believe that this little gentleman, who enjoys an income of 40,000 livres and whose only claim to public recognition is that he put Paris in prison by cutting off the fresh air with a wall that cost the poor 33 millions and that he removed the powder from the Arsenal to the Bastille on the night of July 12 and 13, is engaged on a devilish intrigue to get himself elected as administrator of the department of Paris? . . . Would to heaven that he had been strung to the lamp-post on August 6. Then the electors of la Culture would not need to blush for having nominated him." And there was more of it. Marat attacked other academicians as well, including Monge and Laplace, accusing them of spending in debauchery the money granted by the State for the study of aeronautics! But Lavoisier was the object of his greatest violence.

The Régie des Poudres was likewise attacked in the

<sup>&</sup>lt;sup>1</sup> Œuvres, IV, p. 360.

newspapers and the clubs, but Lavoisier protested in a letter to the Ministers of War and Marine that all their demands had been satisfied and that there was still a large reserve in the magazines. As a further reply to the unjust criticisms that were current, the four régisseurs published a defence of their work and pointed out that under their administration the nation had been relieved of an inequitable burden of 600,000 livres by the restriction of the right of excavation for saltpetre, that the output of saltpetre had increased from 1,700,000 to 3,700,000 livres and that the range of the powder had been doubled; and concluded with particulars of their public work, excusing themselves for giving these personal details on the grounds that their loyalty and their honour were involved.

When the Ferme was suppressed by decree of the National Assembly on March 20, 1791, Lavoisier was disappointed to find that none of the Ministers supported his claims to a position in the new financial administration. Presently however Delessart, the Minister of Finance, nominated him to the commission of the Treasury. Lavoisier declined to accept any salary for this work on the grounds that he was already paid as a régisseur des poudres. "Je demande," he wrote, "qu'il me soit permis de remplir gratuitement les nouvelles fonctions qui me sont confiées. Les émoluments dont je jouis comme régisseur des poudres, précisément parce qu'ils sont modiques, conviennent à ma manière de vivre, à mes goûts, à mes besoins, et dans un moment où tant de citoyens honnêtes perdent leur état, je ne pourrois, pour rien au monde, consentir à profiter d'un double traitement." This letter was published in the Moniteur for April 7, 1791: it may have been intended to disarm the critics of Lavoisier's wealth. In any event, Lavoisier wished to keep his position as régisseur des poudres; but, when the National Assembly reduced the number of régisseurs by one, he lost it because he was engaged at the Treasury. He protested that his entry into the Treasury had been conditional on his retention in the Régie: "Je compterai," he wrote to the Minister, "sur l'assurance que vous m'avez donnée de vive voix, au nom du roy, que mon droit à une place de régisseur des poudres ne seroit pas suspendu et que je conserverois la faculté de l'exercer dès qu'une place de régisseur deviendroit vacante. Je compterai également sur l'assurance du logement que j'occupe et dans lequel je me suis établi à mes frais, où j'ai fait des dépenses considérables en laboratoire, en cabinets relatifs aux sciences que je cultive et en instruments."2 The justice of his protest was admitted and he was allowed to retain his house at the Arsenal.

At the Treasury Lavoisier's skill was greatly appreciated by his colleagues; but disturbed by the march of events and fearing that the National Assembly would plunge the nation into war, he decided to limit his public activities to unsalaried work and resigned from the Treasury in 1792. He was at once recalled to the Régie des Poudres to replace Clouet, but the system was still being

<sup>1</sup> Grimaux, pp. 209-10.

<sup>&</sup>lt;sup>2</sup> Ibid., p. 210.

criticised and the régisseurs were suspect because they were originally nominees of the King. Accordingly Lavoisier resigned, but undertook to carry on his researches on gunpowder and saltpetre. He evidently wished to efface himself from all paid public offices. On June 15, 1792, he had already declined the King's invitation to become Ministre des Contributions Publiques, because he felt that he would not be able to discharge the duties of that office, since the legislature had already gone beyond the legal limits of the constitution. Lavoisier moved on August 15, 1792, from the Arsenal, where almost all his chemical researches had been carried out, to 243 Boulevard de la Madeleine. Three days later the other régisseurs were arrested and the aged Faucheux killed himself in despair. Lavoisier's premonitions had proved correct. As regards his work in the Régie, it is enough to say that the success of the new kind of war that was waged by the armies of Revolutionary France owed much to the ample supplies of good powder with which the labours of Lavoisier had stocked her magazines.

Since December 1791, Lavoisier had been treasurer of the Academy, but declined the salary while he was régisseur des poudres. From now onwards, until its suppression, the history of the Academy is largely the history of the Commission of Weights and Measures: and throughout this period Lavoisier, going in constant dread for his life, displayed his indomitable courage and tireless vigilance in defending the interests of science against the apathetic ignorance of the new rulers of France and,

as he was to realize later, the shameless intrigues of Fourcroy. The use of his private fortune in the affairs of the Academy, when State grants remained unpaid, and the payment of the pensions of the aged and infirm Academicians were but details in his widespread activities on behalf of science at this time.

The Academy however provided only temporary shelter from the storm. At the meeting of April 25, 1792, Fourcroy pointed out that the Society of Medicine had removed from its list of members the names of various noted counter-revolutionary émigrés and surprised his colleagues by proposing that the Academy should act similarly with regard to certain of its members well known for their incivisme. The menace of Fourcroy's words threatened every member of the Academy; for it was clearly not the proscription of mere émigrés that he had in mind. Several members promptly obiected that the Academy had no jurisdiction over the political views of its members. Fourcroy however persisted in his proposal, but consideration was postponed until April 29, on which date a motion was carried to submit a list of the members to the Minister of the Interior who would order dismissals, if there were to be any, while the Academy "would apply themselves, as was their custom, to more intellectual occupations." Fourcroy did not forget the humiliation of this defeat: he soon proved the most bitter and inflexible of the Academy's enemies.

In the autumn of 1792 Lavoisier paid his last visit to Fréchines and then returned to Paris to carry on

his work at the Academy and on the Commission of Weights and Measures, of which he was secretary and treasurer and for which the National Assembly had voted a grant of 300,000 livres, in the prompt payment of the instalments of which Lavoisier seems to have had some considerable difficulty. The Minister of the Interior returned the modified list of Academicians at this time and thus the Academy received some semblance of official recognition. All institutions deriving from the monarchy were however suspect and the Academy attempted a demonstration of their loyalty by appearing in a body at the bar of the National Convention on November 25, 1792, to present a volume of their Mémoires together with a report of the progress of the work on the new system of weights and measures. The Academy were favourably received and congratulated on their work: but three days later hostile elements in the Convention secured the passing of a decree forbidding the Academy to fill any of the vacancies in their ranks until further orders.

The Academy thus entered on the year 1793 under the menace of political interference and indeed of possible suppression. Lavoisier redoubled his efforts to save science from extinction in France: he was the life and soul of the dying Academy. He responded to the appeals of one Minister after another for scientific advice and assistance on technical problems; he continued the laborious work on the weights and measures: he secured new State grants and the payment of those overdue;

and he was constantly lobbying with the members of a government that was always ready to reap without sowing. Finally, the officials responsible for handing over the sums allocated to the Academy refused payment in accordance with the decree suppressing all corporations, until a new order passed the National Convention. Lavoisier therefore proached Lakanal, who was able to carry the necessary proposals through the Convention with some difficulty on May 25, 1793. Yet on July 10 no money had been paid over by the Treasury and again Lavoisier helped the Academy from his own means. There was however no open act of hostility towards the Academy, who on August 1 received the thanks of the National Convention for their continued labours on the weights and measures and were actually asked to give their advice on coinage problems. On August 8, a week later, the Convention voted for the suppression of all learned societies, after receiving a report from the Comité de l'Instruction Publique, which was intended to except the Academy from the operation of this decree, and to allow the continuance of the work on which the Academicians were engaged and of the State grants for that work, until a satisfactory scheme for the foundation of a new scientific society could be prepared. It appears that Lavoisier had some hand in devising this report. But when it came before the Convention, the painter David called for the suppression of all the funestes Académies, qui ne peuvent subsister sous un régime libre, and carried the Convention with him.

Lavoisier was however not yet defeated. He proposed to Lakanal the continuation of the official work undertaken by the suppressed Academy through such a new society as had been suggested in the report of the Comité de l'Instruction Publique, this new body to be under the control of a commission of the National Convention and to receive the emoluments previously paid to the Academy. On August 14, Lakanal succeeded in persuading the Convention to pass a decree that "La Convention nationale décrète que les membres de la ci-devant Académie des sciences continueront de s'assembler dans le lieu ordinaire de leurs séances pour s'occuper spécialement des objets qui leur ont été et pourront leur être envoyés par la Convention nationale. . . . Les attributions annuelles faites aux savants qui composaient la ci-devant Académie leur seront payées comme par le passé."1

But Lavoisier's victory was short-lived. When the Academicians met on August 17, it was to find their rooms sealed under the decree of August 8 and the decree of August 14 ignored. The Academy was defunct. It had been done to death by the venomous Fourcroy, who had carried the Comité de l'Instruction Publique against Lakanal at the very moment when the Academy had achieved satisfactory relations with the National Convention. Between August 20 and September 9, Fourcroy's proposals for continuing the work on the weights and measures under a commission were approved by the Comité and Lavoisier was called upon to assist. On September 14

<sup>&</sup>lt;sup>1</sup> *Ibid.*, p. 241.

Lavoisier's house was searched for the first time; such visits were shortly to become more frequent.

In this year also Lavoisier made his most notable contribution to the Bureau de Consultation des Arts et Métiers, which had been established in 1791 and of which he was a member. In his Réflexions sur l'instruction publique présentées à la Convention nationale par le bureau de consultation des Arts et Métiers (Paris, 1793), Lavoisier set out a detailed scheme for state education in the arts and sciences, involving free primary education as "a duty that society owes to the child." This remarkable document envisaged the complete democratization of opportunity, while recognizing differences in talent, urged the exclusive advantage of science as an educative factor not only for youth but for the whole human race, and included detailed curricula for the primary and secondary schools, and for twelve lycées nationaux in various parts of France and a Lycée central at Paris. Even the punishments in primary schools were discussed: punishment was only to be given after the instructor had referred the question to a jury of children.

Meanwhile, since 1789 the Ferme had been attacked as a pack of thieves and brigands, robbers of the people, and the cry had been raised that their fortunes should be confiscated by the State. But it was not until March 20, 1791, that the National Assembly suppressed the Ferme and called for accounts of all transactions since July 1, 1789. The preparation of these accounts was delegated to a commission of liquidators, appointed from the lease-holders. Lavoisier was however not chosen

as one of these and in 1791 he ceased all work with the Ferme. But the difficulty of preparing the accounts while having to carry out their normal current official duties prevented the commission from completing their work by January 1, 1793, the date originally fixed by the National Assembly. On June 5 the National Convention suppressed the commission. A few months later a decree ordered that the personal papers of the Fermiers Généraux should be sealed: and on September 10 and 11 two members of the Revolutionary Committee visited Lavoisier's house to search his papers and affix the necessary seals, Romme and Fourcroy accompanying them to identify and to avoid the sealing of anything relating to the work on weights and measures being carried out for the Comité de l'Instruction Publique. Lavoisier protested with dignity that he had left the Ferme some years previously, that he had declined to receive what was owed to him, that he had since acted as a commissioner of the Treasury gratuitously, that he had given up this office to prosecute scientific researches on matters of public utility, and that he could not believe that he was among those to whom the decree was intended to be applied, but that he was prepared to submit to the searching of his papers although he wished to state his objections. The searchers declared that there was nothing suspicious in any of the papers written in French, but decided to hand a packet of letters written in English to the Comité de Sûreté Générale, Lavoisier insisting on affixing his seal to these alongside the official seal.

This packet was afterwards found to consist of letters from Priestley, Black, Wedgwood and other scientists. On September 24, the Convention ordered the removal of the seals from the papers of the Fermiers Généraux, so that they might prepare their accounts by April 1, 1794. Lavoisier received notice of the raising of the seals on his papers on September 28 as follows:

"Les secrétaire-greffier de la section des Piques au citoyen Lavoisier:

"CITOYEN,

"Je m'empresse de vous faire parvenir le procès-verbal relatif à la levée des scellés qui avaient été apposés chez vous ; tout ce qu'il contient rend hommage à votre civisme et est susceptible de dissiper toute espèce de soupçon.

Paris, 28 Septembre 1793, l'an 2<sup>e</sup> de la République.

BAILLIE."1

But a successful clamour was now raised in the National Convention by Dupin for an examination of the accounts of the Ferme by a commission, the members of which offered to denounce the various abuses of which the Ferme had been guilty and to present information upon the matter. Dupin's nominees were all former employees of the Ferme; and the chief amongst them was Gaudot, a convicted thief and forger. Even with this threat upon them, the Fermiers Généraux expected to be faced only with concocted evidence that could be easily

rebutted and at the worst they feared the confiscation of their property: it is possible that it was at this time that Lavoisier remarked that he expected to lose his fortune, in which case he would work as a pharmacist. It seems clear that the Ferme had no suspicion either of the hatred against them or of the peril that was about them. On November 14, the National Convention, roused by an angry speech on the delay in the presentation of the Ferme's accounts, ordered the arrest of the Fermiers Généraux, although the recently passed decree of September 24 had fixed the date for the rendering of the accounts as April 1, 1794. Meanwhile, Lavoisier was denounced in the Convention for being simultaneously in the Ferme and the Régie des Poudres by a former salpétrier, who had probably other interests than justice on his mind; later an anonymous informer accused him of conducting a correspondence with the émigré Blizard, but this was disproved before the Revolutionary Committee; and at the instigation of the toadying Fourcroy his name was removed from the list of founders of the Lycée on the grounds that he was a counter-revolutionary.

When the decree of arrest was confirmed by the Convention on November 24, Lavoisier was presiding at the Bureau de Consultation. He heard the news in the evening when on guard-duty at the Arsenal as a member of the National Guard and, fearing immediate arrest, took refuge with Lucas, a former usher at the Academy. He appealed to the Comité de Sûreté Générale to be allowed to carry on his work on the weights and measures. But the appeal

went unanswered and no friend seems to have spoken for him. Accordingly he gave himself up on November 28 and joined the other Fermiers Généraux including Paulze, his father-in-law, in the prison of Port-Libre, formerly the Convent of Port-Royal. The older prisoners such as Paulze suffered much from the cold and the prison was overcrowded: the prisoners were allowed to order their own meals and to take them together and were expected to prepare the accounts of the Ferme while thus in custody. Lavoisier wrote to his wife that he was making his room more comfortable and was preparing his memoirs on chemistry for publication in complete form: and he advised her not to spend her strength in useless attempts to secure his release. Mme. Lavoisier, however, continued her efforts, but all that she could gain was permission to visit her husband at Port-Libre.

The situation of the Fermiers Généraux was desperate. They could not prepare their accounts without documents, and the mass of documents was so large as to prohibit their being brought from the Hôtel des Fermes to Port-Libre. Accordingly they appealed to be removed to the Hôtel des Fermes. Lavoisier had little hope of the success of this plea.

On December 18, Borda and Haüy appealed on behalf of the Commission of Weights and Measures for Lavoisier's release to carry on the work, but the Comité de Sûreté Générale ignored the appeal. On December 20, after receiving a report from the Comité de l'Instruction Publique, which included Fourcroy and de Morveau among its members, the

Comité de Salut Public removed Borda, Laplace, Lavoisier and others from the Commission of Weights and Measures. The Comité des Assignats et Monnaies also appealed in vain on December 21 for Lavoisier's release. Meanwhile, there was no reply to the appeal of the Fermiers Généraux to the Convention and a new petition was presented on December 11, on receipt of which the Convention, in the apparent belief that the Ferme was likely to disgorge 300 to 400 millions, ordered the removal of the prisoners to the Hôtel des Fermes under surveillance and the confiscation of all their property and possessions and the sealing of their residences. The seals were placed on Lavoisier's house on December 17. They were raised twice, once for de Morveau and Fourcroy to remove objects belonging to the Commission of Weights and Measures, Lavoisier being brought from prison in charge of two guards to witness the removal, an encounter of which Grimaux wrote: "Qui nous dira les regards que purent échanger le prisonnier et les conventionnels? Ouels sentiments devaient agiter ceux-ci en voyant devant eux, dans la condition humiliée d'un criminel, l'ancien directeur de l'Académie, l'opulent fermier général, le savant illustre dont ils avaient si souvent sollicité l'appui et les suffrages?"1

On December 19, Lavoisier wrote to his wife:

<sup>&</sup>quot;Tu te donnes, ma bonne amie, bien de la peine, bien de la fatigue de corps et d'esprit, et moi je ne puis la partager. Prends garde que ta santé ne

<sup>&</sup>lt;sup>1</sup> Ibid., p. 277.

s'altère, ce seroit le plus grand des malheurs. Ma carrière est avancée, j'ay joui d'une existence heureuse depuis que je me connois, tu y as contribué et tu y contribues tous les jours par les marques d'attachement que tu me donnes; enfin je laisserai toujours après moi des souvenirs d'estime et de considération. Ainsy ma tâche est remplie, mais toi qui as encore droit d'espérer une longue carrière, ne la prodigue pas. J'ay cru m'apercevoir hier que tu étais triste; pourquoi le serois-tu, puisque je suis résigné à tout et que je regarderai comme gagné tout ce que je ne perdrai pas. D'ailleurs nous ne sommes pas sans espérance de nous rejoindre et, en attendant, tes visites me font encor passer de doux instans."

The Fermiers Généraux were transferred to the Hôtel des Fermes on December 24 to much worse conditions than those of Port-Libre, some of the prisoners having no beds. Most of them now had no money and ate at the charity of their fellows. But at last they had access to their papers and toiled for ten hours daily at the preparation of their accounts, the completion of which most of them supposed would enable them to go free. The work was completed and sent to the Comité des Finances on January 27, 1794, with a refutation of the charges preferred by Dupin's nominees, Gaudot and his associates. The Fermiers Généraux now claimed their liberty, but in vain, because Gaudot objected that it was necessary to examine the accounts. After this examination, even Gaudot could not assess the sum owed by the Ferme to the State at more than 130 millions; but various further charges were preferred of abuses of privilege, excessive rates of interest and adulteration of the tobacco by increasing the moisture added, the mouillade, beyond the legal amount permitted and en faisant payer l'eau introduite au prix du tabac, manœuvre aussi dangereuse pour la santé du consommateur que nuisible à ses intérêts! It was the mouillade that brought death to the Fermiers Généraux and the title of Dupin-Mouillade to their enemy. Faced with this new charge, Lavoisier showed from the records that the greatest part of the water necessarily added in the manufacture evaporated before the tobacco was sold and reminded the critics of the Ferme that the price at which it was sold to retailers was, on the instruction of the Ferme, calculated according to the dry weight. Meanwhile, Fouquier-Tinville sent Danton and other founders of the Republic to the guillotine. Lavoisier prepared his defence, certificates of his services from the Bureau de Consultation and the Régie des Poudres, an attestation, delivered to him in prison by his former fellow-Academicians, Cadet and Baumé (passionate opponent of the new chemistry), of his opposition to the adding of moisture to the tobacco, and a personally compiled résumé of his scientific work and official labours. Some of his friends wished to take steps on his behalf, but, fearing that they might thereby endanger their safety, he refused to accept their help. Mme. Lavoisier, in defiance of the decree forbidding all ex-nobles from entering Paris, made even at the last moment a spirited attempt to save her husband from the dreaded Revolutionary tribunal. Pluvinet, the pharmacist of the Rue des Lombards who provided Lavoisier with the chemicals for his laboratory. conceived the idea of saving Lavoisier by soliciting the help of Dupin himself! This bold plan deserved a better fate. Pluvinet was acquainted with Dupin's sister-in-law, a lady of unconventional manners, who at Pluvinet's suggestion persuaded Dupin to arrange for Lavoisier to be separated from his fellowprisoners and transferred to another prison and for the report to contain nothing that might be in his disfavour. Dupin, however, complained that Mme. Lavoisier had not called upon him, but had relied on her aristocratic friends. Mme. Lavoisier, warned by Pluvinet, promptly called upon Dupin, but, instead of appearing as a suppliant, declared that she had not come to beg for pity for her husband, who was innocent, that only scoundrels could accuse him. and that he would be dishonoured if his case was separated from those of his colleagues, whose lives were forfeit for their fortunes and who, if they died, would all die innocent men. Dupin, in irritation at this attitude, then turned a deaf ear to all appeals.

Dupin brought the report of his commission before the National Convention on May 5, 1794, and, after a speech rank with injustice, obtained a decree that the Fermiers Généraux should be brought before the Revolutionary tribunal. This tribunal had been set up expressly to deal with acts of incivisme, of which, it is hardly surprising to note, the Fermiers Généraux had not been accused. The

decree was passed about four o'clock and the information was immediately carried to the Fermiers Généraux by one who was present and who, on arrival at the Hôtel des Fermes, met Lavoisier and broke the news to him. To Lavoisier thus fell the unpleasant task of informing his associates. No further visitors were allowed to see the prisoners, who were moved to the Conciergerie at five o'clock. The decree was passed to the Revolutionary tribunal next day, May 6: but Fouquier-Tinville had prepared the charge sheets before Dupin had spoken in the Convention on the previous day and there is little doubt that Dupin had helped him to do so.

At half-past seven on the morning of May 7, the Fermiers Généraux were put separately through some empty formalities of interrogation, but they had no notice of when they were to appear before the tribunal. It was probably at this time that Lavoisier wrote the following letter to his cousin Augez de Villers: "I'ai obtenu une carrière passablement longue, surtout fort heureuse, et je crois que ma mémoire sera accompagnée de quelques regrets, peut-être de quelque gloire. Qu'aurais-je pu désirer de plus? Les événemens dans lesquels je me trouve enveloppé vont probablement m'éviter les inconvéniens de la vieillesse. Je mourai tout entier, c'est encore un avantage que je dois compter au nombre de ceux dont j'ai joui. Si j'éprouve quelques sentimens pénibles, c'est de n'avoir pas fait plus pour ma famille; c'est d'être denué de tout et de ne pouvoir lui donner ni à elle ni à vous aucun gage de mon attachement et de ma reconnaissance. Il est

donc vrai que l'exercice de toutes les vertus sociales, des services importans rendus à la patrie, une carrière utilement employée pour le progrès des arts et des connaissances humaines ne suffisent pas pour préserver d'une fin sinistre et pour éviter de périr en coupable! Je vous écris aujourd'hui, parce que demain il ne me serait peut-être plus permis de le faire, et que c'est une douce consolation pour moi de m'occuper de vous et des personnes qui me sont chères dans ces derniers momens. Ne m'oubliez pas auprès de ceux qui s'intéressent à moi, que cette lettre leur soit commune. C'est vraisemblablement la dernière que je vous écrirai." And it was now, too, that a deputation from the Lycée des Arts visited Lavoisier in prison to confer on him a now unidentifiable distinction awarded to him three days previously. This display of courage is refreshing amidst the miserable knavery that surrounded Lavoisier in his last hours.

At one o'clock next morning the thirty-two prisoners were each given a copy of the charges laid against them. The writing was illegible and, in any case, they were ordered to extinguish their lights. At dawn thirty-one of them were taken to await their trial before the tribunal: one had been removed from the list by the influence of friends. They were introduced to their four official defenders who were given a quarter of an hour to complete their consultations. At ten o'clock they were brought before the tribunal, Coffinhal presiding. There was the customary display of National Guards with fixed bayonets and the usual interested crowd.

<sup>&</sup>lt;sup>1</sup> *Ibid.*, pp. 296-7.

Coffinhal asked some brief questions to establish the identity of the prisoners, whose replies were received with derision from both judges and jury. Then the public-prosecutor, Liendon, got the worst of several exchanges with one of the accused, St.-Amand, who was thereupon brutally ordered by Coffinhal to answer only yes or no. The next moment an order arrived from the National Convention for the release of three of the accused, to whom the decree did not apply since they had not been engaged in the Ferme during the period specified. After this interruption, Liendon rose to ask a few trivial questions and then to make the usual ranting appeal with the peroration that la mesure des crimes de ces vampires était au comble, qu'ils réclamaient vengeance, que l'immoralité de ces êtres êtait gravée dans l'opinion publique, et qu'ils étaient les auteurs de tous les maux qui pendant quelque temps avaient affligé la France. Then the four counsel for the defence were heard—for twenty-eight prisoners en masse after a consultation of fifteen minutes! And Hallé submitted to the tribunal the report of the Bureau de Consultation on Lavoisier's scientific work, on which Coffinhal is said to have remarked: "La République n'a pas besoin de savants, il faut que la justice suive son cours." The tribunal however had no authority to deal with such offences as the Fermiers Généraux were accused of, if committed before the Revolution. Coffinhal with his cunning genius disposed of any possible scruples about such a mere formality by inviting the jury to ask themselves whether or no there had been a plot against the people involving

unfair exactions, adulteration of the tobacco with water and ingredients deleterious to the health of the citizens, and supplying the enemies of the Republic with immense sums of money withheld from the Treasury of the nation. Coffinhal's genius had its imaginative side: plotting with foreign nations was punishable by death.

The jury unanimously declared the accused guilty. Time pressed. The tumbrels were waiting for the day's batch of victims. Judgment of death and confiscation of property was pronounced at once, so instantly that it had to be recorded later. The section of the penal code under which these penalties were inflicted and which Coffinhal read in accordance with procedure was as follows: "Toute manœuvre, toutes intelligences avec les ennemis de la France tendant, soit à faciliter leur entrée dans les dépendances de l'empire français, soit à leur livrer des villes, forteresses, ports, vaisseaux, magasins ou arsenaux appartenant à la France, soit à leur fournir des secours en soldats, argent, vivres ou munitions, soit à favoriser d'une manière quelconque le progrès de leurs armes sur le territoire français ou contre les forces de terre ou de mer, soit à ébranler la fidélité des officiers, soldats ou des autres citoyens envers la nation française, seront punis de mort." The formal reading of this irrelevant section of the code, the final touch to a sublime farce, was followed by an order to the public prosecutor to carry out the judgment within forty-eight hours. The victims were then taken back to the Conciergerie and promptly conveyed thence to the Place de la Revolution and guillotined forthwith in the order in which they were named in the list. Paulze was third and Lavoisier fourth: and thus, while awaiting his own fate, Lavoisier saw his father-in-law executed before his eyes. The bodies of the victims were thrown into nameless graves in the cemetery of la Madeleine.

Thus perished Lavoisier on May 8, 1794. "Only a moment to cut off his head," said Lagrange to Delambre next day, "and perhaps a hundred years before we shall have another like it." But criticisms and appreciations had to be whispered when the Terror was raging through France. Mme. Lavoisier accused Lavoisier's fellow-scientists of her husband's death. Certainly, there is no evidence that Monge or Hassenfratz or de Morveau or Fourcrov, all of whom possessed either authority or influence, raised a hand to help him, while Borda, an ex-noble, and Haüy, a former Abbé, both of whom were therefore suspect, courageously protested against Lavoisier's arrest and Hallé appeared for him even before Coffinhal. Fourcroy, indeed, climbed to eminence on Lavoisier's scientific work: he had even gone so far as to describe the new chemical theory as la théorie des chimistes français, which had drawn from Lavoisier the protest: "Cette théorie n'est pas, comme je l'entends dire, celle des chimistes français, elle est la mienne : c'est une propriété que je réclame auprès de mes contemporains et de la postérité."

Mme. Lavoisier, intent on avenging her husband's death and having subsequently suffered

imprisonment and the confiscation of her property, had no small hand in a pamphlet denouncing Dupin and published on July 10, 1795. Dupin could make only a wretchedly inadequate defence and retired from a public life that had been distinguished only by his iniquitous pursuit of the Fermiers Généraux. It was recognized that a cruel and irreparable injustice had been perpetrated: and the personal possessions of the victims were returned to their relatives, the inventories made at the time of the confiscations being so carefully drawn up that Lavoisier's library at Fréchines lacked only three volumes when it was re-assembled. Finally, a new examination of the accounts of the Ferme showed that, instead of the Ferme owing the nation 130 millions, the nation owed the Ferme 8 millions; but no claim for this money was preferred by their relatives. Mme. Lavoisier was for some time engaged in preparing her husband's uncompleted memoirs for publication. These were published in two volumes in 1805 under the title Mémoires de Chimie, Mme, Lavoisier justifiably refusing to accept a pretentious introduction from Séguin who claimed for himself a great share in the work that was described in these two volumes. Copies of the Mémoires were presented to various eminent scientists throughout Europe: none were sold, except those left at Mme. Lavoisier's death. On October 22, 1805, Mme. Lavoisier married Count Rumford, but the marriage was unhappy and was followed by a mutual agreement separate: she died suddenly in Paris on February 10, 1836, at the age of seventy-six.

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