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U. S. DEPARTMENT OF AGRICULTURE,
OFFICE OF PUBLIC ROADS—Circular No. 97.
LOGAN WALLER PAGE, Director.

COKE-OVEN TARS OF THE UNITED STATES.

BY

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UNIVERSITY OF CALIFORNIA
AT LOS ANGELES
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LETTER OF TRANSMITTAL.

UNITED STATES DEPARTMENT OF AGRICULTURE,
OFFICE OF PUBLIC ROADS,
Washington, D. C., October 19, 1911.

SIR: I have the honor to transmit herewith the manuscript of a circular by Mr. Prévost Hubbard, chemist in this office, entitled "Coke-Oven Tars of the United States." This publication gives the results of examinations of all the coke-oven tars at present manufactured in this country, together with a brief discussion of their properties in relation to their use as road materials. I respectfully request that this manuscript be published as Circular 97 of this office.

Respectfully,

LOGAN WALLER PAGE,
Director.

HON. JAMES WILSON,
Secretary of Agriculture.

COKE-OVEN TARS OF THE UNITED STATES.

The rapidly increasing use in this country of refined coal tar in the treatment and construction of roads and the fact that an immense quantity of coal tar will ultimately become available for this purpose through the installation of by-product coke ovens make it highly desirable to obtain accurate information as to the properties of coke-oven tars which are being produced at present. Attention was called by the author to the importance of this subject in a recent publication,¹ as follows:

While, in the manufacture of coal gas, the production of tar is absolutely unavoidable, this is not true of the manufacture of coke for metallurgical purposes. There are two general types of coke ovens in use at present, in one of which no attempt is made to recover the volatile products of the coal. This is the oldest form of oven, known as the "beehive," and is extensively used in this country to-day. It is constructed of brick and as its name implies has the form of a beehive. Bituminous coal is placed in this oven or kiln and a part of it burned in order to carbonize the remainder, while the volatile products, such as gas, ammonia, and tar, are allowed to escape through an opening in the top of the kiln, where they are lost in flame and smoke.

Coke ovens in which the by-products are saved are now used to some extent in this country, and sooner or later will undoubtedly replace the old-style oven entirely, and thus increase our output of tar enormously. The reason that they have not been more generally adopted in this country is that in the United States tars are of much less economic importance than in the European countries, where great chemical industries are based upon the utilization of this material. Germany in particular is far in advance of us in this field and exports to this country alone coal-tar products to the value of several million dollars each year. With the development of the road-tar industry, which promises to consume vast quantities of tar, and the necessity for refining such tars before use, the general adoption of by-product ovens is only a matter of time. What this will mean in the increase in tar production can be imagined from the fact that in 1908, out of a total of over 26,000,000 tons of coke produced in coke ovens, only a little over 4,000,000 tons were obtained from by-product

¹ Dust Preventives and Road Binders, pp. 239-240, New York.

ovens. About 22,000,000 tons of coke were, therefore, produced without recovery of the tar. As the average yield of coke per ton of coal was 66 per cent, this would represent the consumption of over 33,000,000 tons of coal. Upon the basis of a yield of 10 gallons of tar per ton of coal, it may be seen that over 330,000,000 gallons of tar were lost in 1908 which might have been saved. As the actual production of coal tar both from coke ovens and gas houses amounted to about 101,000,000 gallons, it is evident that over three-fourths of our possible production of tar as a by-product was lost during that year. At a valuation of $2\frac{1}{2}$ cents per gallon, this means a loss of over \$8,000,000. With such an increase in production, however, the monetary value of coal tar would have dropped, so that this figure may be somewhat exaggerated. In any event, at a conservative estimate, the tar lost each year from nonrecovery coke ovens is sufficient to build 9,000 miles of tar macadam road 15 feet wide.

This estimate was based on data taken from reports of the United States Geological Survey. In a later report by Parker,¹ it is shown that over 53,000,000 tons of coal were consumed in beehive ovens in 1910, so that on the same basis it would appear that over 530,000,000 gallons of tar were lost during that year. The output of tar from by-product coke ovens, however, has also continued to increase, as shown by the following figures taken from this report:

| | Gallons. |
|-----------|--------------|
| 1908..... | 42, 720, 609 |
| 1909..... | 60, 126, 006 |
| 1910..... | 66, 303, 214 |

The tar thus produced in 1910 was valued at \$1,599,453, or about 2.4 cents per gallon. It is evident, therefore, that the value of the tar lost during that year by the use of beehive ovens amounted to approximately \$12,000,000. That the use of by-product ovens is increasing in greater proportion than the use of the beehive oven is shown by the fact that for the former type the increase in 1910 over 1909 in tons of coke produced was 14.13 per cent, while the production from beehive ovens increased by only 4.57 per cent. Parker adds, however:

While noteworthy progress has been made in the substitution of modern retort-oven practice for the wasteful and what should be obsolete beehive or partial-combustion method of coke making in the United States, this country is still much behind European countries in this regard.

He says further:

The yield of coal in coke in retort ovens exceeds that obtained in beehive or other partial-combustion ovens by about 15 per cent, and generally the quality of the coke is improved.

¹ The Manufacture of Coke in 1910, United States Geological Survey.

In order to determine the character of coke-oven tars at present being produced in the United States, it was first necessary to obtain samples from all of the known plants, and for this purpose reference was made to a list of by-product and retort coke-oven plants of the United States and Canada (Jan. 1, 1910), which was kindly loaned to the author by Mr. Parker in advance of its publication.¹ Letters were then written to each plant asking for the following information:

- (1) a. At what maximum temperature are your retorts fired in your _____ ovens?
In your _____ ovens?
- b. What is the maximum temperature to which the charge of coal is brought in your _____ ovens? In your _____ ovens?
- (2) What is the specific gravity of your crude tar in your _____ ovens? In your _____ ovens?
- (3) What percentage of free carbon is found in your crude tar from your _____ ovens?
From your _____ ovens?
- (4) Will you furnish us for examination a 1-gallon sample of your crude tar from your _____ ovens? From your _____ ovens? To be sent at our expense.

Very courteous replies were received from the manufacturers, and in practically every case samples of tar were also forwarded. The blanks in these questions were filled out by inserting the names of the type or types of ovens operated by the manufacturer to whom they were sent. It was found that, where two types of ovens were operated at the same plant, no attempt was made to separate the tar, and the entire output was run into a common well. In such cases the sample of tar submitted was a mixture obtained from both ovens. The report of these samples is given at the bottom of Table I.

The questions concerning temperature were asked because criticism from an authoritative source had been received with regard to a statement made by the author in a former publication² to the effect that in the production of tar from by-product coke ovens "*carbonization is conducted at a lower temperature than in the manufacture of coal gas.*" The resulting tar, therefore, contains a smaller amount of free carbon, averaging from 3 to 10 per cent * * *." But little reliable information on this subject could be obtained from published literature, although the opinion seemed to prevail that carbonization in by-product coke ovens is conducted at a lower temperature than in modern gas-house practice. Thus, according to Lunge,³ "Hilgenstock (J. Gasbeleucht., 1902, 617) attributes the notorious difference between gas-tar and coke-oven tar with respect of their contents of free carbon and other products of pyrogenetic decomposition to the fact that in coke ovens the escaping vapors do not attain temperatures above 600°C., and that they are, moreover, protected against decomposition by the dilution of the heavy vapors, slowly given off from the

¹ Mineral Resources of the United States, 1909, Part II, pp. 210-242, United States Geological Survey.

² Circular 93, Office of Public Roads, U. S. Department of Agriculture, p. 9.

³ Coal Tar and Ammonia, 4th ed., part 1, p. 23, Van Nostrand.

TABLE I.—Analyses of crude coke-oven tars

| Serial No. | General information. | | Answers to questions. | | | |
|------------|---------------------------------------------------------------------|-------------------------------|--------------------------------------------------|--------------------------------------------------|----------------------------------------|---------------------------------|
| | Company and location. | Type of oven. | Maximum temperature of firing retorts. | Maximum temperature to which coal is brought. | Specific gravity of crude tar. | Per cent of free carbon in tar. |
| 5126 | Solvay Process Co., Syracuse, N. Y. | Semet-Solvay. | 1050-1450° C. | 950-1150° C. | 1.12-1.21 | 3-12 |
| 5123 | Semet-Solvay Co., Pennsylvania Steel Co., Steelton, Pa. |do..... | 1050-1450° C. | 950-1150° C. | 1.12-1.21 | 3-12 |
| 5124 | Semet-Solvay Co., National Tube Co., Benwood, W. Va. |do..... | 1050-1450° C. | 950-1150° C. | 1.12-1.21 | 3-12 |
| 5137 | Semet-Solvay Co., Milwaukee Coke & Gas Co., Milwaukee, Wis. |do..... | 1050-1450° C. | 950-1150° C. | 1.12-1.21 | 3-12 |
| 5121 | Semet-Solvay Co., Pennsylvania Steel Co., Lebanon, Pa. |do..... | 1050-1450° C. | 950-1150° C. | 1.12-1.21 | 3-12 |
| 5125 | By-Products Coke Corporation, South Chicago, Ill. |do..... | 1050-1450° C. | 950-1150° C. | 1.12-1.21 | 3-12 |
| 5128 | Semet-Solvay Co., Detroit, Mich. |do..... | 1050-1450° C. | 950-1150° C. | 1.12-1.21 | 3-12 |
| 5200 | Semet-Solvay Co., Empire Coke Co., Geneva, N. Y. |do..... | 1050-1450° C. | 950-1150° C. | 1.12-1.21 | 3-12 |
| 5189 | Semet-Solvay Co., Dunbar Furnace Co., Dunbar, Pa. |do..... | 1050-1450° C. | 950-1150° C. | 1.12-1.21 | 3-12 |
| 5150 | Semet-Solvay Co., Central Iron & Coal Co., Tuscaloosa, Ala. |do..... | 1250° C. | 1150° C. | 1.17 | 5.72 |
| 5074 | Philadelphia Suburban Gas & Electric Co., Chester, Pa. |do..... | 1050° C. | 1000° C. | 1.16 (20° C.) | |
| 5081 | Semet-Solvay Co., Ensley, Ala. |do..... | 1250° C. | 1150° C. | 1.17 (15° C.) | 8 |
| 5095 | The New England Gas & Coke Co., Everett, Mass. | Otto Hoffman. | 1100° C. | 1200° C. | 1.17 | 8-10 |
| 5083 | Lackawanna Steel Co., Lackawanna Iron & Steel Co., Lebanon, Pa. |do..... | 1000° C. (1800° F.) | 1000° C. (1800° F.) | 1.10 | 16-24 |
| 5159 | Dominion Tar & Chemical Co., Sydney, Nova Scotia. |do..... | (?) | (?) | 1.170 | 10-15 |
| 5107 | Hamilton Otto Coke Co., Hamilton, Ohio. |do..... | 1111° C. (2000° F.) | 1111° C. (2000° F.) | 1.14 | 6 16.6 |
| 5086 | Carnegie Steel Co., South Sharon, Pa. | United Otto. | 1666° C. (3000° F.) | 1444° C. (2600° F.) | 1.2 | 7.09-10.64 |
| 5078 | Maryland Steel Co., Sparrows Point, Md. |do..... | 1333° C. (2400° F.) | 1222° C. (2200° F.) | 1.19 | 8-10 |
| 5087 | Citizens' Gas Co., Indianapolis, Ind. |do..... | 1222° C. (2200° F.) | 1222° C. (2200° F.) | 1.14-1.15 | 4-5 |
| 5109 | Pittsburg Gas & Coke Co., The United Coke & Gas Co., Glassport, Pa. |do..... | (?) | (?) | 1.207 (50° F.) 1.169 (10° C.) | 16.59 |
| 5122 | Zenith Furnace Co., Duluth, Minn. |do..... | 1222-1277° C. 2200 - 2300° (F.) | (?) | (?) | (?) |
| 5188 | Illinois Steel Co., Joliet, Ill. | Koppers. | 1444° C. (2600° F.) | 1388° C. (2500° F.) | 1.16-1.20 | 12-15 |
| 5404 | Illinois Steel Co., Indiana Steel Co., Gary, Ind. |do..... | 1100° C. | 880 - 950° C. | 1.174 1.169 | 4.35 |
| 5108 | Camden Coke Co., Camden, N. J. | Otto Hoffman. United Otto. | 1000° C. (1800° F.) 1222° C. (2200° F.) | 833° C. (1500° F.) 1055° C. (1900° F.) | 1.20-1.30 (1.221) | 7-9 (7.3) |
| 5127 | Cambria Steel Co., Johnstown, Pa. | Otto Hoffman. United Otto. | 1111° C. (2000° F.) 1111° C. (2000° F.) | 1111° C. (2000° F.) 1111° C. (2000° F.) | 1.12 | 15 |
| 5089 | Lackawanna Steel Co., Buffalo, N. Y. | United Otto. Rothberg. | 1000° C. (1800° F.) 1000° C. (1800° F.) | 1000° C. (1800° F.) 1000° C. (1800° F.) | 1.16 | 16-24 |

¹ Approximately.² No information.³ Varies with coal. Coal with 28 per cent of volatile matter used.⁴ With H₂O.⁵ At present.⁶ Variable.⁷ Trace.⁸ Trace of solids.⁹ Distillate, solid.¹⁰ Distillate, one-fourth solid.¹¹ Distillate, nine-tenths solid.¹² Distillate, three-fourths solid.¹³ Distillate, eight-ninths solid.¹⁴ Distillate, one-half solid.

produced in the United States and Canada.

| Examination, Office of Public Roads. | | | | | | | | | | | | | | Serial No. | | |
|--------------------------------------|--------------------------|------------------|-------------------------------------------------------------------|-----------------------|---------------------|--------------------------|---------------------|---------------------------|---------------------|--------------------------|---------------------|--------------------------|---------------------|---------------------|---------------------|------|
| Specific gravity of tar, 25° C. | Per cent of free carbon. | Per cent of ash. | Per cent soluble in CS ₂ , including H ₂ O. | Distillation results. | | | | | | | | | | | | |
| | | | | Water. | | Light oils up to 110° C. | | Middle oils, 110°-170° C. | | Heavy oils, 170°-270° C. | | Heavy oils, 270°-315° C. | | | Pitch. | |
| | | | | Per cent by volume. | Per cent by weight. | Per cent by volume. | Per cent by weight. | Per cent by volume. | Per cent by weight. | Per cent by volume. | Per cent by weight. | Per cent by volume. | Per cent by weight. | Per cent by volume. | Per cent by weight. | |
| 1.195 | 7.76 | 0.12 | 92.12 | 1.0 | 0.8 | ⁸ 0.3 | 0.3 | 0.8 | 0.7 | ¹² 13.1 | 11.5 | ¹⁹ 8.2 | 7.3 | ²⁵ 76.6 | 79.1 | 5126 |
| 1.206 | 8.77 | .07 | 91.16 | 1.0 | .8 | .4 | .3 | ⁹ 2.0 | 1.7 | ⁹ 14.0 | 12.3 | ²⁰ 7.9 | 6.9 | ²⁶ 74.7 | 77.6 | 5123 |
| 1.176 | 7.14 | .04 | 92.82 | 1.1 | 1.0 | 1.9 | 1.5 | .7 | .6 | 14.9 | 13.2 | ²¹ 11.9 | 10.6 | ²⁷ 69.5 | 73.1 | 5124 |
| 1.168 | 6.10 | .05 | 93.85 | 1.8 | 1.5 | 1.4 | 1.2 | .8 | .6 | ¹³ 21.1 | 18.9 | ²⁰ 5.5 | 4.9 | ²⁵ 69.4 | 72.5 | 5137 |
| 1.173 | 4.71 | .06 | 95.23 | .6 | .5 | 1.6 | 1.3 | .8 | .6 | ¹⁴ 17.5 | 15.5 | ¹⁹ 9.4 | 8.4 | ²⁵ 70.1 | 73.7 | 5121 |
| 1.191 | 7.49 | .03 | 92.48 | (7) | (7) | .4 | .3 | ¹² 1.1 | .9 | ¹⁵ 23.6 | 20.7 | ⁹ 9.8 | 8.9 | ²⁷ 65.1 | 68.9 | 5125 |
| 1.169 | 6.56 | .11 | 93.33 | 6.9 | 5.9 | ⁹ 2.8 | 2.3 | ⁹ .4 | .3 | ¹¹ 14.6 | 13.0 | ⁸ 6.9 | 5.7 | ²⁶ 68.4 | 72.0 | 5128 |
| 1.159 | 6.07 | .08 | 93.85 | 4.0 | 3.4 | 2.6 | 2.1 | .6 | .5 | ¹⁰ 17.6 | 15.5 | ²² 11.4 | 10.4 | ²⁷ 63.8 | 67.7 | 5200 |
| 1.181 | 8.85 | .02 | 91.13 | 2.0 | 1.7 | 1.7 | 1.4 | .2 | .2 | ¹⁶ 20.0 | 17.8 | ²¹ 6.5 | 5.7 | ²⁵ 69.6 | 73.1 | 5189 |
| 1.159 | 5.05 | .02 | 94.93 | 3.2 | 2.8 | 2.4 | 1.9 | .3 | .3 | 18.6 | 16.3 | ¹⁹ 7.5 | 6.8 | ²⁷ 68.0 | 71.5 | 5160 |
| 1.141 | 3.96 | .05 | 95.99 | 2.3 | 2.0 | 2.3 | 1.3 | 1.2 | .8 | 22.8 | 19.5 | ¹⁹ 13.6 | 12.5 | 57.8 | 62.6 | 5074 |
| 1.175 | 6.90 | .06 | 93.04 | 3.3 | 2.8 | ⁸ 1.4 | 1.0 | .2 | .2 | ¹⁷ 16.5 | 14.1 | ¹⁴ 9.3 | 8.2 | ²⁷ 69.3 | 73.2 | 5081 |
| 1.160 | 13.94 | .00 | 86.06 | 2.2 | 2.0 | 2.9 | 2.3 | .6 | .5 | 23.5 | 20.4 | ¹⁷ 15.6 | 14.4 | ²⁷ 55.2 | 59.7 | 5095 |
| 1.214 | 14.05 | .13 | 85.82 | 5.4 | 4.4 | ⁹ 1.4 | 1.4 | ⁹ .1 | .1 | ¹¹ 13.0 | 10.9 | ²¹ 9.4 | 8.1 | ²⁷ 70.7 | 74.6 | 5083 |
| 1.143 | 10.81 | .05 | 89.14 | 3.2 | 2.8 | 1.9 | 1.5 | .6 | .5 | 27.2 | 24.2 | ¹⁹ 7.3 | 6.7 | ²⁷ 59.8 | 63.5 | 5159 |
| 1.160 | 8.37 | .06 | 91.57 | 3.4 | 3.0 | 3.1 | 2.5 | .7 | .6 | 27.9 | 24.4 | ¹⁹ 3.8 | 3.5 | ²⁷ 61.1 | 64.9 | 5107 |
| 1.191 | 7.89 | .03 | 92.08 | 1.0 | 1.0 | ⁹ 1.6 | 1.2 | ⁹ .6 | .4 | ¹⁶ 12.1 | 10.2 | ¹⁹ 11.0 | 9.7 | ²⁸ 73.7 | 77.5 | 5086 |
| 1.179 | 8.49 | .03 | 91.48 | 1.6 | 1.3 | 1.3 | .9 | .6 | .4 | ¹² 17.2 | 15.1 | ²¹ 9.6 | 8.5 | ²⁸ 69.7 | 73.2 | 5078 |
| 1.133 | 5.21 | .07 | 94.72 | 1.2 | 1.1 | 1.1 | .9 | 1.4 | 1.3 | 23.9 | 21.4 | ¹⁰ 11.6 | 10.4 | ²⁷ 60.8 | 64.7 | 5087 |
| 1.176 | 10.53 | .04 | 89.43 | 1.1 | 1.0 | 1.1 | .9 | .5 | .4 | ¹⁸ 26.9 | 23.6 | ¹⁴ 6.9 | 6.3 | ²⁷ 63.5 | 67.6 | 5109 |
| 1.195 | 12.18 | .05 | 87.77 | 3.6 | 3.0 | 1.7 | 1.3 | .4 | .3 | ¹¹ 18.1 | 15.9 | ¹⁹ 12.5 | 11.1 | ²⁷ 63.7 | 67.8 | 5122 |
| 1.171 | 3.89 | .06 | 96.05 | 1.9 | 1.6 | ⁹ 1.7 | 1.2 | ⁹ .2 | .2 | ⁹ 20.0 | 18.0 | ¹¹ 13.4 | 12.0 | ²⁶ 62.8 | 66.3 | 5188 |
| 1.169 | 2.73 | .04 | 97.23 | 3.5 | 3.0 | ⁹ 1.3 | 1.0 | ⁹ .4 | .3 | ⁹ 20.6 | 18.5 | ⁹ 7.1 | 6.5 | ²⁶ 67.1 | 70.2 | 5404 |
| 1.182 | 11.30 | .06 | 88.64 | 2.2 | 1.9 | 1.8 | 1.4 | .6 | .5 | ¹⁴ 20.5 | 18.2 | ²³ 8.5 | 7.5 | ²⁵ 66.4 | 70.1 | 5108 |
| 1.211 | 12.40 | .16 | 87.44 | 10.1 | 8.3 | ⁹ 3.1 | 2.3 | ⁹ .3 | .2 | ⁹ 7.1 | 6.1 | ¹² 7.4 | 6.0 | ²⁶ 72.0 | 74.8 | 5127 |
| 1.210 | 16.80 | .00 | 83.20 | 2.7 | 2.2 | ¹⁰ .5 | .3 | ⁹ 2.2 | 1.7 | ⁹ 11.7 | 9.9 | ²³ 11.8 | 10.2 | ²⁷ 71.1 | 75.0 | 5089 |

¹² Distillate, two-thirds solid.

¹⁶ Distillate, four-fifths solid.

¹⁷ Distillate, seven-eighths solid.

¹⁸ Distillate, one-ninth solid.

¹⁹ Distillate, one-third solid.

²⁰ Distillate, one-sixth solid.

²¹ Distillate, one-fifth solid.

²² Distillate, two-fifths solid.

²³ Distillate, one-seventh solid.

²⁴ Distillate, three-fifths solid.

²⁵ Pitch, soft and sticky.

²⁶ Pitch, very soft and sticky.

²⁷ Pitch, hard and brittle.

²⁸ Pitch, plastic.

interior of the coal block, with the fire-resisting gases escaping at the same time from the outer zone of the block. On the other hand, in the case of gas retorts, the heavy vapors escape at once undiluted by the fire-resisting gases which are set free afterwards, and the heavy gases are thus exposed to the white heat of the upper part of the retort."

Answers given by the manufacturers with respect to this question indicate only an approximate knowledge. In general it may be said that carbonization below 970° C. is considered low temperature; from 970° C. to $1,100^{\circ}$ C., medium temperature; and from $1,100^{\circ}$ C. to $1,540^{\circ}$ C. high temperature; and that modern gas-house practice involves the use of high temperatures. However this may be, it is not the purpose of this circular to compare coke-oven tars with gas-house tars, but to consider the former with relation to their utility as road materials.

From a total of 31 manufacturers to whom the questions were submitted 30 replies were received, but 4 of these reported their plants as not in operation. The remaining 26 furnished samples of their crude tar for examination and answered the questions in so far as they were able. Upon receipt of each sample the entire contents of the package were thoroughly mixed and a representative sample taken for analysis. The results of these analyses, together with the information furnished by the manufacturers, are given in Table I. In this table the different tars are grouped according to the type of oven in which they were produced.

In columns 4 and 5 all temperatures are expressed in degrees Centigrade, although where the manufacturers gave the temperatures in degrees Fahrenheit their statements are shown in parentheses. In column 5 it will be noticed that statements relative to the maximum temperature to which the coal is brought during distillation indicate that two of the plants run below 970° C., that a total of 22 run not over $1,150^{\circ}$ C., that 9 run from 950° C. to $1,150^{\circ}$ C., and that only 5 run above $1,150^{\circ}$ C. The maximum temperature of firing the retorts is, however, reported in most cases as being higher than the maximum temperature to which the coal is brought.

The maximum percentage of free carbon reported is from 16 to 24 per cent, but 17 manufacturers reported the maximum percentage of free carbon as being 12 per cent or under, and only 4 as 16 per cent or over.

Analyses of the samples received were made in accordance with the methods described in a former publication¹ of the office. The work consisted in determining the specific gravity, free carbon, or organic matter insoluble in *c. p.* carbon disulphide upon a 15-minute digestion at room temperature, material soluble in carbon disulphide, percentage of ash, and percentage of different fractions obtained by distilling a 250 *c. c.* sample in a 750 *c. c.* tubulated glass retort with the ther-

¹ Bulletin 38, Office of Public Roads, U. S. Department of Agriculture.

nometer so placed that the top of the bulb was level with the bottom of the juncture of the stem and body of the retort.

It will be noted that the gravities of the samples examined range from 1.133 to 1.214 and that the great majority are lower than 1.200. This in itself indicates low percentages of free carbon. The minimum percentage of free carbon was 2.73, the maximum 16.80, and the average for the 26 samples 8.38. Eighteen samples contained less than 10 per cent of free carbon and 8 more than 10 per cent. About two-thirds of these products might, therefore, be considered as low-carbon tars and the other third as medium-carbon tars. The amount of ash in no case exceeded 0.16 per cent, and in most cases it was practically nil. This is, of course, also true of practically all gas-house coal tars. The percentage of water present varied from a trace to 10.1 per cent by volume, but in only 3 instances did it exceed 5 per cent. Water is a variable, depending upon a number of conditions, and, as it is not a part of the true tar, has been eliminated in Table II. Before leaving Table I, however, it is of interest to note that 14 of the pitch residues, remaining after distillation had been carried to 315° C., were either soft or plastic—a condition which has seldom been noticed by the author in the distillation of gas-house coal tars. The amount of solids which crystallized or precipitated out of the different fractions was found to vary greatly, as shown in the foot-notes to Table I.

TABLE II.—Analysis of coke-oven tars upon a water-free basis.

| Serial number. | Type of oven. | Percentage of free carbon. | Fractions by weight. | | | | |
|----------------|------------------------------|----------------------------|-------------------------|----------------------------|----------------------------|----------------------------|----------------------|
| | | | Percentage up to 110°C. | Percentage from 110-170°C. | Percentage from 170-270°C. | Percentage from 270-315°C. | Percentage of pitch. |
| 5126 | Semet-Solvay | 7.82 | 0.30 | 0.70 | 11.59 | 7.35 | 79.73 |
| 5123 | do. | 8.84 | .30 | 1.71 | 12.39 | 6.95 | 78.22 |
| 5124 | do. | 7.21 | 1.55 | .60 | 13.33 | 10.70 | 73.83 |
| 5137 | do. | 6.19 | 1.21 | .60 | 19.18 | 4.97 | 73.69 |
| 5121 | do. | 4.73 | 1.30 | .60 | 15.57 | 8.44 | 74.07 |
| 5125 | do. | 7.49 | .30 | .90 | 20.70 | 8.90 | 68.90 |
| 5128 | do. | 6.97 | 2.44 | .31 | 13.81 | 6.05 | 76.51 |
| 6200 | do. | 6.28 | 2.17 | .51 | 16.04 | 10.76 | 70.08 |
| 5189 | do. | 9.00 | 1.42 | .20 | 18.10 | 5.79 | 74.39 |
| 6160 | do. | 5.19 | 1.95 | .30 | 16.76 | 6.99 | 73.55 |
| 5074 | do. | 4.04 | 1.32 | .81 | 19.89 | 12.75 | 63.87 |
| 5081 | do. | 7.09 | 1.02 | .20 | 14.50 | 8.43 | 75.30 |
| 5995 | Otto Hoffman | 14.22 | 2.34 | .51 | 20.81 | 14.06 | 60.91 |
| 5083 | do. | 14.69 | 1.46 | .10 | 11.40 | 8.47 | 78.46 |
| 5159 | do. | 11.12 | 1.54 | .51 | 24.89 | 6.59 | 65.32 |
| 5107 | do. | 8.62 | 2.57 | .61 | 25.15 | 3.60 | 66.90 |
| 5086 | United Otto | 7.96 | 1.21 | .40 | 15.29 | 9.79 | 78.28 |
| 5078 | do. | 8.60 | .91 | .40 | 19.30 | 8.43 | 74.16 |
| 5087 | do. | 5.26 | .91 | 1.31 | 21.63 | 10.51 | 65.41 |
| 5109 | do. | 10.63 | .90 | .40 | 23.83 | 6.36 | 68.28 |
| 5122 | do. | 12.55 | 1.34 | .30 | 16.39 | 11.44 | 60.80 |
| 5188 | Koppers | 3.95 | 1.21 | .20 | 18.29 | 12.19 | 67.37 |
| 5401 | do. | 2.81 | 1.03 | .30 | 19.07 | 6.70 | 72.37 |
| 5108 | Otto Hoffman and United Otto | 11.51 | 1.42 | .60 | 18.55 | 7.64 | 71.45 |
| 5127 | do. | 13.52 | 2.50 | .21 | 6.65 | 7.52 | 81.57 |
| 5089 | United Otto and Rothberg | 17.17 | .30 | 1.73 | 10.12 | 10.42 | 76.68 |

The results given in Table II are calculated upon a water-free basis—i. e., the percentages are expressed in terms of the actual tar exclusive of water. Considering these products according to type,

it will be seen that the tar produced by the Koppers ovens contains the lowest percentage of free carbon, the Semet-Solvay tars the next lowest, the United Otto next, the Otto Hoffman next, and the mixed tar from the United Otto and Rothberg ovens contains the highest percentage of free carbon. For the sake of comparison the minimum, maximum, and average percentages of free carbon for each of these types are shown in Table III.

TABLE III.—Percentage of free carbon in coke-oven tars.

[Water-free basis.]

| Type of oven. | Percentage of free carbon. | | |
|-------------------------------------------|----------------------------|----------|----------|
| | Minimum. | Maximum. | Average. |
| Koppers..... | 2.81 | 3.95 | 3.38 |
| Semet-Solvay..... | 4.04 | 9.00 | 6.74 |
| United Otto..... | 5.26 | 12.55 | 9.00 |
| Otto Hoffman..... | 8.62 | 14.69 | 12.16 |
| Otto Hoffman and United Otto (mixed)..... | 11.51 | 13.52 | 12.51 |
| United Otto and Rothberg (mixed)..... | 17.17 | 17.17 | 17.17 |

The percentages of various fractions for the different types of tars overlap to such an extent that no detailed comparison will be made. The maximum, minimum, and average total distillates to 315° C. for the different types are, however, given in Table IV.

TABLE IV.—Percentage by volume of total distillate to 315° C. in coke-oven tars.

[Water-free basis.]

| Type of oven. | Percentage by volume. | | |
|-------------------------------------------|-----------------------|----------|----------|
| | Minimum. | Maximum. | Average. |
| Koppers..... | 30.5 | 36.0 | 33.3 |
| Semet-Solvay..... | 22.6 | 40.8 | 29.9 |
| United Otto..... | 25.6 | 38.5 | 32.6 |
| Otto Hoffman..... | 25.3 | 43.6 | 36.0 |
| Otto Hoffman and United Otto (mixed)..... | 19.9 | 32.1 | 26.0 |
| United Otto and Rothberg (mixed)..... | 26.9 | 26.9 | 26.9 |

From this Table it is evident that wide variations exist in the relation of total distillate to pitch residue in the coke-oven tars produced in this country, and this is even true of different tars produced by the same type of oven.

Straight coal-tar road binders or refined coal tars are usually manufactured by subjecting the crude material to a process of distillation with or without steam or air agitation. Distillation is carried to the point at which the residuum remaining in the still has obtained the desired consistency at normal temperatures, and this involves the removal of certain of the more volatile oils present in the crude material. For use in construction work a soft and almost fluid pitch is often produced, and the consistency of this pitch is

controlled by means of a melting point or float test. When the crude tar runs abnormally high in free carbon, it is sometimes mixed with crude water-gas tar before distillation. Water-gas tar contains a very low percentage of free carbon, and by properly proportioning the two a product is obtained, upon distillation, which does not carry more than the maximum limit of free carbon set by manufacturers. What the maximum limit should be is a much mooted question among those who have given thought to this matter. The governing considerations are: (1) What is the most economical limit from the standpoint of manufacture? and (2) What is the proper limit with regard to the utilization of the product as a road material? For a number of reasons, which it is unnecessary to mention in this circular, an excessively high-carbon tar is difficult to distill properly and, with other things equal, the lower the percentage of carbon the easier and shorter the distilling process. From this standpoint, therefore, by-product coke-oven tars are well adapted to the manufacture of road binders. Moreover, because of their low percentage of free carbon, they may be employed in a manner similar to water-gas tars, when it is desired to utilize a crude high-carbon tar in the production of a medium-carbon tar road binder.

In an ordinary road tar for use in construction work where free carbon is present to the extent of about 20 per cent, the proportion of total distillate, below 315° C., to pitch residue is approximately 1 to 4. Where this relation exists the pitch residue is hard and brittle. A residue which is soft or plastic is to be preferred, as it would indicate longer life during service, and where such a residue is present the proportion of distillate would naturally be lower for a given consistency, as the distillates may be considered as fluxes for the residues. If such is the case, it is evident from the foregoing tables that coke-oven tars offer a valuable source of supply for tar road binders. As an example, even the highest-carbon tar, No. 5089, if distilled to the point where the proportion of distillate, below 315° C., to the pitch residue was as 1 to 4, would contain less than 19 per cent of free carbon, which is at present considered as not excessive for a refined coal tar.

In conclusion it may be said that indications point strongly to the fact that by-product coke ovens will eventually play a most important part in the road-material industry, and it is to be hoped that their general adoption in this country will be rapid. The future demand for economical bituminous road binders in the United States will undoubtedly exceed the supply, and this in spite of the natural increase in petroleum and asphalt road binders. If such is the case, the present loss of enormous quantities of tar, to say nothing of gas and ammonia, because of the use of beehive ovens, is a matter worthy of the utmost consideration on the part of all who are interested in the conservation of our resources.

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