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# Proceedings of the 23rd Forum on the Geology of Industrial Minerals

Randall E. Hughes and James C. Bradbury, editors



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ILLINOIS STATE GEOLOGICAL SURVEY  
May 11-15, 1987  
North Aurora, Illinois

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Cover photo: State-of-the-art mining at the Ottawa Silica Company Quarry in July 1940. Workers used scrapers to remove thin overburden down to the sandstone, then shoveled out (by hand) the loose overburden remaining in depressions and loaded it onto this brand-new dump-truck. The final cleaning was done with stiff brushes and compressed air or steam. The men (left center) are drilling a blast-hole on the cleaned sandstone surface; in the background is the processing plant.

*Graphics: Pamela Foster*

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# **Proceedings of the 23rd Forum on the Geology of Industrial Minerals**

**Randall E. Hughes and James C. Bradbury, editors**

**May 11-15, 1987  
North Aurora, Illinois**

**ILLINOIS STATE GEOLOGICAL SURVEY  
Morris W. Leighton, Chief**


**Natural Resources Building  
615 East Peabody Drive  
Champaign, IL 61820**

#### **FORUM DEDICATION**

Dr. Ralph E. Grim, the keynote author in this proceedings, died August 19, 1989, shortly after returning from a consulting trip. Because of his lifelong interest in industrial minerals and the help and guidance he gave to many of us, we dedicate this volume to Dr. Grim's memory.

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# Keynote Address: Industrial Minerals Research

*Ralph Grim, Professor Emeritus  
Department of Geology  
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*Our keynote speaker, Ralph Grim, is a father of clay mineralogy. His career has been marked with outstanding scientific achievements and service to the State of Illinois, the University of Illinois, and the national and international scientific and industrial communities. Dr. Grim earned his bachelor's degree from Yale University and his doctorate from the University of Iowa. He began his career in geology more than 60 years ago as Assistant Professor at the University of Mississippi when he was also Assistant State Geologist of Mississippi. Dr. Grim joined the Illinois State Geology Survey (ISGS) in 1931 as a petrographer and became widely known for his work on clays and clay minerals. He became Principal Geologist of the ISGS in 1945, and from 1948 to 1967 was a research professor of geology at the University of Illinois. Dr. Grim has been widely recognized. Among his honors are the Gold Medal and honorary membership in the Clay Minerals Society of Spain in 1972; the Medal of the National Order and designation as a Chevalier of the National Order of the Republic of the Ivory Coast in 1973; the prestigious Roebling Medal of the Mineralogical Society of America in 1974, the highest honor that can be bestowed by that society in his field; and in 1984, an honorary doctoral degree from the University of Illinois. He continues to be active in scientific circles, and has published more than 120 articles and three books. Dr. Grim doesn't know this, but many of his books were my primers when I began my research work in the X-ray diffraction laboratory of the Carter Oil Research Company in Tulsa, Oklahoma. His life has touched many of us in many ways for many years.*

*Morris W. Leighton, Chief  
Illinois State Geological Survey*

## INTRODUCTION

Many industrial minerals such as sands and gravels require very little preparation before they are used commercially. Some require a moderate amount of preparation—perhaps grinding, perhaps a little purification, perhaps classification. Others profit from substantial preparation before they are put on the market or used, and still others are the raw materials for a variety of manufacturing processes. This paper is concerned with research to improve methods of preparing industrial minerals for use and to extend and improve products made from them.

## BENTONITE

Bentonite is a peculiar kind of clay composed entirely of montmorillonite, a member of the smectite group of clay minerals. In the United States, bentonite is produced mostly in Wyoming but also

in the Gulf Coast areas of Texas, Mississippi, and Alabama and in the Rocky Mountain states. The development of the bentonite industry is an American event. Bentonite was recognized and developed for industrial use here years before Europeans and others even realized that it existed. In England, bentonite was produced as fuller's earth for many years; however, it was the Americans who recognized the equivalence of bentonite and fuller's earth.

Bentonite has unique properties: it swells in water, produces gel-type structures, and has high adsorptive capacity. It can adsorb various organic and inorganic ions that are commonly replaceable one for another. It is exceedingly plastic. The properties of bentonite obtained from different places can vary widely.

## Development of bentonites

The development of bentonites in this country was primarily the result of the efforts of two men. In the 1920s and 1930s George Radcliffe, head of the Baroid Corporation (now part of the NL Industries), realized, with the development of rotary drilling for oil in the Gulf Coast region, that drilling muds were essential in that process. He also realized that bentonites could produce superb drilling fluids, and he developed that business. Bentonites set into a gel when drilling is stopped and tend to hold cuttings in suspension so they do not drop to the bottom and freeze the drill stem. Bentonites also tend to produce an impervious wall on the inside of the drillhole.

Much research has since been done on the use of bentonites in drilling fluids. One interesting problem is that when you drill a well through clays and shales you unavoidably grind up a lot of that clay and shale and add it to the mud. The mud becomes increasingly viscous—so viscous that it has to be modified. Obviously, you could dilute the mud with water to reduce the viscosity, but this would reduce the weight of the mud, which is not desirable. Research has been conducted on how to control the viscosity of the mud. Various chemicals, particularly certain phosphates, have proven valuable for this purpose.

Paul Bechner, head of the American Colloid Company in Chicago for many years, also helped to develop the bentonite industry. During the 1920s and 1930s, when foundries become mechanized and shifted over to synthetic molding sands, he appreciated the possibility of using bentonite as a bonding agent for synthetic molding sands.

## Properties and uses of bentonites

One of the factors that causes variation in the properties of bentonite from place to place is the particular cation that is adsorbed by the bentonite. A bentonite that is carrying sodium will have somewhat

different properties from a bentonite carrying calcium. The properties preferred for drilling and bonding clays are commonly developed in the sodium bentonites rather than in the calcium bentonites. Bentonite producers worldwide add soda ash to bentonites to change their characteristics and control their properties. A good deal of research has gone into developing ways to add soda ash to get maximum improvement.

Some bentonites decolorize oil and thus can be used as fuller's earth—as is one of the English bentonites. By treating certain bentonites with acids you can enhance the decolorizing property and you can manipulate this process, by varying time, temperature, and acid concentrations, to produce adsorbents that are specific for a particular kind of oil (cottonseed oil, for instance).

**Catalytic properties** In some cases, bentonites have catalytic properties—the ability to change the character of organic molecules because of their reaction with the montmorillonite surface. The process for producing gasoline by refining crude oil changed around the 1920s from distillation processes to the use of catalysts. The early catalysts used in the petroleum industry were synthetic granular materials (a little coarser than sugar or small pellets) such as silica compounds with magnesium. When the catalysts are mixed with the feed stock and heated to a temperature of a couple of hundred degrees centigrade, they break down the feed stock into gasolines and into carbon that is coated on the catalysts.

A successful catalyst must be hard. It must be able to convert a high percentage of the feed stock to gasoline, and the gasoline formed must have an acceptable octane number. A great deal of research has been done on developing catalysts for the petroleum industry that can achieve these desired results.

In the 1940s, around the time of World War II, Wright Gary, who was head of the Filtrol Corporation in Los Angeles, conceived of the idea that bentonites might be used to make a satisfactory catalyst for producing aviation gasoline. After about two years of research, he succeeded in producing the first satisfactory catalyst from clay material.

Since that time, catalysts for the petroleum industry have been produced by treating other clay minerals. Plants in Georgia are producing catalysts for the

petroleum industry from kaolinite, and recently an excellent catalyst was made by a totally different process in which dehydroxylated kaolin is treated with caustic soda to create a zeolite-type structure having superb catalytic properties. This process results in a gasoline with an exceedingly high octane number.

**Reactions with organics** In the early 1940s there was a furious effort on the part of the Army Air Corps to find some way to quickly solidify soil, so that if its men captured a Pacific island they could go in and harden the soil quickly in order to land planes on it. This research involved some work with organic materials and soil, much of which was done at the Massachusetts Institute of Technology (MIT) by Ernest Hauser.

About 1942, George Radcliffe set up a fellowship at Mellon Institute to investigate other potential uses of bentonite. He chose a group of people to select the topic of research and advise the fellow. The research topic this group selected was the reaction of bentonite with various organic compounds. Soil chemists, agronomists, and others were particularly interested in the reaction of organics in soils with clay materials.

The fellow chosen, John W. "Spike" Jordan, was a bright, young organic chemist. He went to work at Mellon and in about three or four years came up with what now are known as "bentones." Bentones are organically treated bentonites in which the individual layer components of the montmorillonite are coated with a thin sheet of organic molecules. The character of the bentonite is thus changed; instead of being water absorbent, it becomes water repellent and adsorbs and gels many organic liquids. Now produced commercially, bentones are used in preparing lubricating greases (in place of soaps) and in many other ways. The work by Spike Jordan opened the door for research into the reaction of all sorts of organic materials with all of the various types of clay minerals.

**Bentonite sealers** Bentonite has the power of fixing many organic and inorganic materials; it also can form a very impervious seal. For example, if you spread a little bentonite along a drainage ditch it becomes impervious to water. I have prevented water from leaking into our house by spreading a layer of bentonite all around the outside wall.

The use of bentonites in the disposal of high-level radioactive wastes is a current interest of mine and many others as well. High-level radioactive wastes and some other materials can probably be disposed of by calcining the waste material with a flux to form a slag in which the hot isotopes are locked up in a relatively insoluble form. The slag is placed in conduits or pipes about a foot in diameter and about 10 feet long. These pipes are then carried to an underground facility, where holes are drilled to a diameter of about 30 inches. The pipes are lowered into the holes and completely surrounded by bentonite, with bentonite beneath them and bentonite on top of them. The bentonite forms an impervious seal around the pipes; also, it will adsorb the isotopes in the event that the pipes should be rusted through or broken, preventing the isotopes from escaping into the groundwater system.

Bentonite probably will not be used in its pure form in such an operation but will be mixed with zeolites, another type of mineral with high capacity for fixation of isotopes. Some ceramic research laboratories have recently developed synthetic materials that have a very high potency for the adsorption and fixation of atomic wastes. These would also be added to the bentonite, probably to form about a 50:50 mixture; the bentonites would form an impervious seal and the other materials added would aid in trapping any isotopes that might escape.

The Swedish government is currently using this method of disposal of atomic waste on a pilot-plant scale. Sweden is intensely interested in atomic energy because it has no coal or oil and has used all of its potential sites for hydroelectric energy. Except for atomic energy, Sweden has few energy sources for making electricity. The Swedes are now investigating the use of bentonites in radioactive waste disposal in an abandoned iron mine located within a mass of granite at a depth of about 1500 feet. This activity is sponsored not only by the Swedish government, but by our own government as well. I have been in the mine, and it is quite a place. One of the largest computers you could imagine is in the mine, 1500 feet underground. The laboratory work for the procedure of atomic waste disposal is being done at Luleo, just south of the Arctic Circle, at a



branch of the University of Sweden and the Swedish Geological Survey.

## KAOLINS

Kaolins are clays composed essentially of the clay mineral kaolinite. They are used in the manufacture of various ceramic products, refractories, chinaware, sanitaryware, and also paper used as fillers and coating agents. It comes as a real surprise to many people to learn that a sheet of paper like that used in *National Geographic* is more than half clay.

In Georgia, many deposits of kaolin are found in an area stretching from near Macon on the southwest to about the South Carolina line on the northeast. They are lenticular masses, 15 feet thick or more, composed essentially of pure kaolinite and commonly found in sandstone. Georgia kaolinite is unique worldwide in that it contains no fine quartz particles—no grit that cannot be easily removed. Therein lies its special value. Hundreds of thousands of tons of kaolin are exported every year from the deposits in Georgia to Germany, Japan, Brazil, and elsewhere because of its unique characteristics. It costs too much to produce a completely grit-free kaolin from deposits in many other parts of the world, and paper manufacturers won't have anything to do with a clay that's at all gritty. If you run an X-ray diffraction pattern on a sample of kaolin and there's even the very slightest indication that quartz is in it, that kaolin isn't any good for the paper industry. The grit tends to wear out the fourdrinier wire in the paper-making machines. It also tends to produce an abrasive surface on the sheet of paper, which tends to wear out the type face of the offset printing masters.

Two varieties of kaolins are found in Georgia. In kaolins of Cretaceous age, the kaolinite is present in sizable vermicules or "books" in crystals that you can sometimes see with a hand lens. The other kaolins, of Tertiary age, are completely different. They are composed of extremely thin, small flakes about a micrometer or so in size that are packed together, one on top of the other, in a kind of random fashion. They break down only to aggregates, not to individual flakes.

Material used for coating paper should contain some relatively large flakes to heal the openings in the fiber that makes up the base of the paper. For some types of printing (rotogravure, for example) a

very smooth surface is required for acceptable printing. The Cretaceous kaolins are excellent for this. The preparation of the coating-grade product involves delaminating the kaolins—that is, they are split on their cleavages during the grinding process. A vast amount of research has gone into making big flakes from tiny flakes in the Tertiary kaolins so that these kaolins might also be used to make a satisfactory surface for book and rotogravure printing. Considerable success has been obtained by tacking these flakes together with an organic, sometimes using a big flake to act as a kind of core to hold the tiny flakes together.

There is far more to research on preparing clay for the paper industry than just removing the little bit of grit present in the kaolin. The goal of such research is to prepare a material that produces a printable surface with acceptable gloss—a sheet of paper that is very, very thin, yet opaque enough so that advertisers can't see a competitor's message through the sheet of paper they are reading.

One small area in Georgia contains some kaolins with smectite, a component in bentonites. How this came about, I don't know—the origin of the Tertiary kaolins is also a mystery to me. I can't understand how such extremely fine material could have been transported a sizable distance from any source area without somewhere along the line having picked up some grit that had been washed into the stream—so that grit was deposited with the very fine kaolin. Now, the origin of the Cretaceous kaolins is reasonably clear: they were transported only a very, very short distance from saprolites in the Piedmont, where alterations produced a material composed of extremely large crystals of kaolinite and only large particles of quartz.

## ZEOLITES

In recent years, there has been a great deal of interest in another type of industrial mineral that nobody had ever paid much attention to—the zeolites. In my days at the university, zeolites were a mineral curiosity. They occurred in very small amounts, sometimes in vugs in igneous rocks, but they were not particularly abundant and nobody considered that they might have commercial use. Recently, however, tremendous deposits of zeolites have been found in our western states. Some of these zeolites are

altered volcanic ashes; this poses the interesting problem of why the volcanic ash is altered to a zeolite rather than to smectite, a clay. The zeolites have a structure completely different from that of the clays. The silica tetrahedral units are tied together in a netlike structure that provides tremendous adsorption capacity. Some of the silicon is replaced by aluminum, which gives the zeolites a high cation exchange capacity. The zeolites have been used as molecular sieves to classify various materials. Zeolites can be made commercially, but, as I said, in recent years huge deposits of zeolites have been found.

Last year, in extreme western Idaho and extreme eastern Oregon, I spent some time looking at these zeolites. There's one deposit up there in which they put a drillhole down through about a hundred feet of solid clinoptilolite zeolite, and if you look at a thin section of this material under the microscope you can still see the structure of the parent ash. This zeolite clearly is altered volcanic ash.

Zeolites have interesting adsorption properties, as I mentioned earlier. They also have the ability to fix various isotopes, so it's possible that a natural or synthetic zeolite will be used as an ingredient in material used to form the seal around the atomic waste containers. Zeolites have other properties, and there's considerable research going on at the present time to discover potential uses for them. Zeolites are granular. They don't tend to mix with water, so they won't form a mud. They tend to fix potassium and various organic compounds, so they've been loaded with nitrogenous materials and potash and used to provide the plant food for plants growing in a tank of water. (Instead of putting your nutrients in solution into the tank, you adsorb them in the zeolites.) The zeolites slowly release the nutrients to the plants over a period of time. When the zeolites are exhausted, you take them out, replenish them, and use them again. Zeolites also can absorb a variety of odors and have been used as deodorants.

A man who lives near the Idaho deposit has taken upon himself the job of finding uses for these zeolites. He recently set up some research projects in the local high school in which a group of students investigated the potential uses of zeolites. One bright young woman got hold of some soil that contained a good

deal of phosphate, but the phosphorous was locked up in a calcium compound and was not readily available to the plants. So she mixed some zeolite with this particular soil and planted radishes and alfalfa in test plots. She reported that the zeolites tended to release the phosphate and that she got increased yields of radishes and alfalfa when the zeolites were added. Zeolites are an interesting resource with a promising future.

## GYPSUM

Another industrial mineral that's a favorite of mine is gypsum. Gypsum is white and occurs in flake-shaped units. Chemically it is hydrated calcium sulfate, currently used as a pigment in paints and in plasters. If you drive off about three-fourths of the water that's present, you get Plaster of Paris. Gypsum is tremendously abundant worldwide and its deposits are relatively pure.

About ten years ago, a very good friend of mine in England—a consultant for a company that produced industrial minerals and chemicals—convinced his company to investigate another possible use for gypsum. Two people in the research laboratory began to conduct some experiments to try to make a "bentone" material out of gypsum. I was pretty deeply involved in this and spent some time in their laboratory, and one of their scientists spent some weeks with me in my laboratory at the University of Illinois. I think we had achieved the intercalation of organics in gypsum. We had some X-ray diffraction data that indicated we were on the threshold of really working that out. Unfortunately, the company in England had financial difficulties and went broke, and the project was killed. I would hope that this type of research on gypsum will continue.

## CONSULTING

For many years I have been a consultant to a great many companies involved in industrial minerals. One of the problems in consulting is that you must get close to management in order to be able to do anything really important. Commonly the company that you're a consultant for is managed by lawyers or accountants or other people who have lit-

tle or no technical background. Consequently, they are reluctant to be frank about their problems, and you don't have a chance to contribute anything. I know of a couple of cases in which a company I was supposed to be a consultant for purchased deposits that I happened to know about. They lost a good deal of money attempting to develop these materials for which there was no hope. I didn't know about their plans to purchase the deposits or their efforts to develop them, and in the end I resigned from the company.

Now this lack of communication carries on down through a whole host of activities: tremendous problems are created and mistakes are made because often the people at the top don't know what to ask or who to ask. Here's just one example of this. For years, I was a consultant for the Bechtel Corporation in San Francisco, a huge engineering and construction outfit that builds everything from dams to atomic energy plants. The Bechtel Corporation has a very large, excellent geology department, and the investigation of the geology prior to construction activity is very thorough in the Bechtel group.

One of my early activities with the company concerned two atomic energy plants that were built in Spain near Barcelona. Bechtel didn't have anything to do with the construction of these plants—they were built by a Spanish public utility on the side of a valley above a sizable stream. They had chopped off the end of the broad ridge in the valley wall, cut it down, and leveled it off. On this newly leveled ground, they built two atomic energy plants. The bedrock in the area is a series of Cretaceous sands, clays, and gravels.

Just before the fuel was to be loaded into the first-completed plant the soil began to swell, and it pushed the plant up a measurable amount. The Spanish engineers called the Bechtel Corporation, wanting to know what caused the rise and what they could do about it. Well, the cause of it, as anybody with any knowledge about clay minerals would suspect right off, was that they had somehow changed the movement of groundwater when they leveled off this hill. You should never do this sort of thing if you're going to build a plant in such an

area. They changed the flow of water through bedrock material, and probably there was bentonite somewhere in these Cretaceous formations. (Cretaceous formations are notorious for the presence of bentonite and smectite types of materials.) Analysis showed that there was about 10 to 15 percent smectite in the materials at the site. So what do you do about it? In the first place, you make sure that you keep all the water away from the formation; there are also ways that a chemical ingredient can be put into the smectite material to help prevent the swelling by binding the particles together.

This procedure has been used very successfully in the construction of highways around Oslo in Norway. In the vicinity of Oslo and elsewhere in the world there are materials that engineers call "quick" soils. These are soils that have a high moisture content, and in an undisturbed condition they are solid and reasonably strong. But if they are agitated they flow like water. In the Oslo area, terraces along streams have suddenly liquefied and flowed catastrophically down into streams. Well, a very bright engineer in Oslo conceived of a way to stabilize these "quick" soils by treating them with various chemicals—and this treatment is so effective that it's been possible to build superhighways across these soils.

## RESEARCH OPPORTUNITIES

Much research remains to be done on improving industrial minerals and expanding their uses. There is tremendous opportunity for fundamental research in many areas. Gypsum, which is so abundant worldwide, is basically an untapped resource. Maybe it could be used to stabilize soft soils—it certainly has more uses than just plaster and pigment. Some of the applications for industrial minerals that I've touched on today are in their early stages—bentonites for radioactive waste disposal, kaolins in the printing industry, and zeolites for increased productivity in agriculture. The future looks promising to me for further research to improve the preparation and expand the uses of this important group of minerals.

# The Current State of Industrial Minerals in China

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## ABSTRACT

*China has a rich and varied mineral resource base and all the requisite conditions that tend to stimulate production and utilization of industrial minerals: a large, growing population, rapidly increasing living standards, an expanding industrial base, a growing need for export earnings, facilities for rail and water transport, and abundant low-cost labor.*

*Because of the variety of geologic terranes in China, virtually every known industrial mineral occurs in important quantities. Three ancient continental cratons are rimmed by plate-collision zones having metamorphic belts, ultramafic terranes, and igneous complexes; the cratons are locally covered by relatively undeformed clastic and evaporitic sedimentary rocks. Therefore, the geological environments that lead to the formation of a wide variety of industrial minerals are broadly distributed around the country. Although mineral production has so far been concentrated in the densely populated eastern half of China, the potential for future production is also high in western China, in spite of transportation limitations.*

*In addition to the huge quantities of construction materials used in China, many minerals of high unit-value—including asbestos, barite, diamond, fluorite, graphite, pyrophyllite, sillimanite, vermiculite, wollastonite, and minerals of beryllium, boron, and titanium—are of current or potential importance in world trade. China leads the world in production of rare earths and barite. Exports to the world are expected to increase as the immaturity of China's mineral resource database and transportation system is offset by a growing labor surplus and increasing need for foreign exchange to finance industrialization.*

*During the current period of rapid flux in China's economic policy, the foreigner interested in China's minerals faces a challenging business environment. Reorganization of central-government agencies into competitive, profit-motivated units has created rivalries that transcend the traditional commodity-bound relationships between organizations. At the same time, the economic autonomy of local governments has increased. Private enterprise, in the form of Chinese or foreign companies, may soon become an important factor in China's industrial minerals industry.*

## INTRODUCTION

China has been a supplier of industrial minerals to the world for centuries. Minerals such as borates from the salt lakes of western China and kaolin (named for Gaoling village in Jiangxi province) have long nourished an export trade for China. About 1273 Marco Polo described the mining and processing of asbestos in what is now Xinjiang province.

A number of factors have combined to create a thriving industrial minerals busi-

ness in China: a rich and varied resource base, a large and increasing population, a rising standard of living, abundant low-cost labor, increasing industrialization, and a growing need for export earnings. Although strained to capacity, rail and waterborne transport systems are adequate in eastern China.

During the past few years, Chinese exports of certain industrial minerals have had major impacts on Western producers and consumers. Negative impacts have

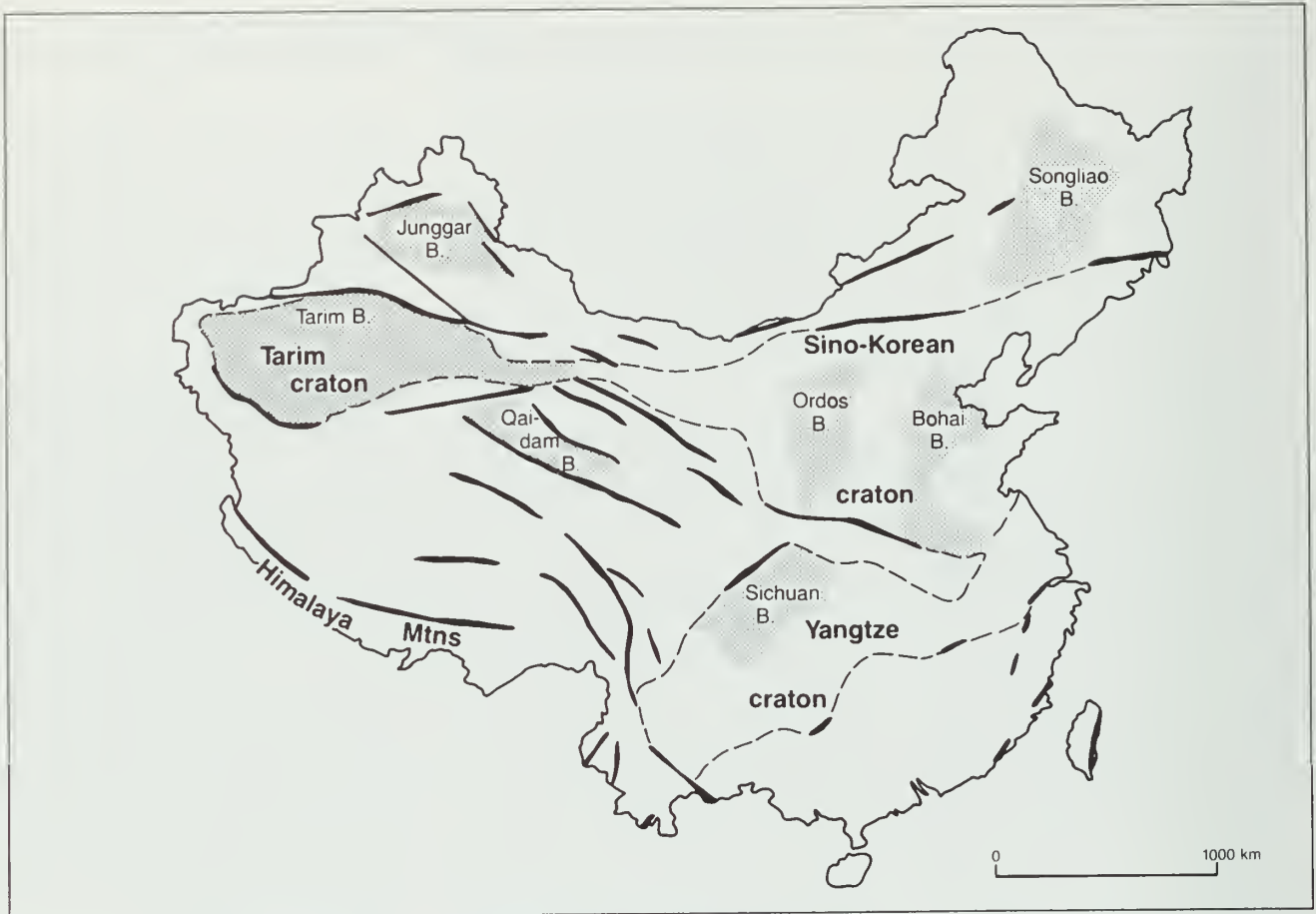
been felt by Western producers of barite, fluorite, and rare earths because of falling prices in the face of Chinese exports during times of weak demand. A positive side to the equation for western producers is that China is supplying some high-demand minerals (wollastonite, magnesite), and purchasing others (phosphates, potash, soda ash, diatomite).

This article provides a geological framework for China's industrial minerals resources, examines the organizational basis of the China's industrial minerals industry, and attempts to discern some trends for the future. Background information for this article was derived from the writer's geologic work, including examination of industrial minerals deposits, in several regions of China.

## GEOLOGIC SETTING

Within China's 9.5 million square kilometers (an area 2 percent larger than the United States), a wide variety of geologic conditions has resulted in environments suitable for the formation of nearly every industrial mineral commodity. Three ancient cratons—the Sino-Korean, the Yangtze, and the Tarim—are rimmed by plate-collision zones with metamorphic belts, ophiolites, and igneous complexes (fig. 1). A collision zone is still forming in the Himalayas. Sedimentary basins, some with evaporite facies, locally cover cratonic and intercratonic rocks. Sedimentation (mostly continental-evaporitic in the west and deltaic-marine in the east) continues in many parts of China.

Some degree of correspondence exists between the tectonic units shown on figure 1 and the distribution of certain minerals (fig. 2). Asbestos and chromite typically occur in ophiolites; diamonds on cratons; bentonite, boron, and potash in sedimentary basins; and rare earths, titanium, and perlite on the margins of cratons.



**Figure 1** Tectonic sketch map of China showing Precambrian cratons, ophiolite belts (dark bands), and selected sedimentary basins (modified from Zhang et al., 1984).

## CURRENT PRODUCTION

Most mineral production figures are officially confidential, and probably no complete statistics exist for minerals produced and consumed locally in rural areas. The chapter on China (Chin, 1987) in the U.S. Bureau of Mines Yearbook includes production estimates for only about 15 industrial mineral commodities, and many commodity review articles in the mining press neglect China altogether. However, it is apparent that about 60 industrial minerals (excluding construction materials) are produced in China and about 50 are exported.

Table 1 is a summary of available data for most industrial minerals produced in China; it was compiled from a wide variety of sources. It is apparent that China is the world's leading producer (and exporter) of rare earths and barite, and an important producer of a number of other minerals. Taken together, the figures in this table indicate China's top ranking, along with the United States and the U.S.S.R., as an industrial minerals

power. Not shown in table 1 is the production trend for each commodity, which is upward in most cases as China continues its drive to industrialize and export.

In terms of foreign trade, China's exports include many minerals important to modern technology, such as refractories, rare earths, and rare metals (beryllium, lithium, niobium, and tantalum). Imported minerals include those used as fertilizers (phosphate, potash, and sulfur) and for housing (soda ash for glass, and cement). The import list underlines the country's priorities to feed and house a population of 1.1 billion people.

## INDUSTRY ORGANIZATION

The past five years have seen increasing decentralization of China's minerals industry. Many responsibilities have been removed from the bailiwick of central-government ministries and delegated to newly established, profit-oriented government corporations. Under the "responsibility system," busi-

nesses are expected to generate a profit and wean themselves from the annual state budget. At the same time, local governments (provincial, prefectural, county, and municipal) have been allowed greater autonomy in producing and selling minerals. The long-term aim of this policy is to promote efficiency and constructive competition.

A side effect of the new system has been blurring of the divisions of responsibility of the various organizations. This has produced a large degree of overlap and competition among minerals-producing organizations. As one example, the Ministry of Geology and Mineral Resources no longer functions solely as a public service, discovering mineral deposits and disseminating information; it may now compete directly with other agencies for participation in minerals development. The result can be interagency competition to the detriment of the agencies and the frustration of the potential foreign investor. In the positive view, the responsibility system has paved

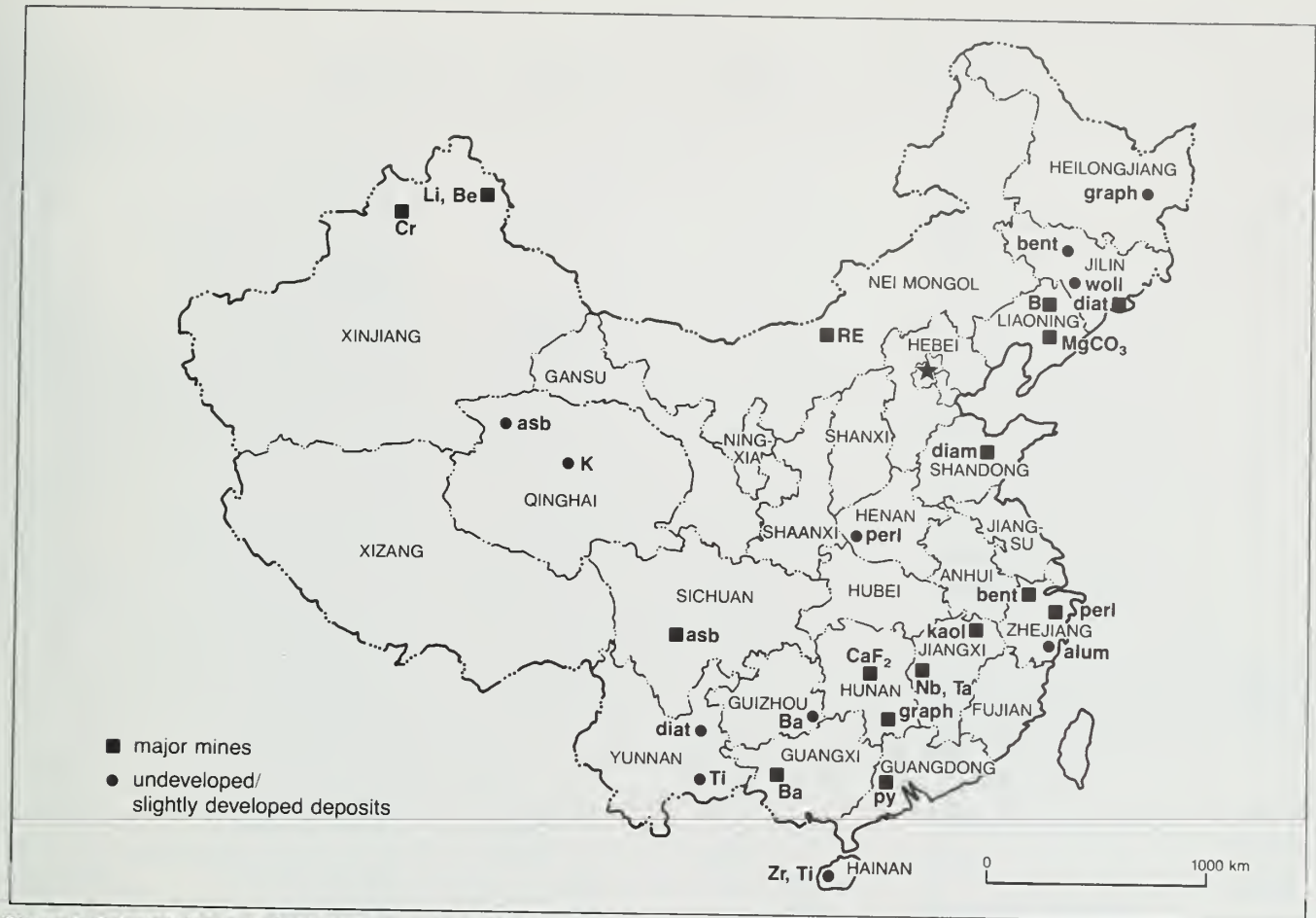


Figure 2 Selected deposits of industrial minerals in China.

the way toward eventual innovative and vigorous management of Chinese industry.

It should be apparent from the following discussion that designation of a "lead agency" on table 1 is not always a simple matter, although in most cases one organization is still dominant.

**China Nonmetallic Minerals Industry Corporation (CNMC)**, the agency most deeply involved in industrial minerals, was formed in 1985 from portions of the Ministry of Construction Materials and is responsible for most of the industrial minerals other than construction materials. When formed, CNMC was largely an umbrella organization with only one mine—an asbestos mine in Qinghai—under its direct ownership. It now has joint-venture or sales agreements with scores of locally managed mines and products plants. In addition, it sells some 80 mineral commodities, ranging from diamonds to mudstone, as well as products such as asbestos brake linings and diatomite bricks.

A major function of CNMC is apparently to provide financing, technical support, marketing, and international contacts to a host of locally managed enterprises in return for joint-venture shares. It also undertakes research on nonmetallic minerals and operates colleges of non-metallic minerals technology.

**State Administration of Building Materials (SABM)** has responsibility for production and processing of aggregates, cement, building stone, glass, and similar materials. SABM produces industrial diamonds and an array of other minerals in addition to construction materials.

**The Ministry of Chemical Industry (MCI)** mines and processes minerals used as chemical feedstocks: potash, phosphate, sulfur, sodium minerals, borates, lithium brines, and a few others. Most of this ministry's activities are in petroleum refining and petrochemicals.

**China National Nonferrous Metals Industry Corporation (CNNC)** mines and processes a host of nonferrous metals (including copper, lead, zinc, and molyb-

denum). Besides having primary responsibility in China for rare earths, titanium, chromite, and some rare metals, CNNC produces a number of industrial minerals as by-products; these include barite, fluorite, pyrite, and others. CNNC has a gemstones division, mainly for marketing of by-product gems from pegmatite and placer mines.

**The Ministry of Geology and Minerals Resources (MGMR)** functions as China's geological survey. During recent years it has become directly involved in mining because of the profit incentive. No longer does MGMR routinely pass on the successful results of its exploration programs to other agencies; now the Ministry often will negotiate an equity interest in its discoveries before disclosing data to an operating agency. For example, MGMR operates a diamond mine in kimberlite, discovered by its own personnel, in Shandong province.

**Local governments** have always been important elements in China's in-

**Table 1.** Industrial minerals production in China.\*

Commodity	Annual prod'n. (metric tons)	Supply status	Lead agency	Comments
Alunite	n.a.	exp	CNMC	Deposits near Wenzhou in Zhejiang province
Andalusite	2,500	exp	CNMC	Includes kyanite, sillimanite
Asbestos	160,000	exp	CNMC	Chrysotile and crocidolite; mines in many provinces
Barite	1,000,000	exp	CNMC CNNC	Guangxi largest producer; also some witherite
Bauxite	500,000	exp	CNMC	Henan largest producer; refractory abrasive, and aluminum ore
Bentonite	500,000	exp	SABM	Ca and Na types; mainly Liaoning
Beryl (BeO)	50	exp	CNMC	Mostly from Altai pegmatites, Xinjiang
Borates	100,000	even	MCI	Szaibelyite mine in Liaoning, resources in W. China playas
Cement	180,000,000	imp	SABM	Official figure; over 5,000 plants, including vertical kilns
Cesium	n.a.	n.a.	n.a.	Pegmatite resource in Jiangxi
Chromite	25,000	imp	CNNC	From Altai Mountains, Xinjiang
Cinnabar	n.a.	exp?	CNNC	Widely used in China as pigment, medicine, gemstone
Clays				
attapulgite	n.a.	exp	CNMC	Exported by CNMC
ball clays	n.a.	exp	CNMC	Exported by CNMC
kaolin	600,000	varies	SABM	Mainly mined in central China
sepiolite	0	n.a.	—	Deposits in Hunan
Corundum	n.a.	exp	n.a.	Most is artificial
Cryolite	n.a.	exp	n.a.	Includes chiolite
Diamond	0.04	exp (gem) imp (industrial)	CNMC MGMR SABM	Placer and kimberlite mines, Mainly Shandong, also Hunan, Liaoning; estimated 80% industrial
Diatomite	50,000	imp	CNMC	Produced in Yunnan, Jilin; insulation grade
Feldspar	n.a.	exp	CNMC	Widespread, mostly K-spar
Fluorite	750,000	exp	CNNC CNMC	Acid and metallurgical grades; largest producer Hunan province
Garnet	4,000	exp	CNMC	Probably alluvial
Gemstones, excluding diamond	n.a.	exp	CNNC CNMC	Includes turquoise, aquamarine, sapphire, ruby, amber, etc.
Graphite	180,000		CNMC	
amorphous		exp		Hunan largest producer
flake		exp		Heilongjiang, Shandong
Gypsum	10,000,000	even	MCI CNMC	Many deposits, also anhydrite
Lithium				
spodumene	15,000	exp	CNNC	Pegmatite mines, Xinjiang
brines	0	—	MCI	Resources in Qinghai and Xizang
Magnesite	3,500,000	exp	MCI	Mainly from Liaoning
Meerschaum	0	—	—	Deposit known in Hunan
Mica	25,000	exp	CNMC	Flake and scrap muscovite, phlogopite, biotite

Table 1. *continued*

Commodity	Annual prod'n. (metric tons)	Supply status	Lead agency	Comments
Nepheline syenite	n.a.	exp?	CNMC	Offered for export
Niobium	n.a.	exp	CNNC	In Nei Mongol REE mine
Olivine	n.a.	exp	CNMC	Offered for export
Perlite	500,000	exp	SABM	Mainly in Henan, Zhejiang
Phosphate	12,000,000	imp	MCI	Largest producer Yunnan
Potash (K <sub>2</sub> O)	50,000	imp	MCI	Brine production in Qinghai
Pyrophyllite	50,000	exp	CNMC	Large deposits in Zhejiang
Rare earths (REO)	15,100	exp	CNNC	Most from Bayan Obo mine, Nei Mongol; includes yttrium; also Th from monazite sands
Rubidium	n.a.	—	—	Pegmatite resources, Jiangxi
Sericite	n.a.	exp	CNMC	Offered for export
Silica, chemical grade	n.a.	exp	—	Silicon metal produced in Qinghai from natural silica
Sodium minerals salt	20,000,000	exp		Sichuan Basin rock salt; also brines and sea salt; rock-salt reserves in Jiangsu
soda ash	2,400,000	imp		Natural (in Nei Mongol) and manufactured
sulfates	300,000	exp		Natural and manufactured; mirabilite in Nei Mongol
Staurolite	n.a.	—	—	No information
Strontium	n.a.	exp	CNMC	Celestite production, Sichuan
Sulfur				
elemental	200,000	imp	MCI	
pyrite	2,300,000	imp	CNNC	Guangdong, Nei Mongol
by-product	350,000	imp	CNMC	
Talc	900,000	exp	CNMC	Largest producer is Liaoning
Tantalum	n.a.	exp	CNNC	Pegmatites, Xinjiang, Jiangxi
Titanium (TiO <sub>2</sub> )	30,000	imp	CNNC	Sponge and oxide production, igneous rocks and beach sands
Vermiculite	small	exp	CNMC	Produced in Hebei
Wollastonite	63,000	exp	CNMC	Jilin and Hubei have most of production and reserves
Zeolites	n.a.	imp?	—	Chabazite production in Hainan; also synthetics
Zircon	15,000	—	—	Mostly from small mines on Hainan beach sands

\*These data were compiled from a wide variety of sources, including U.S. Bureau of Mines (Chin, 1987), *Mining Journal*, *Engineering and Mining Journal*, Chinese press reports, and the author's notes. Data for fuels, metallic ores, and most construction material are not included. Most production figures are estimates for 1987 from unofficial sources. See footnotes for abbreviations.

#### Abbreviations:

CNMC	China Nonmetallic Minerals Industry Corporation
CNNC	China National Nonferrous Metals Industry Corporation
MCI	Ministry of Chemical Industry
MGMR	Ministry of Geology and Mineral Resources
SABM	State Administration of Building Materials
n.a.	data not available
exp	net export commodity
imp	net import commodity

dustrial minerals business because of the local use of many commodities and the labor-intensive nature of many operations. With the decentralization and profit incentive stimuli of the past few years, local governments have increased their participation in processing and selling the commodities produced. A typical small operation may be entirely local in nature; production is by manual methods, and bagged ore is sold to buyers from a higher level of government. Larger operations may be partly mechanized (mainly transport, possibly milling) and may have a higher level joint-venture partner (such as CNMC, CNNC, or MCI) to provide technical input and coordinate purchasing and product sales.

**Local private enterprise** has appeared at the lowest levels of mineral production in the form of individuals and small cooperatives producing small quantities of minerals. Generally these enterprises consist of placer operations or reworking of dumps. The products are usually sold to the local village or county government.

**China National Metals and Minerals Import and Export Corporation (CNIEC)** is the key agency involved in foreign trade of minerals. The corporation deals mainly with semifinished products and large-tonnage commodities, including fuels, metals, and some industrial minerals.

#### **FOREIGN VENTURES IN CHINA**

Foreign interests are still a minuscule element in China's industrial minerals picture. Foreign participation is usually at the level of mineral processing rather than mining. Foreign investments in ventures involving a variety of minerals, including kaolin, talc, diatomite, graphite,

diamonds, and others, are being discussed or already under way.

China's business environment has been in a state of flux since 1976, but since 1984 many laws and regulations pertaining to contracts, joint ventures, foreign exchange, and justice have become codified. Several forms of ventures, each with its own particular tax and operational advantages, are open to foreign investors in China. Besides the commonly used equity joint venture, other vehicles for foreign participation are the contractual joint venture, the wholly owned foreign enterprise, the foreign company, and the representation office.

To the foreigner who opts for involvement in China's industrial minerals business, China offers the advantages of abundant raw materials, a desire to develop and export its minerals, and a potential for enormous growth in internal markets. However, there are aspects of China's business climate that are difficult for the foreign investor to assimilate, and these factors must be taken into consideration in developing a business strategy: (1) cultural and bureaucratic obstacles to doing business; (2) a thinly stretched infrastructure of transportation, communications, and energy supplies; and (3) uneven technical backgrounds of Chinese counterpart personnel, including a low level of awareness of the importance of marketing and quality control.

#### **CURRENT TRENDS**

Huge strides have been made in the development of industrial minerals in China during the past 10 years, starting from a base that consisted mainly of construction materials and a few traditional export items. Fulfillment of development plans naturally depends on a host of

economic and political factors, but certain trends do appear likely over the next decade:

- The import/export balance for some minerals will shift as China's industrial base and consumer segment grow.
- Processed mineral products will further displace raw minerals as export items.
- Quality control will improve in response to market forces.
- Expansion of mining and processing facilities will require increasing infusions of Western technology, capital, and management.
- Chinese exports will dominate trade and will affect the prices of a number of commodities, including barite, fluorite, graphite, pyrophyllite, natural magnesite, and rare earths.
- Domestic demand will require continued imports of fertilizer minerals and increasing imports of soda ash, borates, and a few other materials.

Regardless of the degree to which each of these trends develops, China will be very much in evidence in the industrial minerals business in the coming years.

#### **ACKNOWLEDGMENTS**

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# Geology of Six Kentucky Carbonates: Sulfur Sorbents for AFBC

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## ABSTRACT

Three limestones and three dolostones from five active quarries and one mine in Kentucky were tested in the 2.7-million-Btu/hr atmospheric fluidized-bed combustion (AFBC) pilot plant at the Kentucky Energy Cabinet Laboratory to investigate their effectiveness as sulfur sorbents. The three limestones consisted of high-calcium oolitic-peloidal calcarenite from the Ste. Genevieve Limestone (Mississippian); crinoidal, bioclastic calcarenite from the Salem and Warsaw Limestones (Mississippian); and fossiliferous, bioclastic calcarenite from the Grier Limestone Member of the Lexington Limestone (Ordovician). The three dolostones consisted of the Oregon Formation (Ordovician), the Laurel Dolomite (Silurian), and the Renfro Member of the Slade Formation (Mississippian).

The dolostones performed better than the limestones as sulfur sorbents, showing higher calcium utilization values, greater sulfur capture capacity, and slightly lower SO<sub>2</sub> emission levels. Conversely, the limestones had lower rates of attrition and elutriation from the bed and slightly lower NO<sub>x</sub> emission levels.

The mean grain sizes of the limestones were 550, 800, and 1200 μm for the Grier, Salem-Warsaw, and Ste. Genevieve, respectively. The mean crystal sizes were 42, 86, and 25 μm for the Oregon, Laurel, and Renfro dolostones, respectively. Chemically, the six carbonates were composed primarily of CaCO<sub>3</sub>, MgCO<sub>3</sub>, and SiO<sub>2</sub> in the following concentrations: Ste. Genevieve (95.9, 2.1, 1.1), Salem-Warsaw (90.9, 4.5, 3.0), Grier (87.4, 3.8, 4.7), Oregon (62.4, 31.6, 4.2), Laurel (51.5, 39.0, 6.5), and Renfro (59.4, 26.2, 10.3).

utilization efficiency. In addition, limited evaluations of attrition and elutriation were conducted in a 2.54-cm (1-inch) diameter reactor. On the basis of this evaluation, three limestones and three dolostones were selected for testing in the pilot plant.

In the research reported in this paper, six Kentucky carbonates were tested in a 2.7-million-Btu/hr AFBC pilot plant at the Kentucky Energy Cabinet Laboratory (KECL) in Lexington, and results of the pilot-plant tests were compared to results of bench-scale tests of the sorbents. This paper includes information on the main features of AFBC technology, results of the pilot-plant tests, and the geology and geochemistry of the limestones and dolostones. The potential importance of dolostones as alternatives to high-calcium limestone AFBC sorbents is emphasized. Test results and engineering information included in this paper were presented at the Ninth International Conference on Fluidized Bed Combustion (Barron and others, 1987) and at "Coal Technology '86" (Barron and others, 1986).

## AFBC TECHNOLOGY

Atmospheric fluidized-bed combustion has been demonstrated at pilot scale to be an efficient, environmentally acceptable method for burning high-sulfur coal (Castleman, 1985; Tang and others, 1983). The AFBC process has a number of advantages, including in-boiler SO<sub>2</sub> removal, low NO<sub>x</sub> emission levels, high heat-transfer efficiency, low combustion temperatures that eliminate slagging and fouling, and a dry, easily handled solid refuse. In addition, an AFBC boiler can burn many different types and qualities of fuel.

Figure 1 is a schematic of the KECL AFBC pilot plant. Washed Springfield coal (Western Kentucky No.9) and the sorbent to be tested were delivered by

## INTRODUCTION

Limestone and lime have been used to control sulfur dioxide (SO<sub>2</sub>) emissions from coal-burning electric utility plants since the early 1970s. At that time, scrubbing represented a new market for limestone producers. In 1980, when the National Crushed Stone Association presented a seminar on "Limestone for Flue Gas Scrubbing" (NCSA, 1980), only one paper dealt with the use of limestones in atmospheric fluidized-bed combustion (AFBC), a technology then in the distant future. This technology came of age with the start-up of the 160-megawatt (MW) AFBC demonstration plant at the Tennessee Valley Authority's (TVA) Shawnee Steam Plant near Paducah, Kentucky, in 1988. (See Bass and others, 1987, for an overview of the development of AFBC technology at this facility.)

Carbonate rocks are an integral part of atmospheric-pressure fluidized-bed combustion of coal. They capture sulfur released from the burning coal in the boiler itself, eliminating the need for downstream scrubbers.

The overall goal of this project was to examine the basic chemical and geological parameters characteristic of some of the major carbonate deposits in Kentucky and to identify, by means of bench-scale and pilot plant tests, parameters that are useful in predicting and ranking the performance of sorbents in AFBC boilers. More than 200 samples, representing a broad range of lithologic types, were collected and analyzed chemically and petrographically. These samples were then characterized in a modified thermogravimetric analyzer (TGA) for sulfur-capture capacity and calcium-

\*Formerly called the Kentucky Energy Cabinet Laboratory.

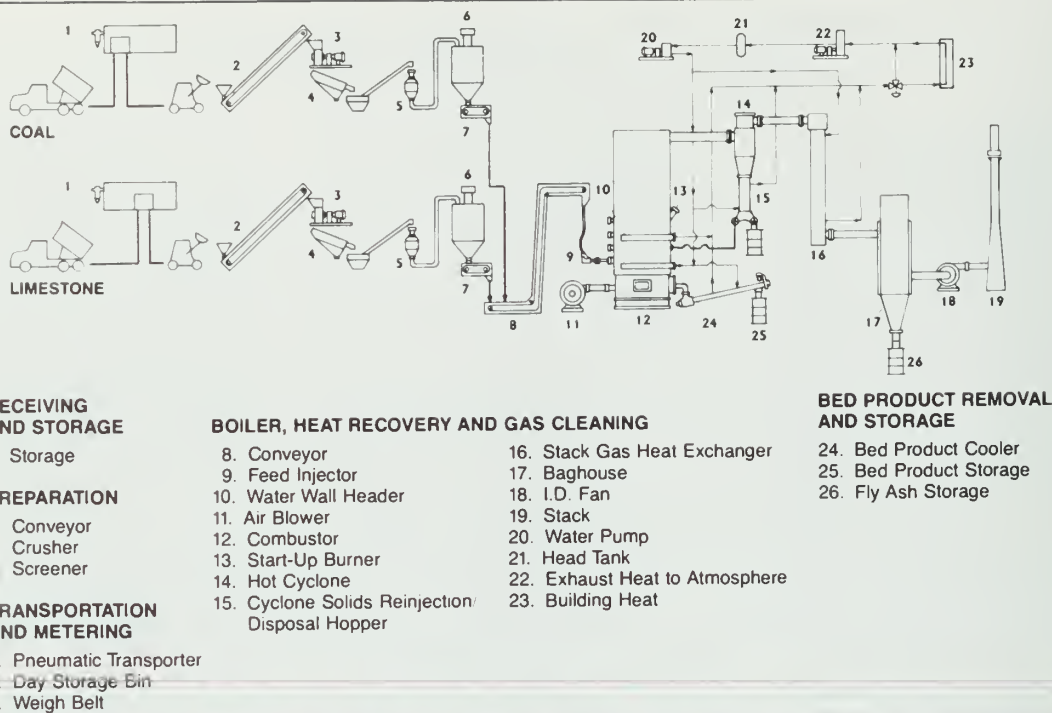


Figure 1 Schematic and flow diagram of the AFBC pilot plant.

truck to storage bins in the preparation building. Sorbent was received as No.9 aggregate (-1/2 inch x 4 mesh). Coal was crushed in the hammer mill to -1/4 inch and sorbent to -1/8 inch. These size differences reflect the different densities and fluidizing properties of the materials. Coal and sorbent limestone were then pneumatically conveyed to day bins located next to the pilot plant. During operations, coal and sorbent were metered across weigh belts and injected under-bed into the firebox. The bed of the boiler was maintained in a fluidized state by a force-draft fan providing a superficial gas velocity of approximately 5.2 feet per second. Combustion gases and entrained particulate matter exit at the top of the combustor and enter a hot-gas cyclone; here the coarse, entrained particulates are removed before the gas enters a heat exchanger, to be cooled before it enters the fine-particulate removal baghouse.

Stack-gas chemistry was monitored with a Beckman gas analyzer. Spent bed material was removed from the bed through a water-cooled auger. A computer-controlled data acquisition system monitored 128 channels of temperature, pressure, flow rates, and chemistry of the process. Operating conditions of the pilot

plant for the sorbent tests are listed in table 1.

As demonstrated in pilot plants and in the construction of demonstration plants and retrofits, AFBC technology can meet current emission requirements and has the potential to meet even stricter requirements. Current operating practices require calcium-to-sulfur ratios above the theoretical stoichiometry of one mole of SO<sub>2</sub> per mole of CaO. These ratios range up to 3:1, CaO:SO<sub>2</sub>, and even higher for more stringent emission controls. For these reasons, research into the effects of chemical, physical, and geologic parameters of limestones and dolostones on AFBC technology has been part of the research program on this technology.

## PILOT PLANT TESTS

The purpose of the pilot plant test was to assess the performance of a broad range of carbonate lithotypes from Kentucky in an industrial-type AFBC unit. The selected sorbents, representing all the major geologic systems that are commercial sources of crushed stone (table 2), were therefore tested under similar conditions in the KECL AFBC pilot plant. Washed Springfield coal (Western Kentucky No. 9) containing approximately 3.5 percent sulfur and 8 to 10 percent ash was used in a test series conducted at 70, 85, and 100 percent load.

### Carbon utilization

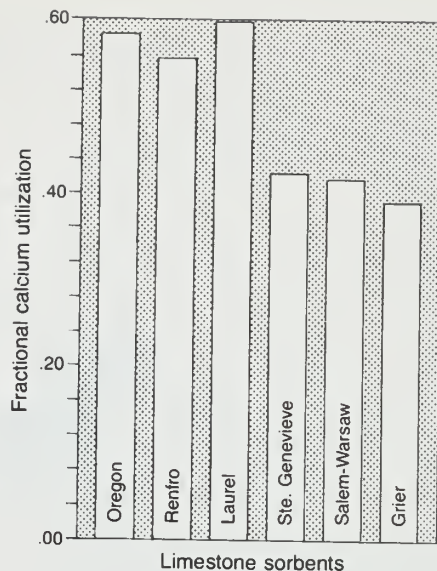
Carbon utilization values ranged from 95 to 99 percent, values comparable to

Table 1 AFBC pilot plant operating conditions

Bed area	6.4 ft (0.6 m)
Fluidizing air superficial velocity	~5.2 ft/s (1.6 m/s)
Excess air	20% (at 100% load)
100% load	2.7 million Btu/hr
Operating temperature	1500-1600°F (830-880°C)
Feedpoint	Under-bed
Cyclone catch	No recycle

**Table 2.** General geologic features of the six pilot plant carbonates tested in the AFBC.

Sorbent Number	Formation Name	Geologic System	Lithologic Type	Grain/Crystal Mean Size
1	Oregon Dol.	Ordovician	Calcareous Dol.	Crystals 42 $\mu$ m
2	Renfro Dol.	Mississippian	Calcareous Dol.	Crystals 25 $\mu$ m
3	Laurel Dol.	Silurian	Calcareous Dol.	Crystals 86 $\mu$ m
4	Ste. Genevieve Dol.	Mississippian	Oolitic Ls.	Grains 1200 $\mu$ m
5	Salem-Warsaw Ls.	Mississippian	Bioclastic Ls.	Grains 800 $\mu$ m
6	Grier Ls.	Ordovician	Fossiliferous Ls.	Grains 550 $\mu$ m

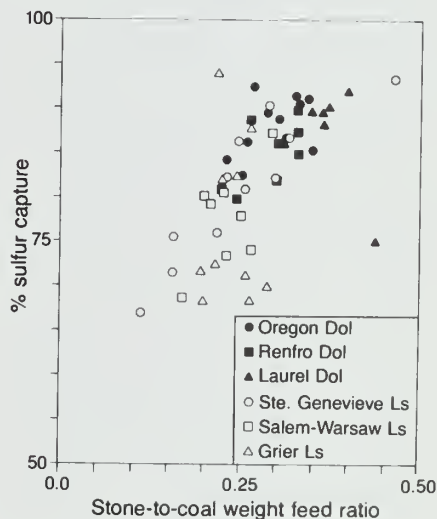


**Figure 3** Maximum fractional calcium utilization observed in the KECL pilot plant for each carbonate rock tested (Barron and others, 1986).

also demonstrates that the dolostones are less sensitive to sorbent feed rate than are the limestones (as reflected by higher calcium utilization and lower SO<sub>2</sub> emissions for the dolostones).

### SELECTION OF SORBENTS

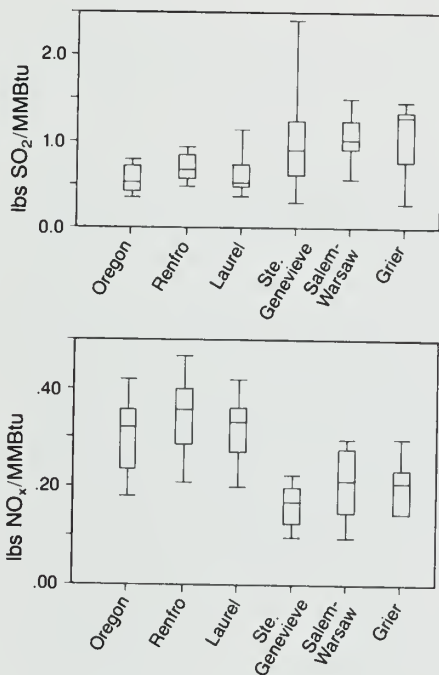
The limestones and dolostones selected for testing were obtained from active quarries or mines in stone-producing areas of Kentucky; they represented a variety of carbonate lithotypes and were lithologically homogeneous.



**Figure 4** Stone-to-coal weight ratio versus sulfur capture percentage. The scatter in the data is due to different operating conditions, such as bed height, excess air, and load (Barron and others, 1986).

### Calcium utilization

Calcium utilization values were higher for the dolostones than for the limestones (fig. 3); the effect of higher calcium utilization was of course reduced somewhat by the higher calcium contents of the limestones.



**Figure 2** Emission levels of SO<sub>2</sub> and NO<sub>x</sub> for the six carbonates tested in the AFBC pilot plant. The uppermost and lowermost lines (whiskers) for each data set denote the range of values, the ends of the boxes mark the quartiles, and the line in the box is the median of the data for that test (Barron and others, 1987).

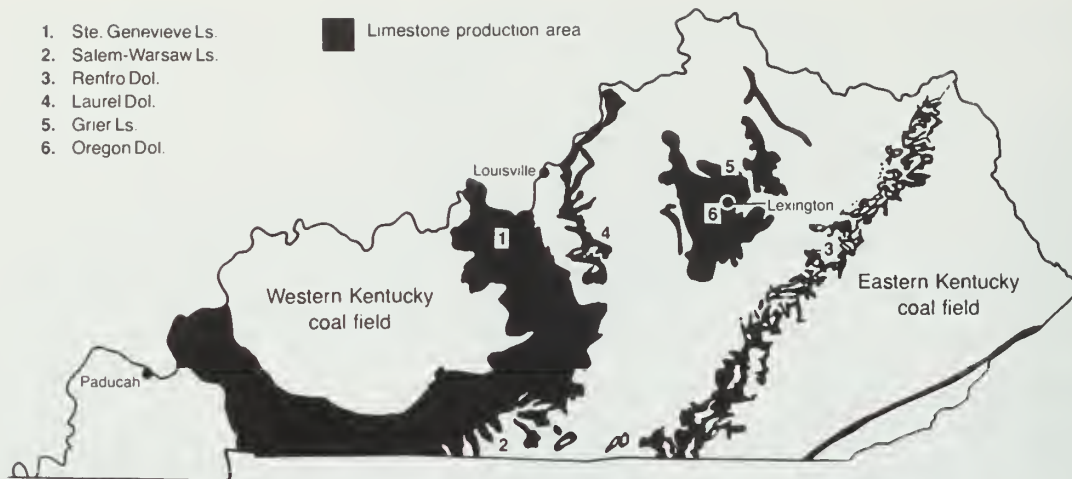
### Sulfur capture capacity

In these tests, sulfur capture was affected by sorbent feed rate (fig. 4), a result consistent with results obtained by others (Castleman, 1985; Tang and others, 1983). This finding supports the concept that increasing the amount of sorbent fed at a given coal feed rate will reduce stone utilization efficiency. The plot in figure 4

those obtained in other tests in this pilot plant. These percentages were higher than those obtained in other AFBC test facilities (Castleman, 1985; Tang and others, 1983). However, the differences in test results can be explained by the absence of a convection-pass heat exchanger in the freeboard area (the area between the top of the bed and the top of the combustor) of this pilot plant. The effect of the heat exchanger in the other units is to lower the temperature of that zone to below combustion levels; thus, any unburned or partly burned coal carried into the zone would not be burned, and carbon utilization, as measured in the ash, would be lower. Because the pilot plant had no heat exchanger to reduce the temperature in this unit, elutriated coal continued to burn in the freeboard, and the carbon content in the ash was lower than it would have been if the temperature had been reduced.

### Emission levels

Data on NO<sub>x</sub> emission levels in the six tests (fig. 2) indicate that all test results were well below the NO<sub>x</sub> emission standard of 0.6 lbs NO<sub>x</sub> per million Btu (MMBtu), as expected with AFBC units (Castleman, 1985; Furusawa and others, 1978; Hansen and others, 1983). The dolostones had slightly higher NO<sub>x</sub> levels than the limestones. SO<sub>2</sub> emission levels for all carbonates were below the 1.2 lbs SO<sub>2</sub>/MMBtu emission standard. Lower SO<sub>2</sub> levels were achieved with the dolostones, partly because of the greater reactivity of dolostones and partly because of the slightly higher sorbent feed rates. Because the dolostones had lower SO<sub>2</sub> levels, the higher NO<sub>x</sub> levels were probably related to increased sulfation of the dolostones. Other researchers (Castleman, 1985; Furusawa and others, 1978) have reported on tests of partly sulfated beds.



**Figure 5** Locations of the quarries that supplied carbonate rocks for the AFBC pilot plant tests, shown in relationship to the major limestone-producing areas and coal fields of Kentucky.

In previous research at the KECL AFBC pilot plant, various Kentucky coals were tested, but only one locally available carbonate sorbent, the Oregon Formation, was used in all runs. In the current phase of research, the sorbent characteristics of six carbonate rocks were determined, but the same high-sulfur Springfield coal (Western Kentucky No. 9) was used in all runs.

Three dolostone and three limestone lithotypes were selected because bench-scale testing of carbonate rocks from across Kentucky had indicated that both dolostones and limestones were potentially suitable for use in AFBC plants. Testing at KECL, and at other laboratories (Borgwardt and Harvey, 1972; Harvey and Steinmetz, 1972; Snyder, Fuchs, and Wilson, 1978), indicated that different carbonate lithologies reacted differently in the calcining/sulfating AFBC environment; therefore, lithologically homogeneous sorbent feedstocks were sought in this study to facilitate interpretation of study results. This requirement eliminated many quarries and mines as potential sources of stone because their active faces contained a mixture of carbonate lithologies.

We obtained stone only from active operations (fig. 5) because relatively large quantities of stone were required for the test runs and because we wanted to document potential sources of sorbent stone for future AFBC plants. No stone was obtained from the western part of the Mississippian Plateau Region in western Kentucky because the Warsaw and Ste.

Genevieve Limestones (the principal sources of stone in the area) had already been used as sorbents in the TVA 20-MW pilot plant near Paducah, Kentucky.

### GEOCHEMISTRY OF SORBENTS

The concentration of  $\text{CaCO}_3$  in the three limestones ranged from 85 to 95 percent; the  $\text{MgCO}_3$  content of each limestone was less than 5 percent (fig. 6). The total content of inerts (mostly quartz and clay) for each limestone was below 5 percent. Two of the limestones, the Ste. Genevieve and the Salem-Warsaw, were high-carbonate stones ( $\text{CaCO}_3 + \text{MgCO}_3 > 95\%$ ); the Ste. Genevieve was a high-calcium limestone ( $> 95\% \text{CaCO}_3$ ). The  $\text{MgCO}_3$  content of the calcareous dolostones ranged from 25 to 36 percent (fig. 6), short of the  $\text{MgCO}_3$  concentration required for true dolomites in the strict industrial sense (40 to 43%  $\text{MgCO}_3$ ). The total content of inerts (mostly  $\text{SiO}_2$ , quartz or chert primarily) for each dolostone ranged from 6 to 14 percent, higher than in the limestones.

### GEOLOGY OF SORBENTS

The three dolostones and three limestones are described separately, in ascending stratigraphic order, in the following sections. Stratigraphic positions of the carbonate units are shown in figures 7 and 8 and general characteristics are summarized in table 2.

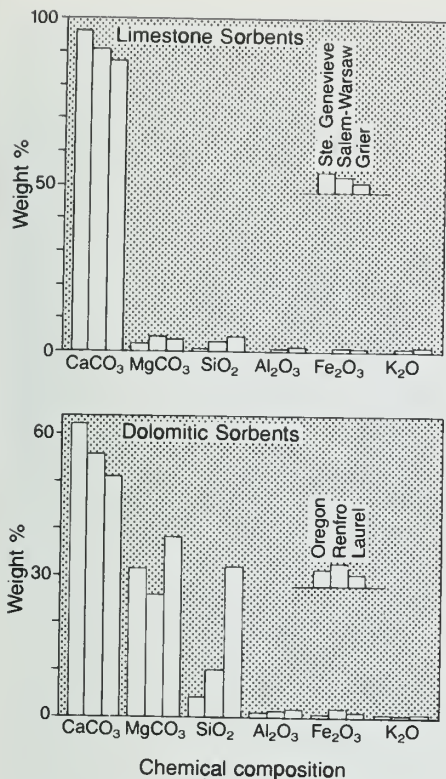
#### Dolostones

**Oregon Formation** The Oregon Formation is part of the High Bridge Group

(Middle Ordovician), a thick (430 to 570 ft) sequence of limestone and dolostone (fig. 7). The Oregon is composed of calcareous dolostone commonly interbedded with calcilitite. The dolostone is yellowish gray to yellowish brown, intricately mottled with dark gray; it is finely crystalline and thick bedded. Microscopically, it consists of a mixture of anhedral and euhedral dolomite crystals with small scattered patches of calcite microspar (fig. 9). The dolomite crystals generally have dark boundaries. The size of the dolomite crystals (measured along apparent long axes) averaged 44  $\mu\text{m}$  in the long dimension.

The High Bridge Group, the oldest strata exposed in Kentucky, crops out on the Cincinnati Arch in central Kentucky along valleys of the entrenched Kentucky River (fig. 10) and its tributaries; it is at minable depth beneath a large part of central and north-central Kentucky. The Oregon Formation ranges from 6 to 65 feet thick (Cressman and Noger, 1976).

Construction and agricultural stone are produced from the Oregon at six underground mines in central Kentucky. The dolostone tested in the AFBC pilot plant was obtained from the Central Rock Company mine in Lexington, Fayette County, where the formation is 28 feet thick. At the time the stone was taken for testing, a bench in the lower 17 feet of the formation was being mined. Five of the operations in the Oregon, including the Central Rock Company mine, also produce stone from the basal part of the overlying Tyrone Limestone.

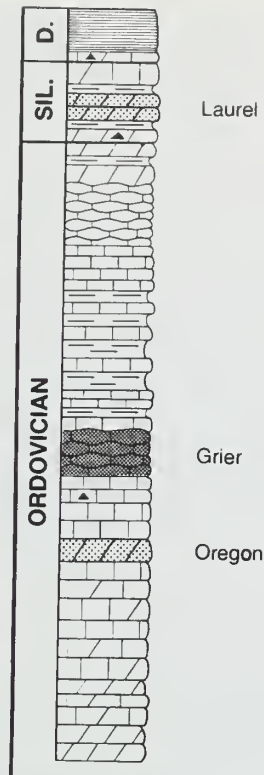


**Figure 6** Chemical compositions (weight percent) of the six carbonate rocks tested in the AFBC pilot plant.

**Laurel Dolomite** The Laurel Dolomite (Middle Silurian, fig. 7) is composed of light gray to olive gray calcareous dolostone with minor amounts of shale in partings and thin beds. The dolostone mainly consists of two types termed informally the "vuggy" stone and the "quarry" stone (Peterson, 1981). The "vuggy" dolostone is finely to medium crystalline, very porous, fossiliferous, and massive to thin bedded; it forms extremely pitted, weathered surfaces. The "quarry" stone is finely to very finely crystalline, porous, and sparsely fossiliferous; it occurs in thin to thick planar beds and forms dense, smooth, rounded surfaces in weathered exposures. In a quarry face of the Laurel shown in figure 11, the "quarry" stone overlies the "vuggy" stone.

Porosity in the formation is mainly biomoldic and partly intercrystalline. In thin section (fig. 12), the Laurel consists primarily of euhedral dolomite rhombohedra with patches of calcite microspar. The average size of the dolomite crystals, measured along apparent long axes, was 88  $\mu\text{m}$ .

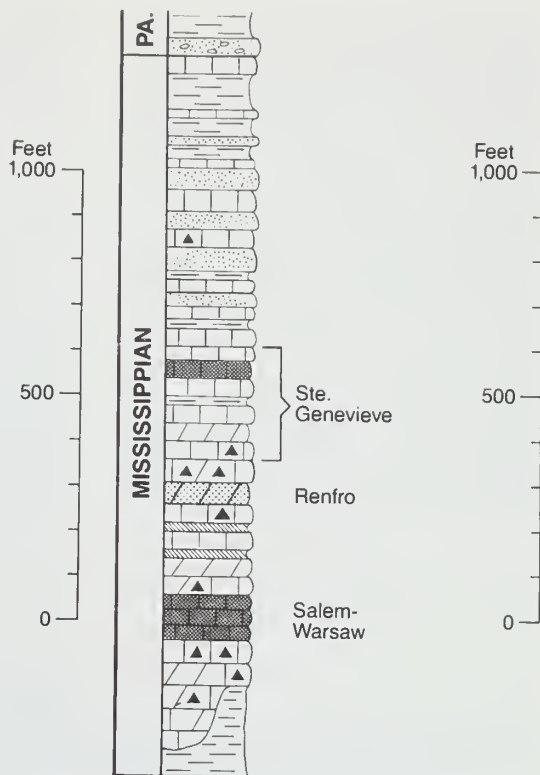
The Laurel crops out in a narrow belt along the western border of the Outer Blue Grass Region in north-central and



**Figure 7** Stratigraphic positions of Ordovician and Silurian carbonate units on a generalized columnar section for surface rocks in Kentucky.

west-central Kentucky; the belt extends from Trimble County on the Ohio River southward for a distance of about 80 miles into western Marion County. (Pre-Middle Devonian erosion truncated the formation at the southern end of the belt.) The Laurel also crops out in south-central Kentucky in a small area along the Kentucky-Tennessee border. In many places the Laurel is 40 to 55 feet thick; its maximum thickness is 65 feet (Peterson, 1981). Construction and agricultural stone are produced from the Laurel at three quarries and two underground mines in the western Blue Grass Region. Working faces encompass both the "vuggy" dolostone and the "quarry" stone. The dolostone tested in the AFBC pilot plant was obtained from a 22-foot face in the upper Laurel (fig. 11) at the Medusa Aggregates Company's Bardstown quarry in Nelson County.

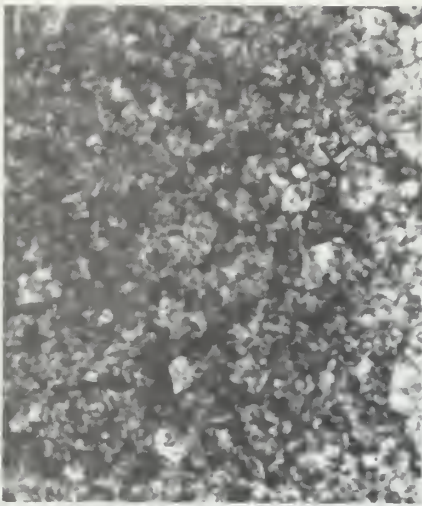
**Renfro Member of the Slade Formation** The Renfro Member, the basal unit of the limestone-dominated Slade Formation (Mississippian; see fig. 8), is composed mostly of calcareous dolostone interbedded with limestone and shale. The dolostone is thick bedded to thin bedded (fig. 13), and very finely to



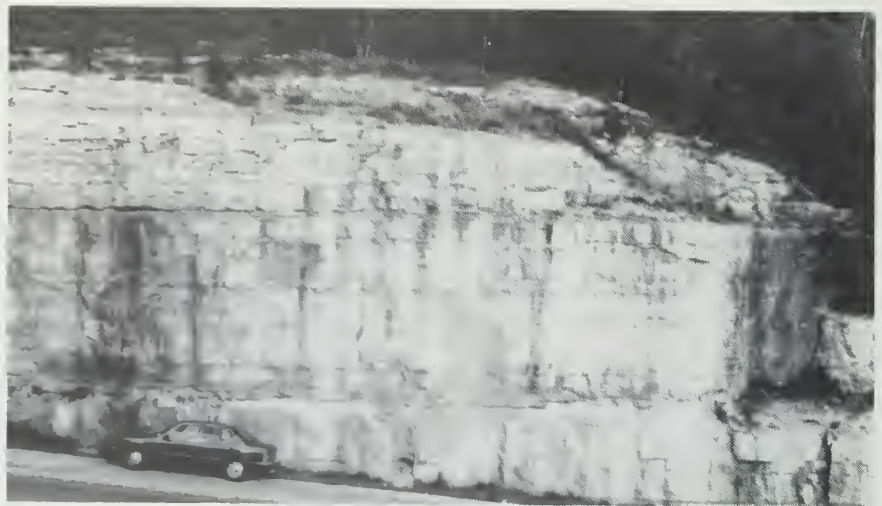
**Figure 8** Stratigraphic positions of Mississippian carbonate units on a generalized columnar section for surface rocks in Kentucky.

finely crystalline; it is light gray and weathers rapidly to yellowish orange or grayish orange. Microscopically, the Renfro consists mostly of euhedral to anhedral dolomite crystals (fig. 14); it has a 4 percent ankerite content and small (10  $\mu\text{m}$ ) crystals of calcite. The average dolomite crystal size (measured along apparent long axes) is 25  $\mu\text{m}$ . The dolomite has a ferroan dolomite content of about 33 percent, as determined by potassium ferrocyanide staining. The dolostone has very fine intercrystalline porosity and irregular patches of calcilitite.

Mississippian limestones and dolostones crop out along the western border of the Eastern Kentucky Coal Field in a northeastward-trending belt extending across south-central, east-central, and northeastern Kentucky from the Tennessee state line to the Ohio River. The name Renfro is applied in east-central and northeastern Kentucky to the dolostone at the base of this carbonate sequence. In south-central Kentucky, rocks correlative with the Renfro are thicker and more varied in lithology; they form, in descending order, middle and lower parts of the St. Louis Limestone, Salem and Warsaw Limestones, and the



**Figure 9** Photomicrograph of the Oregon Formation from the Central Rock Company mine in Fayette County, Kentucky. Average crystal size is 44  $\mu\text{m}$  in the long dimension.



**Figure 10** Roadcut in the Ordovician High Bridge Group along U.S. Highway 27 as it crosses the Kentucky River in Jessamine County, Kentucky. Base of the Oregon Formation is near the shale parting just above the top of the car. The next significant shale parting is in the base of the overlying Tyrone Limestone.

Muldraugh Member of the Borden Formation (Dever and Moody, 1979).

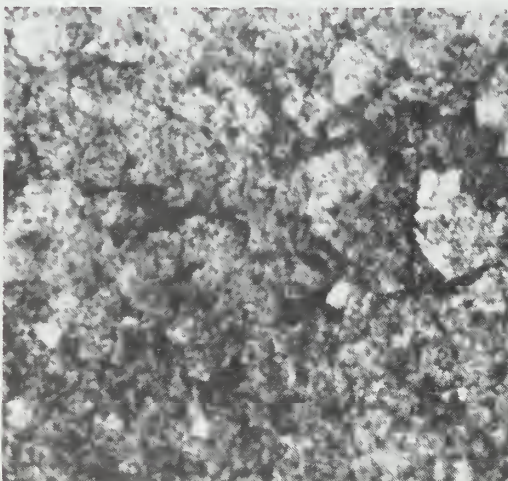
The Renfro thins northeastward across the outcrop belt, from 120 feet in southern east-central Kentucky to 2 feet in northeastern Kentucky. Intra-Mississippian erosion removed the Renfro from parts of northeastern and northern east-central Kentucky.

The dolostone tested in the AFBC pilot plant was obtained from the Natural Bridge Stone Company quarry in Powell County, where Slade limestones overlying the Renfro are quarried for construction stone. A bench of about 15 feet of upper Renfro dolostone was excavated from below the quarry floor (fig. 13) but the stone did not meet construction-stone specifications. Stone from a stock pile of the waste dolostone was prepared as No. 9 aggregate for use in the AFBC pilot plant.

Commercial use of the dolostone in the region presently is limited to one underground operation in south-central Kentucky, where St. Louis dolostone and limestone correlative with the Renfro are mined for agricultural and construction stone. Renfro dolostone would be readily exploitable at many active quarries and mines in the region where the top of the member is a short distance below or at the present floor. Potential exploitation of the Renfro will be somewhat restricted in southern east-central Kentucky, however, because in this region its lower part consists mostly of cherty and geodiferous dolostones. Nodular quartz, associated



**Figure 11** Quarry face of the Silurian Laurel Dolomite in the Medusa Aggregates Company Bardstown quarry in Nelson County, Kentucky, showing bedded "quarry stone" overlying pitted "vuggy" dolostone.

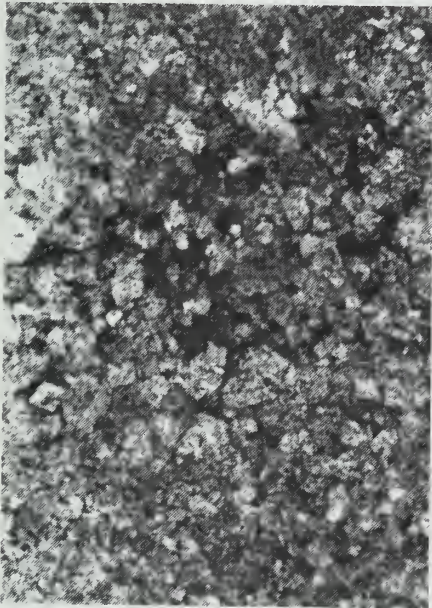


**Figure 12** Photomicrograph of the Laurel Dolomite from the Medusa Aggregates quarry. Average crystal size is 88  $\mu\text{m}$  in the long dimension.

with brecciated dolostone, also occurs in the upper part of the member. These deleterious constituents generally are absent in the Renfro of northern east-central and northeastern Kentucky.



**Figure 13** Upper part of the cut in the Renfro Member of the Slade Formation in the Natural Bridge Stone Company Quarry in Powell County, Kentucky. The hand of the person in the photograph is at the contact between the Renfro and the overlying St. Louis member.



**Figure 14** Photomicrograph of the Renfro Member of the Slade Formation. Average crystal size along the longest grain axis is 25  $\mu$ m.

## Limestones

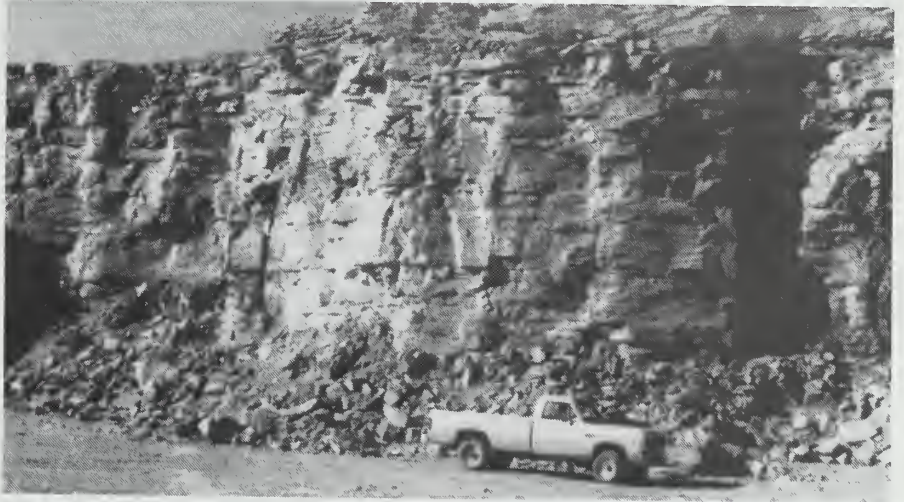
**Grier Limestone Member of the Lexington Limestone** The Grier Limestone Member of the Lexington Limestone (Middle Ordovician, fig. 7) consists of fossiliferous calcarenite and calcisiltite and minor amounts of calculutite. Shale partings are common. The limestone, light brownish gray to pale yellowish brown and medium dark gray to light olive gray, is cemented by medium to coarsely crystalline sparry calcite; whole and fragmented fossils are partly recrystallized. Beds are thin to medium, irregular, nodular, and tabular (fig. 15), and scattered fossils are replaced by gray chert. In thin section (fig. 16), the Grier is composed of about 50 percent medium to coarse calcite spar and 40 percent calcite microspar; the remaining 10 percent consists of dolomite and microcrystalline calcite. Mean grain size (measured along apparent long dimensions) is 550  $\mu$ m,

reflecting the large size range in fossil fragments.

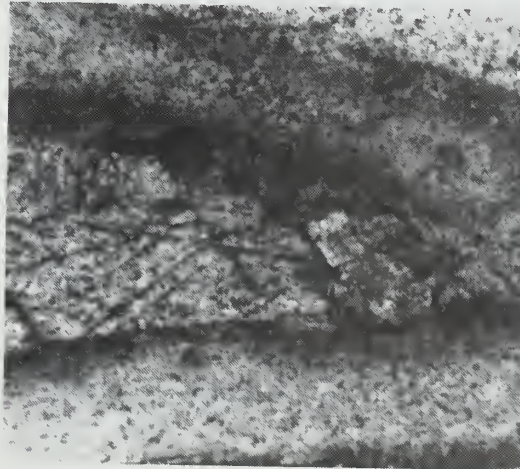
The Grier crops out in central and north-central Kentucky, across the Inner Blue Grass Region and along the valley of the Licking River. Its wide variation in thickness (60 to 180 ft) reflects intertonguing with adjacent members of the Lexington Limestone (Cressman, 1973).

Construction and agricultural stone are produced from the Grier, commonly along with adjacent members of the Lexington, at seven quarries in central and north-central Kentucky. The limestone for the AFBC pilot plant was obtained from the Nally & Gibson Georgetown, Incorporated quarry from a 55-foot face in the upper Grier (fig. 15). The base of the member is not exposed at the quarry, but the Grier is about 130 feet thick in the vicinity (Cressman, 1973).

**Salem and Warsaw Limestones** The Salem Limestone and Warsaw Limestone (Mississippian) of south-central Ken-



**Figure 15** Quarry face of the Grier Limestone Member of the Ordovician Lexington Limestone in the Nally & Gibson Georgetown quarry in Scott County, Kentucky.



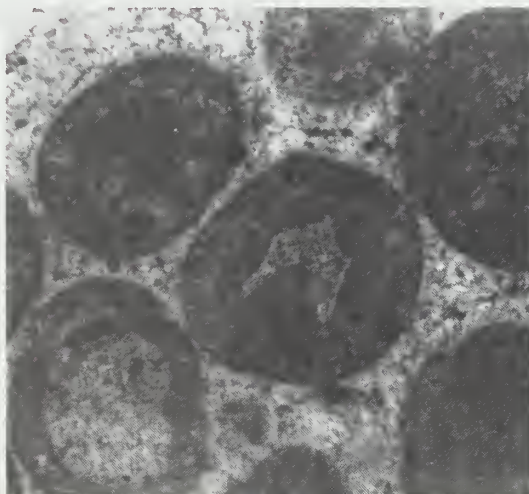
**Figure 16** Photomicrograph of the Grier Limestone Member of the Lexington Limestone showing dolomite crystals within a folded trilobite fragment.



**Figure 17** Quarry face of the Mississippian Salem-Warsaw Limestones in the Southern Aggregates quarry in Allen County, Kentucky. The top of the petroleum-stained zone at the base of the face is approximately 12 feet above the quarry floor.



**Figure 19** Quarry face of the Mississippian Ste. Genevieve Limestone in the Kentucky Stone Company Irvington quarry in Breckinridge County, Kentucky. The high-calcium ledge is the crossbedded, lighter colored unit at the base.



**Figure 20** Photomicrograph of the Ste. Genevieve Limestone showing oolites, micrite-enveloped grains, and spar cement. Average grain size in the longest dimension is 1200  $\mu\text{m}$ .



**Figure 18** Photomicrograph of the Salem-Warsaw Limestones showing pelmatozoan fragments. Average grain size is 800  $\mu\text{m}$  along the longest dimension.

tucky were combined into one mapping unit, Salem-Warsaw Limestones (fig. 8), during the U.S. Geological Survey-Kentucky Geological Survey cooperative mapping project (1960 to 1978). The unit is composed of crinoidal calcarenite and contains zones of sandstone, argillaceous dolomite, and shale. The calcarenite is medium gray to olive black, partly fossiliferous and locally dolomitic and contains minor amounts of chert; it is thin bedded to thick bedded (fig. 17) and commonly cross bedded. In thin section, pelmatozoan particles—the principal constituent (fig. 18)—and smaller amounts of bryozoan particles are both cemented with clear sparry calcite. About one-third of the pelmatozoan fragments have syntaxial rim cement; minor constituents include mollusks, algae, and dolomite crystals. Mean grain size (measured along apparent long dimensions) is 800  $\mu\text{m}$ .

The Salem and Warsaw Limestones crop out in the Mississippian Plateau Region across south-central Kentucky, from Simpson and Warren Counties eastward into Wayne and Pulaski Counties. The unit thickness ranges from 35 to 170 feet.

Construction and agricultural stone are produced from the Salem-Warsaw at



**Table 3** TGA experimental conditions.

Sample mass	45-60 mg
Sample particle size	850 $\mu\text{m}$ - 1 mm (No. 18 X 20 seive)
Operating temperature	1600°F (870°C)
Gas flow rate	8.3 ml/s
Calcining gas composition	N <sub>2</sub> , 81.5%; CO <sub>2</sub> , 16%; O <sub>2</sub> , 2.5%
Calcining time	30 minutes
Sulfating gas composition	N <sub>2</sub> , 75.2%; CO <sub>2</sub> , 16%; O <sub>2</sub> , 2.5%; SO <sub>2</sub> , 0.3%
Sulfating time	160 minutes

two quarries in south-central Kentucky. The limestone for the AFBC pilot plant was obtained from the Southern Aggregates, Incorporated Scottsville quarry in Allen County, from an 80-foot face (fig. 17) in a unit having a total thickness of about 110 feet near the quarry (Ketner, 1962).

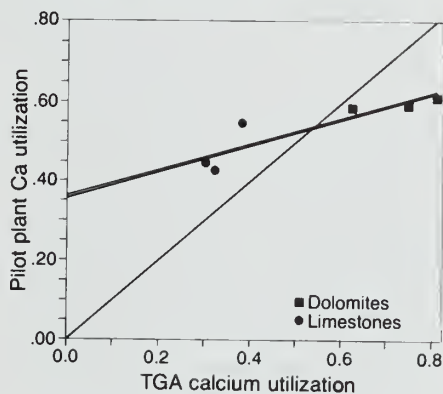
**Ste. Genevieve Limestone** The Ste. Genevieve Limestone (Upper Mississippian, fig. 8) consists mostly of limestone (calcarene and calcilitite), with smaller amounts of dolostone, shale, sandstone, and chert. The limestone is white to olive gray, partly fossiliferous, thinbedded to thickbedded, and partly crossbedded (fig. 19). Calcarenes are composed of bioclastic, micrite-enveloped peloidal and oolitic grains.

The Ste. Genevieve crops out in the Mississippian Plateau Region of western and west-central Kentucky. Equivalent limestones in the Monteagle Limestone, Slade Formation, and Newman Limestone crop out in south-central, east-central, and southeastern Kentucky, respectively. The thickness of the Ste. Genevieve and its equivalents ranges from 320 feet in western Kentucky to less than 10 feet in east-central Kentucky. Intra-Mississippian erosion removed the Ste. Genevieve equivalent from northeastern and parts of northern east-central Kentucky.

Construction, agricultural, and industrial stone are produced from the Ste. Genevieve (commonly together with adjacent limestone units) at 38 quarries and mines in the state. The limestone for the AFBC pilot plant was obtained from the Kentucky Stone Company's Irvington quarry in Breckinridge County, where a 17-foot deposit of high-calcium "oolitic" calcarenite (fig. 19) is selectively quarried for industrial use. The top of the calcarenite is 34 feet below the top of the Ste. Genevieve, which is about 200 feet thick

near the quarry (Dever and others, 1979). The quarry produces construction stone from the lower part of the formation. A lithologically similar high-calcium calcarenite from the Ste. Genevieve in Caldwell County has been used in the TVA 20-MW AFBC pilot plant at the Shawnee Steam Plant near Paducah in western Kentucky.

The light gray to white "oolitic" calcarenite at Irvington consists of micrite-enveloped bioclastic grains and peloids (micritized, algal-bored grains). True oolites were rare. In thin section (fig. 20) the principal volumetric constituents, in descending order, are sparry calcite ce-



**Figure 21** Comparison of calcium utilization data obtained from the TGA with that observed in the pilot plant tests. The single line represents ideal 1:1 relationship; the double line represents a least-squares regression through the six data points (Barron and others, 1987).

ment, micrite-enveloped grains, and peloids. Minor constituents include bioclasts, dolomite, and trace amounts of chalcedony. Mean grain size in the longest dimension is 1200  $\mu\text{m}$ .

### THERMOGRAVIMETRIC ANALYSIS RESULTS

Thermogravimetric analyses (TGA) were conducted on an apparatus patterned after Ruth, Squires, and Graff

(1972) and Snyder, Wilson, and Johnson (1978) (see Barron and others, 1987). In a TGA test, a sample of limestone is placed in a quartz pan on a microbalance. The arm of the balance is placed in a furnace operated isothermally at 87°C. The sample is calcined (table 3) for 30 minutes and then exposed to SO<sub>2</sub>-bearing gas for almost 3 hours. During calcination and sulfation, the microbalance records weight loss and weight gain. The resulting data can be used to calculate sulfation capacity and calcium utilization (combined with chemical content). Table 3 outlines the experimental conditions.

One of the parameters derived from the TGA tests is calcium utilization, which provides a simple, first look at performance of candidate limestones. Calcium-utilization values determined by TGA differ from calcium-utilization values obtained in the pilot plant. The data plotted in figure 21 demonstrate the differences between limestone and dolostone values obtained from TGA data and from pilot plant performance results. For the dolostones, calcium utilization values obtained in pilot plant tests were lower than expected on the basis of TGA data. Conversely, for the limestones, calcium utilization values were higher in the pilot plant results than in the TGA results. Although only six data points are available to work with at this time, it appears that the TGA-derived calcium utilization number is useful for screening purposes.

### SUMMARY

- Six commercially produced Kentucky carbonates were successfully tested as AFBC sulfur sorbents in the AFBC pilot plant at the Kentucky Energy Cabinet Laboratory; all limestones and dolostones achieved NO<sub>x</sub> and SO<sub>2</sub> levels at or below the emission standards.
- The three calcareous dolomites demonstrated greater sulfur dioxide capture and calcium utilization and lower sulfur dioxide emission levels than the limestones did.
- The three limestones exhibited lower NO<sub>x</sub> emissions and required lower feed rates of stone than the dolomites.
- The high-calcium Ste. Genevieve limestone performed better than the bioclastic and fossiliferous limestones.
- TGA calcium-utilization data corresponded well with actual pilot plant performance for all six carbonate rocks.

## ACKNOWLEDGMENTS

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# Possible Underground Mining of Limestone and Dolomite in Central Illinois

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## ABSTRACT

An 11-county area in central Illinois that produces little or no crushed limestone or dolomite for use as construction aggregate includes several major downstate metropolitan centers: Bloomington-Normal, Champaign-Urbana, Decatur, Peoria, and Springfield. In much of this area quarriable thicknesses of limestone or dolomite either do not exist or lie beneath excessive thicknesses of drift and younger, predominantly clastic bedrock of the Pennsylvanian System.

Preliminary studies indicate that underground mining of sub-Pennsylvanian limestone or dolomite formations could provide new sources of limestone and dolomite in central Illinois to meet the growing need for crushed stone. Potential targets include the Valmeyeran (middle Mississippian) Ste. Genevieve, St. Louis, and Burlington; the Middle Devonian Cedar Valley, Wapsipinicon, and Grand Tower; the Niagaran (upper Silurian) Racine; and Champlainian (middle Ordovician) Galena Group.

A detailed investigation of the depth to minable stone, the chemical and physical characteristics of the stone, and the specific factors that favor underground mining operations could lead to profitable mining operations in central Illinois, where both the resources and the market are present.

## INTRODUCTION

Illinois has been a leading producer of limestone and dolomite rock products for many years. In 1988, Illinois ranked sixth among crushed stone-producing states with production of 54.3 million tons. Major quarries serving the metropolitan Chicago area are among the largest in the United States (Mikulic, 1988). However, although quarriable deposits in Illinois may appear to be practically inexhaustible, the resources are not at all evenly distributed geographically, and it is becoming increasingly difficult for producers to supply the demands of major downstate markets for high-quality aggregates.

Major production of limestone for aggregate in Illinois (fig. 1) has occurred in counties near the southern and western borders of the state—in or adjacent to the bluffs of the Ohio and Mississippi Rivers and, to a lesser extent, along the Illinois River. Most dolomite is produced in the northernmost counties; approximately one-fourth of the crushed stone produced

in Illinois in recent years has come from dolomite quarries in Cook County alone.

An 11-county region in central Illinois has little or no crushed stone—limestone or dolomite. In this region, construction companies needing material for concrete and bituminous aggregate traditionally used natural or crushed gravel of glacial (Quaternary) origin (fig. 2) or trucked in stone from quarries in nearby Livingston, Vermilion, Montgomery, Clark, Coles, Christian, Menard, Logan, and Douglas counties. (These sources also provide agricultural limestone.) But demand for limestone and dolomite is growing and transportation costs are escalating; moreover, many local gravels are not suitable for highway construction and some other uses.

Most of the stone produced in Illinois is won by open-pit mining methods, and without doubt, when conditions are favorable, surface mining is more profitable than underground methods. However, in some cases, geological, environmental, and economical factors can make underground mining profitable,

and in some situations underground mining becomes the preferred mining method (Baxter, 1980).

This preliminary assessment of the feasibility of underground mining of limestone and dolomite is based largely on published information of the Illinois State Geological Survey and interpretations by the author.

## LIMESTONE AND DOLOMITE OUTCROPS AND QUARRIES

The pattern of stone production in Illinois reflects the distribution of outcrops imposed by the structural configuration of the Illinois Basin, the sedimentary history of the Basin, and subsequent glacial events. The following discussion of outcrops and quarries is limited to stratigraphic units considered to be targets for underground mining in central Illinois.

The bedrock surface is covered by glacial drift (fig. 3) that either obscures or deeply buries the bedrock (fig. 4) and is thickest in the central Illinois area. Strata of Pennsylvanian age, which are dominantly clastic, underlie the drift in a large part of the central region. Limestones of Pennsylvanian age are generally thin and discontinuous and commonly contain interbedded shale. Pennsylvanian limestones are quarriable where they attain workable thicknesses of acceptable quality and the overburden is manageable (Goodwin, 1983).

Mississippian limestones, particularly the thick formations of Valmeyeran age, are prime sources of crushed stone in southern and western Illinois (Goodwin, 1983); the limestone quarries are largely restricted to areas in and adjacent to the bluffs of the Ohio, Mississippi, and Illinois Rivers. Some of these limestones tend to be soft and lack the soundness required for use in Portland cement and bituminous mixes.

Quarries in rocks of Devonian age are restricted mostly to areas adjacent to the Mississippi River in Rock Island and ad-

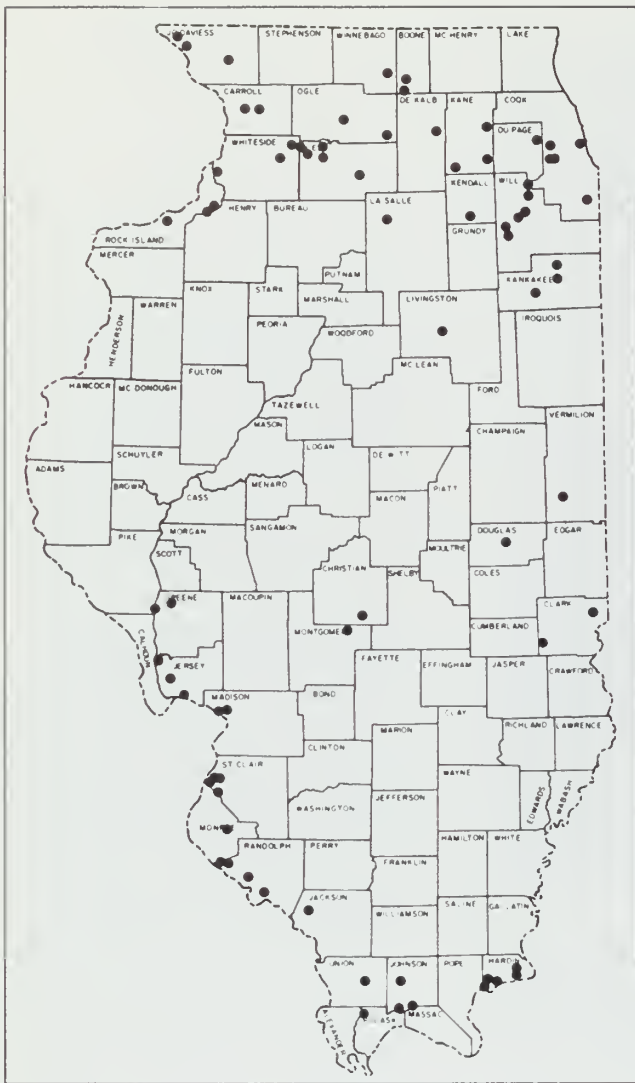


Figure 1. Sources of crushed limestone or dolomite concrete aggregate in Illinois (IDOT, 1986).

joining counties (Goodwin, 1983). Middle Devonian limestone is quarried in the vicinity of the Quad Cities in Rock Island County and near Tuscola in Douglas County, where units of this age reach the bedrock surface at the crest of the La Salle Anticlinal Belt.

Carbonate rocks of the Silurian System (Willman, 1972) are found at the bedrock surface in northern Illinois, where they are predominantly dolomite, and at places along the Mississippi River in southern Illinois, where they are limestone (Goodwin, 1983). High-purity dolomite (greater than 42% magnesium carbonate and more than 97% magnesium carbonate and calcium carbonate together) like that found at the Thornton Quarry in Cook County (Mikulic, 1988)

is the backbone of the aggregate and lime industries in northeastern Illinois.

Carbonate strata of the Ordovician System crop out extensively in northwestern and north-central Illinois (Willman and Kolata, 1978) and are exposed at a few sites along the Mississippi River in western and southern Illinois. Formations of the Galena and Platteville Groups are quarried at numerous locations in northern Illinois (Goodwin, 1983). A high-quality stone at the top of the Galena is mined underground at Elmhurst in Du Page County.

The distribution of limestone outcrops and the subsequent location of quarries are functions of the configuration of the Illinois Basin. The basin, as viewed in a north-south cross section, is roughly spoon-shaped, with the bowl of the spoon

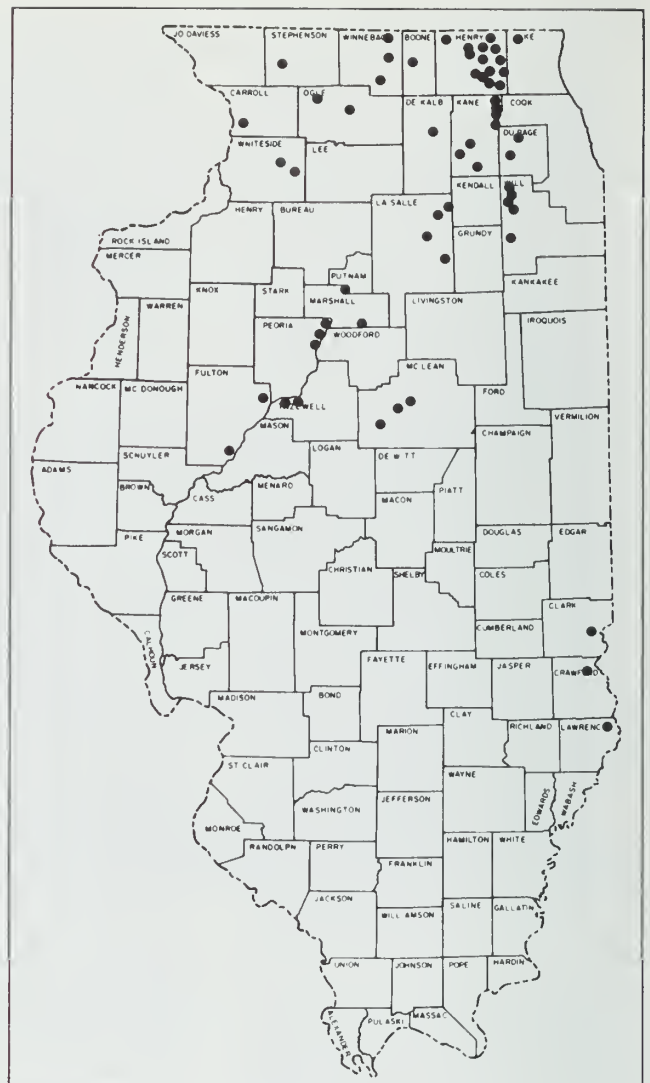


Figure 2. Sand and gravel pits.

to the south and the handle to the north (Willman et al., 1967). As the surface of the Precambrian basement gradually sank, infilling Paleozoic sediments were being deposited. The Paleozoic units dip toward the center of the Basin, and the aggregate thickness of infilling strata generally increases toward the deeper portions of the basin, as illustrated by an east-west cross section through central Illinois (fig. 5). This relatively simple picture is complicated by the major erosional unconformity at the base of the Absaroka (base of the Pennsylvanian). From south to north, progressively older stratigraphic units are present just beneath the unconformity because of beveling by post-Mississippian/pre-Pennsylvanian uplift and erosion (see north-south cross section, Willman et al., 1967). As a result, strata

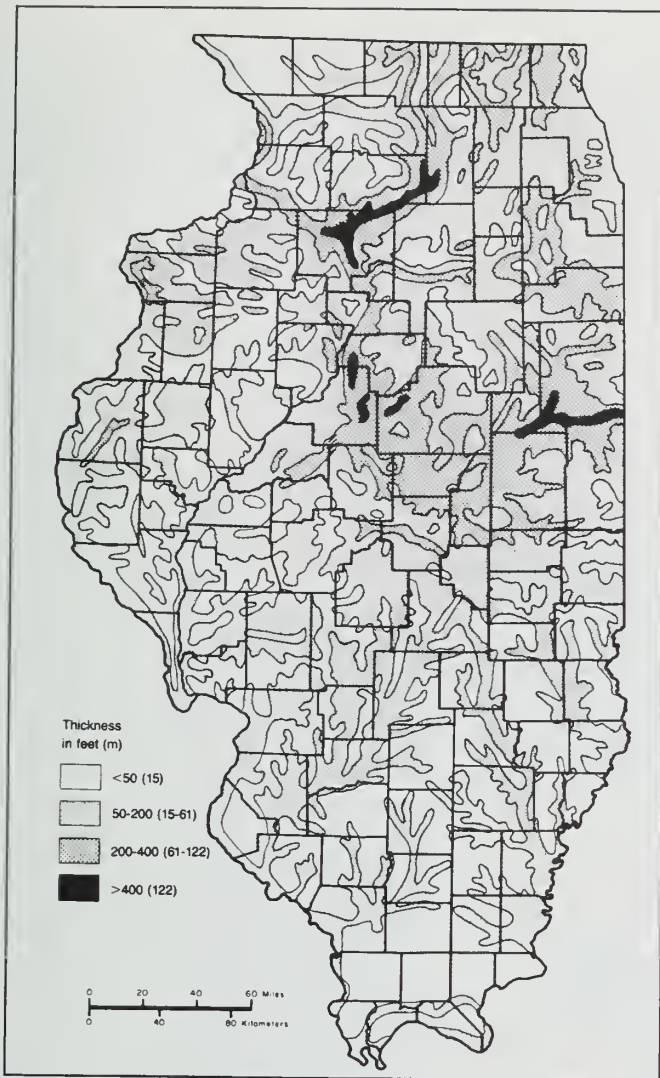


Figure 3. Simplified geologic map of surficial, unconsolidated material, Quaternary and older (from Masters, 1983; after Lineback, 1979, 1981).

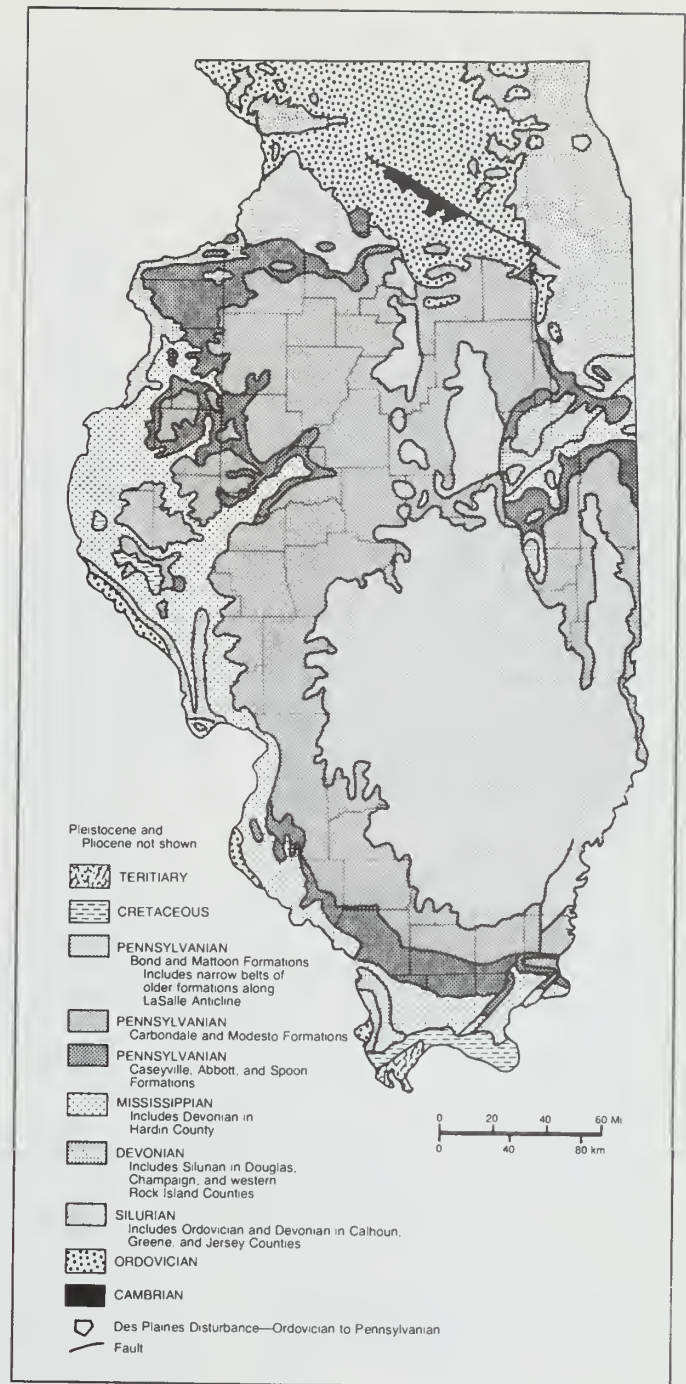


Figure 4. Simplified bedrock geologic map (from Goodwin, 1983; after Willman and Frye, 1970).

that could have been potential targets for underground mining have been partly or completely eroded in the northern part of the study area. Moreover, some carbonate units show depositional thinning and/or a decrease in quality in more shoreward positions.

### POTENTIAL TARGETS FOR UNDERGROUND MINING

An 11-county area in central Illinois that produces little or no crushed stone includes five major downstate urban areas

considered to be prime markets for aggregate: Bloomington-Normal, Champaign-Urbana, Decatur, Peoria, and Springfield (fig. 6). This central Illinois area lies within and near the northern edge of Pennsylvanian bedrock terrane generally overlain by glacial drift of variable thickness. Mississippian, Devonian, Silurian, and Ordovician carbonate rock units that supply the bulk of the crushed stone produced elsewhere in Illinois are preserved in subcrop belts either beneath Pennsylvanian bedrock or beneath the

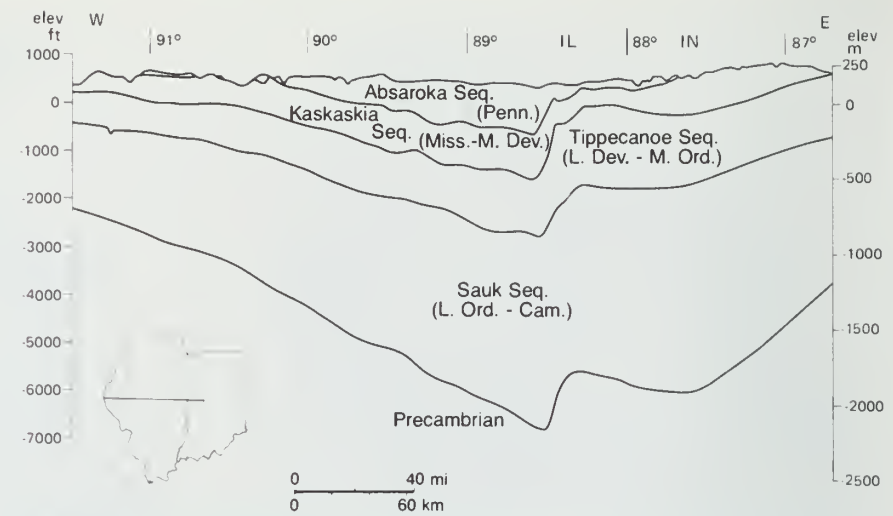
glacial drift where the Pennsylvanian is eroded, as in Champaign County.

In this paper subsurface stone resources are discussed in terms of three major "bundles" of predominantly carbonate rock units: in descending order, the Mammoth Cave, Hunton, and Ottawa Megagroups (fig. 7). The underground mining potential for each unit is considered in terms of (1) elevation within the study area with respect to mean sea level, as shown on published structure contour maps prepared by the ISGS, and (2) depth

of the unit near central Illinois metropolitan areas. Depths were calculated from the structure contour data and surface elevations from topographic maps. Surface elevations (fig. 6) generally range from 700 to 850 feet above mean sea level in the eastern part of the study area (Bloomington Ridged Plain) and from 575 to 625 feet above mean sea level to the west (Springfield Plain).

### Mammoth Cave Megagroup

Potentially minable carbonates of the Mammoth Cave Megagroup in central Illinois are essentially restricted to the Ste. Genevieve and St. Louis Limestones of late Valmeyeran (middle Mississippian) age (fig. 7). The Mammoth Cave Megagroup, commonly referred to in subsurface as the "thick Mississippian lime," includes limestones and some dolomite. The Ste. Genevieve is extensively quarried along its outcrop in southern Illinois and the St. Louis is quarried in the East St. Louis-Alton area and mined underground at East Alton. The Keokuk and Burlington Limestones (fig. 7), quarried



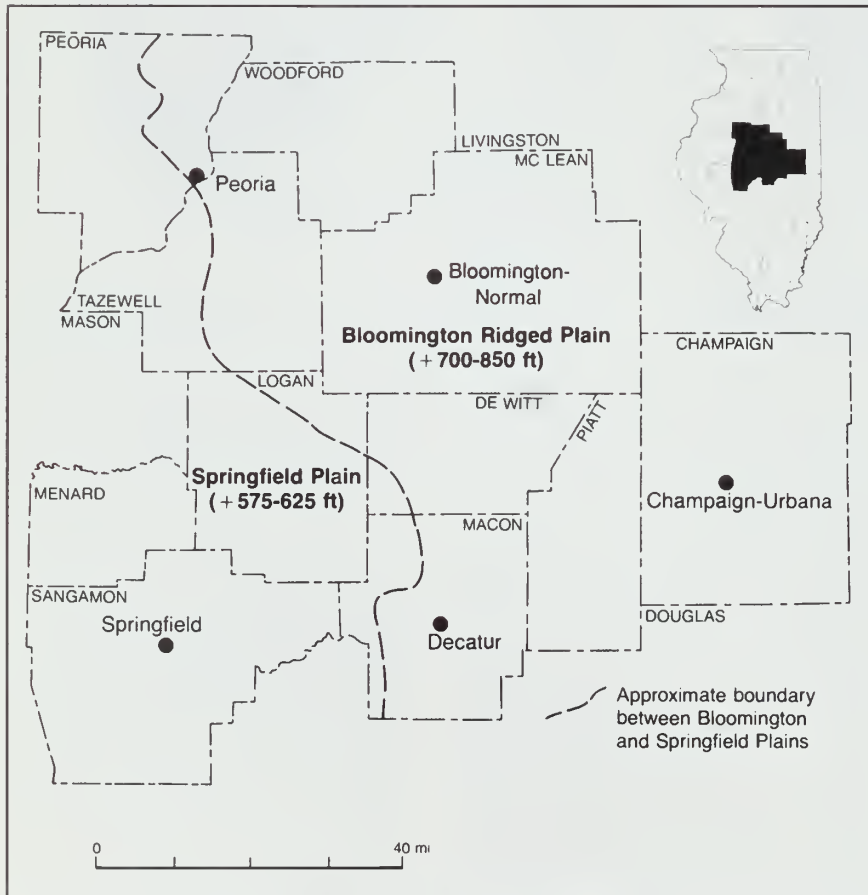
**Figure 5.** East-west geologic cross section through central Illinois showing the dip of Paleozoic strata into the Illinois Basin (Kolata and Nelson, in press).

in western Illinois, generally do not meet specifications for Class A aggregate because of softness and/or excessive amounts of chert; they are therefore not prime targets for underground mining in

central Illinois. However, the Dolbee Creek Member at the base of the Burlington is a high-calcium, soft, crinoidal limestone mined underground at Quincy in Adams County; it extends into the subsurface of the 11-county area. Preliminary study (Cloos and Baxter, 1981) indicates that chert-free limestone assignable to the Dolbee Creek is generally less than 20 feet thick; however, careful exploration may reveal minable thicknesses of chemical-grade limestone at the base of the Burlington—particularly at some localities in Sangamon County.

**Ste. Genevieve and St. Louis Limestones** The Ste. Genevieve and St. Louis Limestones are found beneath Pennsylvanian strata and beneath Chesterian (Upper Mississippian) strata in the southern portion of the 11-county area. Figure 8 shows the elevation of the top of the Karnak Member of the Ste. Genevieve Limestone, as constructed by Bristol and Howard (1976), and the northern extent of the Ste. Genevieve and St. Louis Limestones. The Ste. Genevieve is characterized by oolite that in some places is high-calcium limestone (greater than 95% CaCO<sub>3</sub>), but some parts of the formation may contain interbedded sandstone or sandy limestone. The St. Louis is predominantly dense, fine-grained limestone, commonly containing light gray to white, high-calcium stone in its upper part.

**Depth to Mammoth Cave Limestone Megagroup** The depth to limestones of the Mammoth Cave Megagroup in the study area is indicated by contours



**Figure 6.** Eleven-county study area in central Illinois.

showing the elevation of the top of the Karnak Member of the Ste. Genevieve. The top of the Karnak lies at elevations of more than 800 feet to less than 200 feet below mean sea level in the southern part of the study area, and eroded Mammoth Cave terrane extends even beyond the limits of the St. Louis. Although the physical character of the limestone beds included in the Mammoth Cave Megagroup has not been thoroughly investigated, the top minable beds of "the thick Mississippian lime" may occur at depths of 600 to 800 feet in southwestern Champaign County, 350 to 750 feet in Sangamon County near Springfield, and 800 to 1250 feet in Macon County (about 1100 feet at Decatur).

### Hunton Megagroup

The Hunton Megagroup includes carbonate rock strata of Silurian through Middle Devonian age. Figure 9 shows the elevation of the top of the Hunton down to the -1000-foot contour line (datum, mean sea level), as constructed by Stevenson and Whiting (1967).

**Middle Devonian formations** Middle Devonian formations in the study area include the Wapsipinicon and Cedar Valley Limestones and equivalent units (fig. 7). On the Sangamon Arch, (southwest corner, fig. 9), rocks of Middle Devonian age were not deposited; the depositional edge of the Cedar Valley overlaps the Wapsipinicon on the flank of the arch, but both units are absent on the crest of the arch.

To the east, in Champaign, Piatt, and Macon Counties, Middle Devonian limestone is present, but beds age-equivalent to the Wapsipinicon display an aspect more characteristic of a deep-water facies prevalent in southern Illinois and are assigned to the Grand Tower Limestone. North of the Sangamon Arch the Wapsipinicon is characterized by oolitic limestone, magnesian limestone, dolomite, and evaporites deposited in a semi-restricted to restricted shallow-water environment (James, 1968). In the areas labeled "eroded" (fig. 9), the Middle Devonian has been partly or completely removed by pre-Pennsylvanian erosion (northeast McLean County) or by a combination of pre-Quaternary and pre-Pennsylvanian erosional events (northeast Champaign County). Where Devonian strata have been completely removed, un-

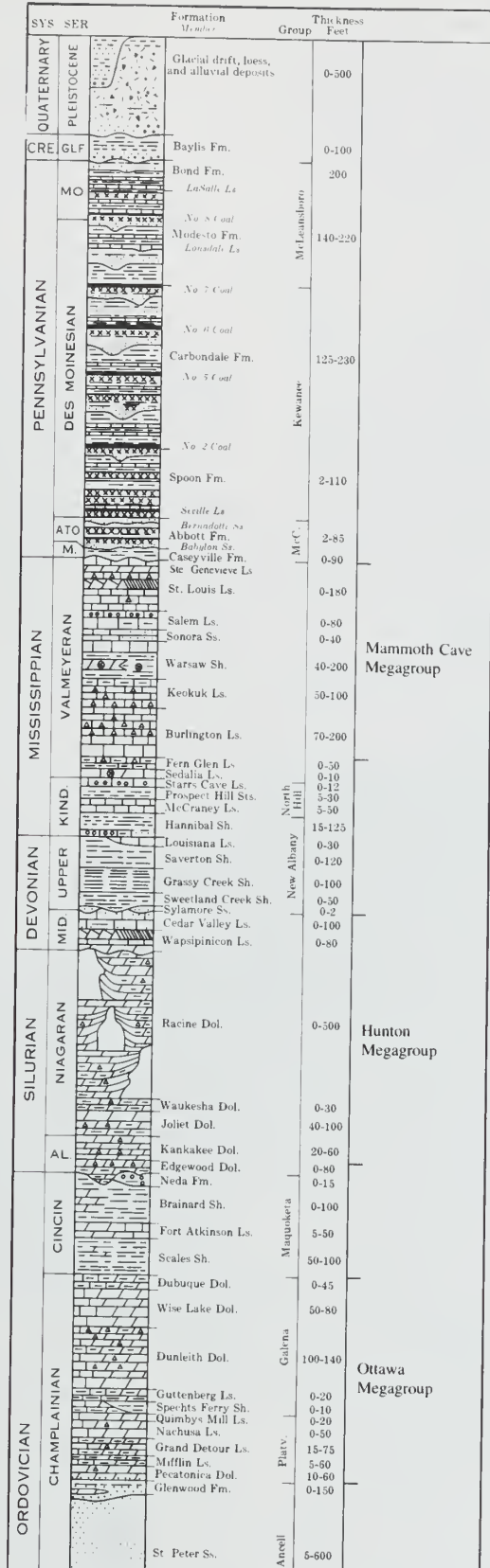
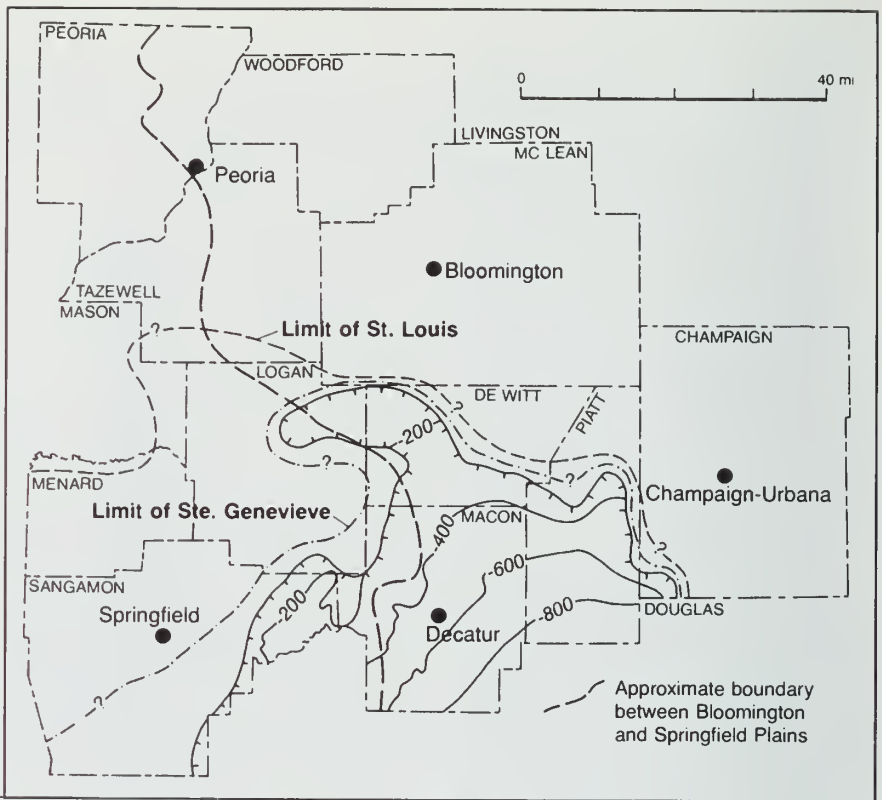


Figure 7. Stratigraphic column for central Illinois.

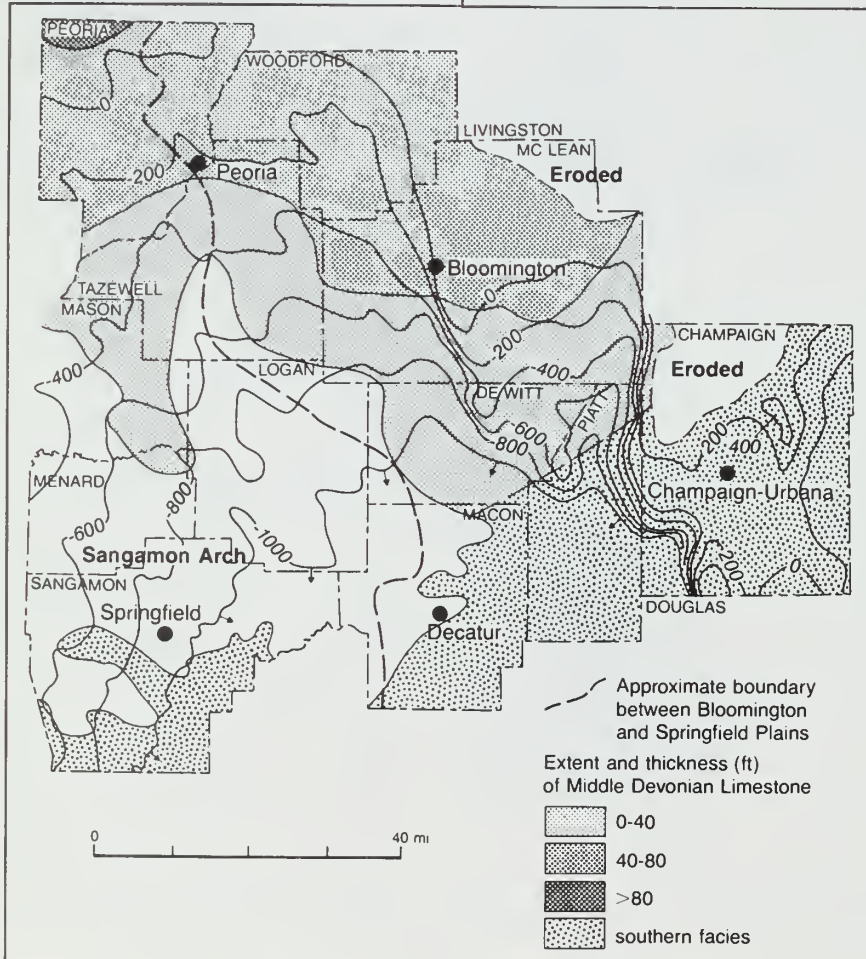
derlying Silurian formations are encountered in the subcrop.

**Silurian formations** Silurian rocks present at depth in the study area include (in descending order) the Racine Formation of Niagaran age, and the Joliet Formation, which is mostly Niagaran although its lowermost 20 to 25 feet are assigned to the Alexandrian (fig. 7). The Racine Formation, from which approximately 30 percent of the stone produced in Illinois is won, is the chief target for underground mining within the Hunton Megagroup. The Racine alone is more than 400 feet thick in southern Champaign County, but the entire Niagaran series is less than 200 feet thick in western Peoria County. Although high-purity, reef-type dolomite has not been reported in central Illinois, the carbonate rocks of Silurian age are predominantly dolomite in the 11-county area and in locations to the north. In southern Illinois limestone predominates.

**Depth to Hunton Megagroup** The depth to carbonate rocks of the Hunton Megagroup can be expected to be less



**Figure 8.** Elevation of the top of the Karnak Member of the Ste. Genevieve and extent of the Ste. Genevieve (see fig. 6) and the St. Louis Limestones in central Illinois (after Bristol and Howard, 1976.)



than 600 feet in the northwest corner of Peoria County within 15 to 20 miles of Peoria; about 450 feet in northeastern McLean County within 25 miles of Bloomington-Normal, where the Middle Devonian is eroded and the uppermost Hunton unit below the Pennsylvanian is the Silurian Racine Formation; and as shallow as 375 feet near Champaign-Urbana.

### Ottawa Megagroup

The Ottawa Megagroup comprises formations of the Galena and Platteville Groups (fig. 7, 10). Of these, the Wise Lake and Dunleith Formations at the top of the Galena (top of Ottawa Megagroup) are composed of high-purity dolomite in much of the outcrop area in northern Illinois. They are mined under-

**Figure 9.** Elevation (ft) of the top of the Hunton Megagroup, extent of middle Devonian strata, and thickness of the Wapsicinicon and Cedar Valley Limestones in central Illinois (after Stevenson and Whiting, 1967; Willman et al., 1975). Arrows along 1000 ft contour show down-dip direction.



ground at Elmhurst in Du Page County, where a high-quality aggregate is produced.

The elevation of the top of the Ottawa Megagroup (Bristol and Buschbach, 1973) is shown in figure 10. The Wise Lake and Dunleith were penetrated in a diamond-drill core hole located near Forrest, in Livingston County (fig. 10), where they are predominantly limestone and contain minable thicknesses of high-calcium limestone (Baxter, 1964). The Galena and underlying Platteville are known to grade laterally from limestone in the south to dolomite in the north in Illinois. This transition, with respect to the total Ottawa Megagroup, takes place within the 11-county area (fig. 10); however, the precise character of the limestone-to-dolomite transitions within the Galena and Platteville Groups individually has not been accurately determined.

The Galena—the top of the Ottawa Megagroup—occurs at depths calculated to be as shallow as 1175 feet within 20 miles of Champaign-Urbana. However, this location is on a structure that is now the site of a natural gas storage project, and the availability of the Ordovician subsurface for underground mining is, at best, questionable. The top of the Galena lies at a similar depth in the northeastern corner of McLean County near Bloomington-Normal and is perhaps as shallow as 1000 feet northwest of Peoria.

### FACTORS FAVORING UNDERGROUND MINING

According to Loofbourow (1983), a combination of favorable conditions is needed to support underground mining operations:

- a rather stable market of at least moderate size
- an accessible, large, rather uniform rock mass, tending to be dry, in which sizable, stable openings can be made
- a "kicker"

Kickers may be factors contributing to high costs of quarrying stone at or near the surface within a given market area. They may include high costs associated with acquisition of land, stripping, waste disposal, reclamation, or other operational or environmental problems. On the other hand, the operative kicker may be some positive factor related to the proposed underground operation, such as

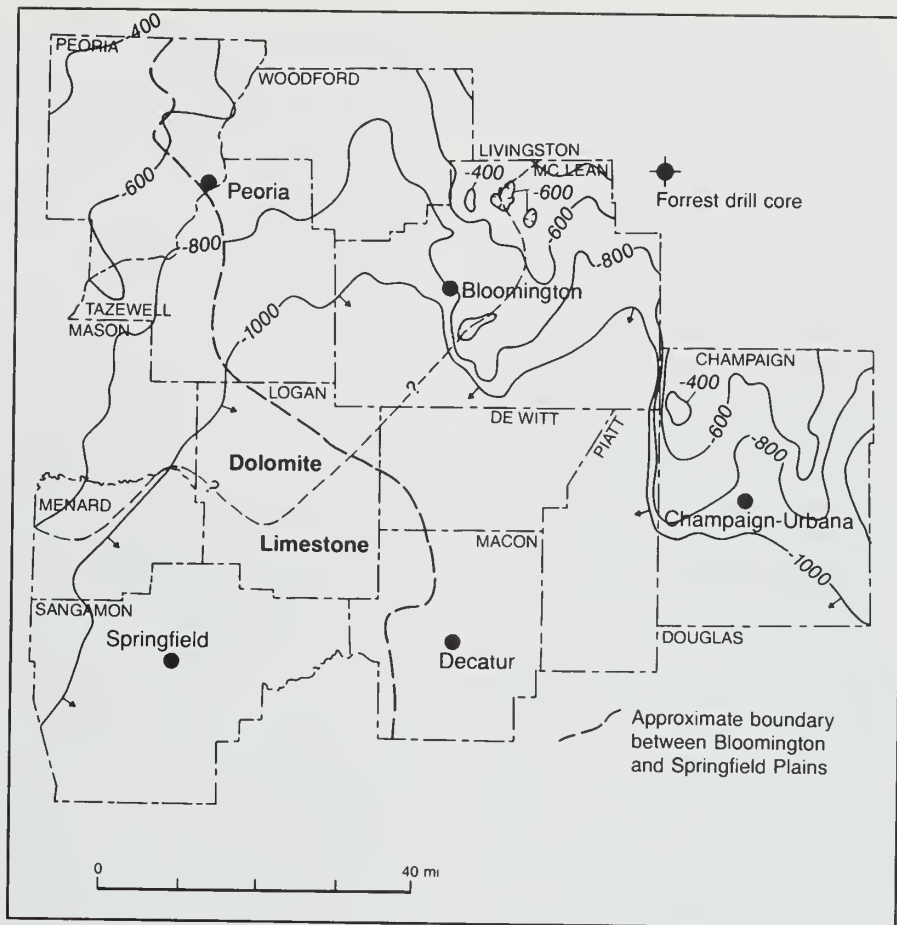


Figure 10. Elevation (ft) of the top of the Ottawa Megagroup (top of the Galena Group) in Central Illinois (see fig. 6) (after Bristol and Buschbach, 1973).

the availability of underground sites nearer the market area than potential surface sites are; an opportunity to negotiate a royalty lease as opposed to incurring a large acquisition cost; uniformly mild underground working conditions; and subsequent use of the underground space created by mining. Although space accessible only by means of vertical shafts would have less potential value for underground storage than would space with horizontal access, permanent income from such uses as energy accumulation (pumped storage for off-peak generation of hydroelectricity), disposal, clarification of water or air, and other potential uses should be considered.

More detailed examination of the depth to minable stone, the chemical and physical characteristics of that stone, and the extent to which the foregoing factors are operative near central Illinois communities is warranted.

A growing need exists for stone that meets the chemical specifications re-

quired for use in flue gas desulfurization, fluidized-bed combustion technology, and other chemical and industrial processes. Such products demand premium prices, and the ability to produce a chemical-grade stone in addition to high-quality aggregate near the major central Illinois markets would increase the odds for the success of an underground mining operation.

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# Model of Construction Aggregates Demand and Supply: A Chicago Area Case Study

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## ABSTRACT

*Construction aggregates—sand and gravel and stone—represent a \$400 million industry in Illinois. Because of their low unit value, construction aggregates are usually sold within 50 miles of the production site. Local economic conditions and demographic factors are thus closely associated with the aggregates industry. In addition, national economic factors and political decisions can influence the construction industry and affect aggregates production. If future demand for aggregates could be successfully predicted, the industry and the economy as a whole would benefit from minimized price fluctuations.*

*In Oregon, Ontario, and California, econometric models were developed to predict aggregates production. The results of the modeling indicated that regional conditions differ enough to warrant development of regional models. The investigation reported in this paper was the first attempt to develop such a model in Illinois. Because two-thirds of the Illinois population is concentrated in the Chicago area, that region was selected as the study area. Econometric models based on demographic and economic information on the six counties in the Chicago area were developed for the projection of future aggregates production. The models indicate that population, employment, gross state product, and mortgage interest rates are the most significant factors affecting the production of aggregates. Short-term projections made with the help of the models were within 5 percent of actual production figures. However, the statistical reliability of the models is low, and the correlations cannot always be explained logically. Further research is essential: detailed community-level data gathering, especially on public construction projects, could improve the models significantly. A comparison with similar attempts at modeling in the United States and Canada indicates that the difficulties encountered in this investigation are not unique to Illinois.*

variations in the reliability of projections, even for individual regions of states. Although statewide models are more desirable, experience has shown that targeting smaller areas within states may be more promising. The present investigation is the first attempt in Illinois to model the demand for construction aggregates.

The Chicago area is defined here to include the six counties of Cook, Du Page, Kane, Lake, McHenry, and Will. This area was selected for study because about two-thirds of the Illinois population is concentrated here; about 45 percent lives in Cook County alone. Currently about 50 percent of the state's stone and sand and gravel production (excluding industrial sands) comes from the six-county Chicago area. Most of the stone used in this area comes from quarries in Cook and Will Counties (fig. 1); however, sand and gravel pits are widely distributed (fig. 2) (Samson and Masters, 1987). Most stone quarries are in northwestern Illinois counties and most sand and gravel pits are in the central parts of northern Illinois, generally several times farther from the Chicago area than are the current suppliers to the Chicago area.

The most densely populated parts of the Chicago area have few pits or quarries because urban sprawl is encroaching on existing and potential quarry sites and reserves are becoming depleted (Mikulic and Goodwin (1986). If the urban sprawl continues, construction aggregates may have to be transported from greater distances; this could drastically affect delivered prices and lead to higher construction costs. The average free-on-board (f.o.b.) mine value of sand and gravel is currently about \$2.80 per ton and that of crushed stone is about \$4.00 per ton. These prices can be virtually doubled within 20 miles of the mine site and tripled at 50- to 60-mile distances (fig. 3).

## INTRODUCTION

Construction aggregates (sand, gravel, and stone) are materials of low unit value that are generally used within 30 to 50 miles of their places of extraction because of shipping costs. The demand for these materials is associated with private and public construction and therefore varies considerably with location. In addition, the construction industry itself is often subject to influences arising from national economic factors such as interest rates or growth in the gross national product, and political decisions with respect to publicly financed construction such as national highways or community roads. Finding ways to predict future demand

for aggregates is a challenging task because of the complex factors (at both local and national levels) associated with aggregates demand. Yet accurate predictions would enable the industry to respond quickly and efficiently to changes in demand and avoid excessive price fluctuations and shortages.

Attempts have been made in Oregon, Ontario, and California to construct econometric models for projecting future demand for aggregates (Friedman, Niemi, and Whitelaw (1979); Matten (1982); and Stinsen, Manson, and Plappert (1983). In all cases, modeling on a regional basis was more successful than statewide modeling, and there were wide

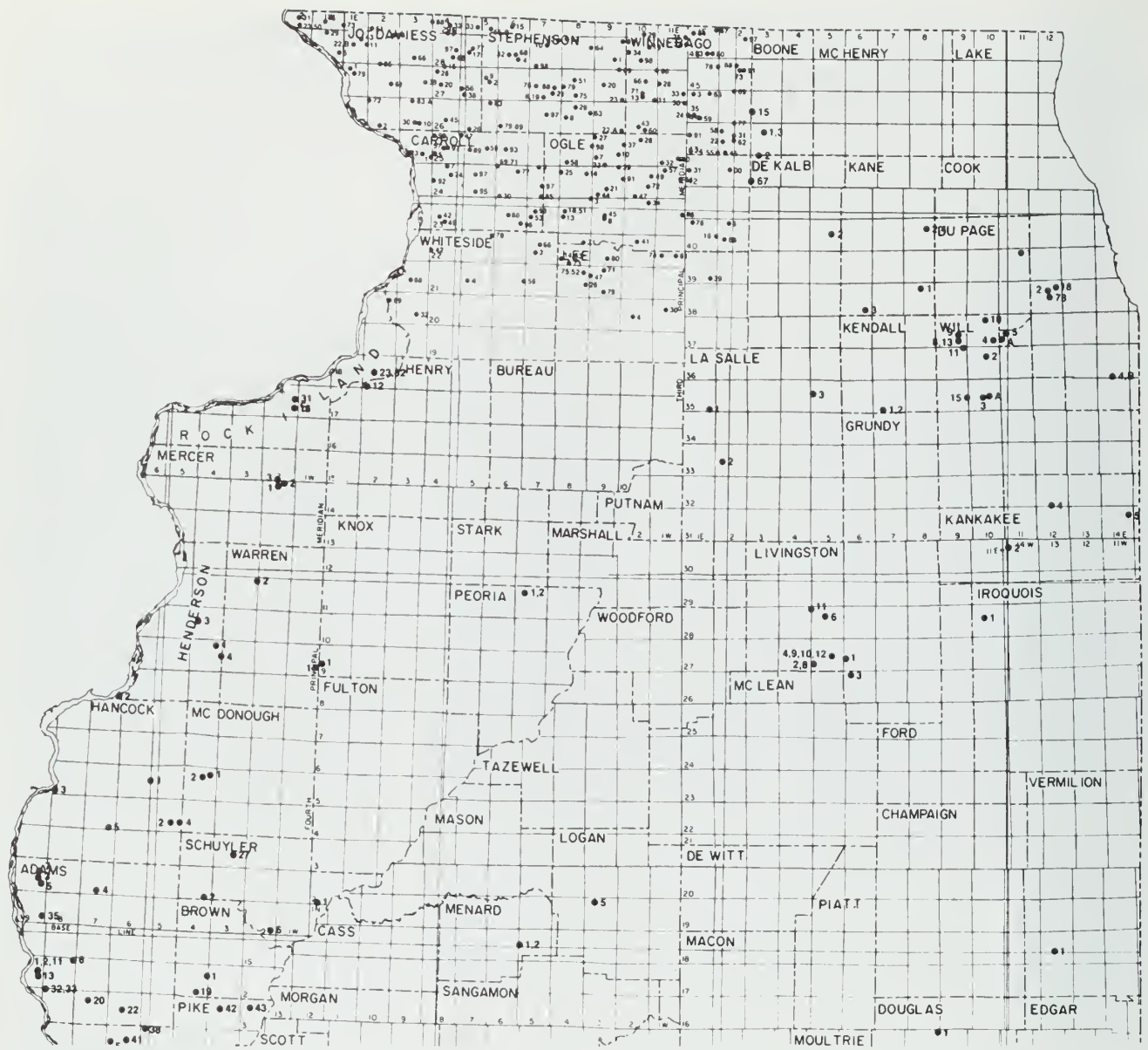


Figure 1 Stone quarries in northern Illinois.

**AGGREGATES PRODUCTION**

From 1970 through 1984 the production of aggregates in the Chicago area fluctuated greatly. Recessions in 1972, 1975, and 1982 affected production severely. The downturn in production since 1980 has been more severe and has lasted longer than any other in the past 15 years (fig. 4). Individual counties differed significantly in their production. McHenry and Kane Counties have been the largest producers of sand and gravel. In Will County, sand and gravel production declined sharply after 1973 and generally remained as low as production in the other Chicago area counties (fig. 5).

Cook and Will Counties produce the most stone; Kane and Du Page Counties produce some stone, and Lake and McHenry Counties (fig. 6) produce only negligible amounts.

Accessibility of the available geologic resources has played an important role in the aggregates industry of the Chicago area. Masters (1978) mapped the sand and gravel deposits of northeastern Illinois, which are located primarily in McHenry and Kane Counties. Bradbury (1977) compiled a map of the dolomite resources in the Chicago area, and his map indicates the concentration of minable stone deposits in Cook and Will

Counties. Urban expansion in these counties has constrained production; consequently, future supplies may have to be shipped from as far as 50 to 80 miles, representing a major cost-escalation factor.

**DEMOGRAPHICS**

From 1970 to 1984 the total population of the six-county Chicago area grew steadily from about 6.99 million to about 7.22 million (Office of Real Estate Research, 1984). This growth occurred outside Cook County; the Cook County population declined from about 5.50 million to about 5.17 million. Since 1980 the population in Cook County appears to

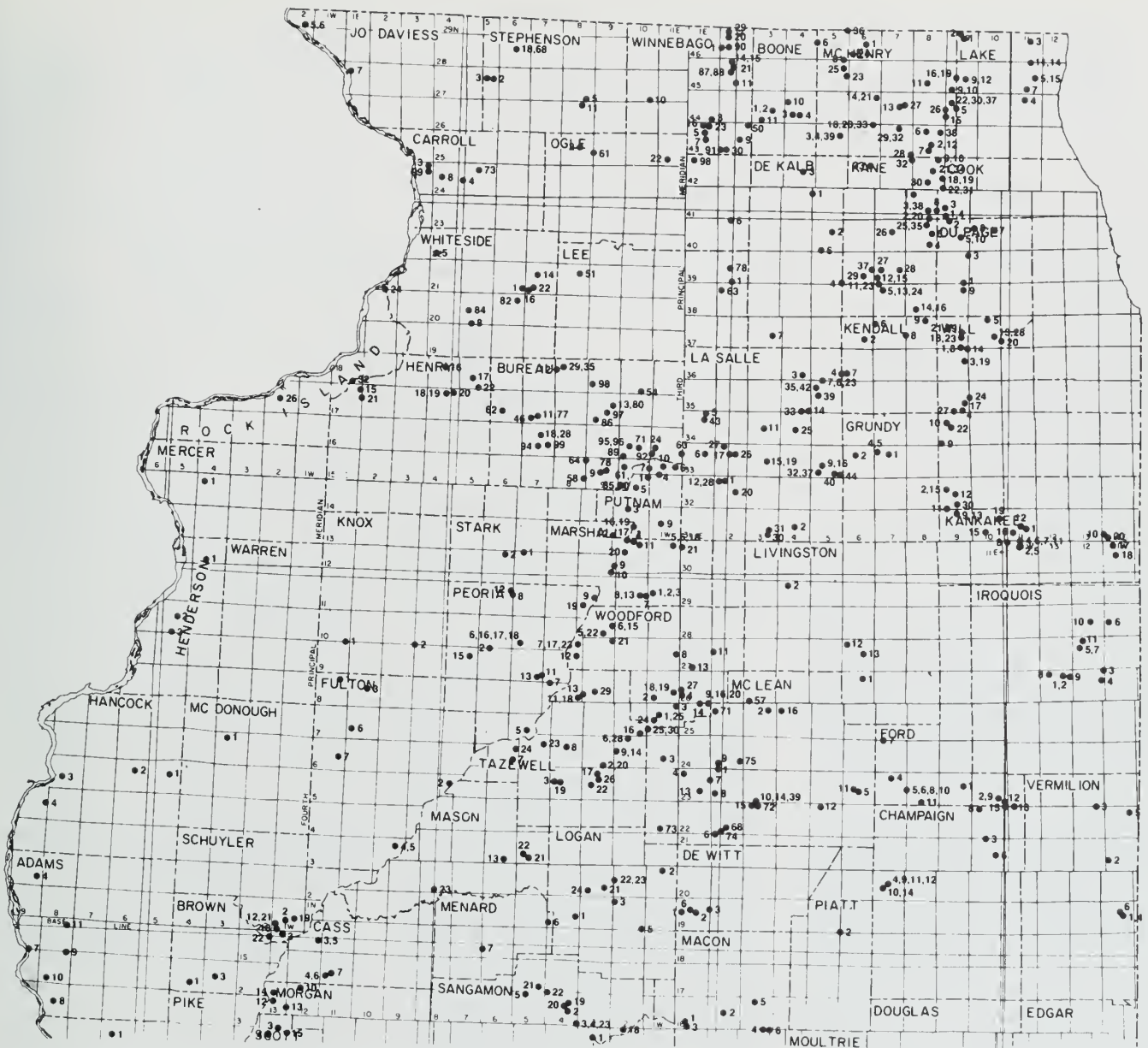


Figure 2 Sand and gravel pits in northern Illinois.

have stabilized at about 5.2 million; the outlying counties continue to grow.

Employment opportunities in the outlying counties have drawn people from Cook County as well as counties outside the area. Figure 7 indicates the rapid employment growth in Du Page County and continuing high employment in Lake County. Figure 8 shows the fluctuating but essentially stagnant employment situation in Cook County. Because of the size of its population, Cook County dominates the employment trend in the six-county area—a trend showing a rising but widely fluctuating employment pattern (fig. 9). Thus, economic activity in

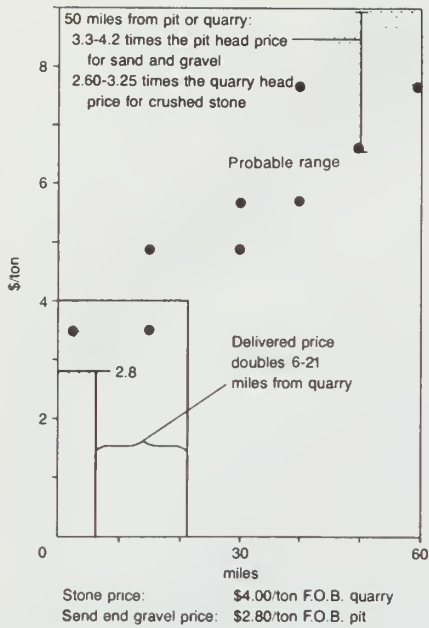
the area depends strongly on demographic and economic conditions in Cook County.

### CONSTRUCTION ACTIVITY

The economic activity linked most directly with the aggregates industry is construction, both private and public. National data on the number of building permits issued, construction completed, and value of construction in place are compiled by the U.S. Bureau of the Census, which uses a sampling technique rather than actual data gathering. At a county level only private construction data are available. Public expenditures by

federal, state, county, and local governments on roads, highways, and other public projects are not compiled by any single agency at the county level. The following analysis therefore excludes public construction. The omission presumably distorts Cook County data more than data from other counties. On the basis of national census data, an estimated 10 to 15 percent of the total construction activity in the Chicago area is publicly financed and thus not included in the analysis.

The value of private construction in place in the Chicago area (in constant 1982 dollars) fluctuated between a 1978 high of about \$4.4 billion and a 1982 low



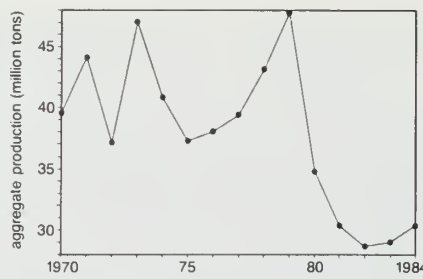
**Figure 3** Cost of transporting aggregates by truck.

of about \$1.3 billion (fig. 10). The 1970-1984 trend in construction value in the Chicago area has been downward (about 33%). The downward trend has been most pronounced in Cook County and least noticeable in Du Page County.

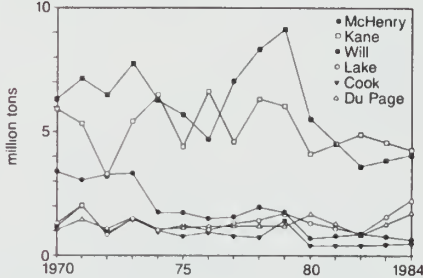
### FACTORS AFFECTING AGGREGATES PRODUCTION

All sand and gravel and most stone produced in the Chicago area are used in the construction industry; therefore, the following factors—which significantly affect the construction industry in this area, also affect the aggregates industry in the Chicago area.

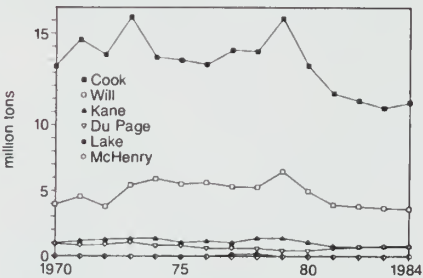
- sand and gravel production (millions of tons per year)
- stone production (millions of tons per year)
- value of private residential buildings in place (billion dollars, 1982)
- value of private nonresidential buildings in place (billion dollars, 1982)
- value of public construction projects in place (billion dollars, 1982)
- population (millions)
- employment (millions)
- personal income (billion dollars, 1982)
- inflation-adjusted mortgage interest rates (percent)
- year-to-year change in gross state product (percent)



**Figure 4** Production of construction aggregates in the Chicago area.



**Figure 5** County sand and gravel production.

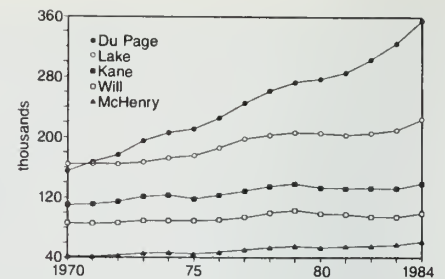


**Figure 6** Stone production by county.

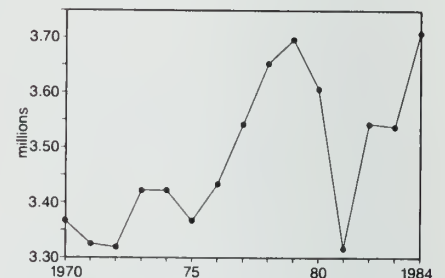
Countywide data on production, population, employment, and income were available; however, data on public spending on construction projects in the Chicago area were available only for highway projects financed or managed by the Illinois Department of Transportation (IDOT). Therefore, the statistical analysis for this study was conducted without information on non-IDOT public spending.

### ECONOMETRIC MODEL

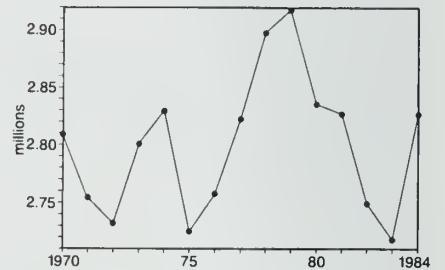
Comparison of figures 4, 9, 10, and 11 suggest that employment, value of construction in place, changes in the GSP, and the real mortgage interest rates may be correlated with the production of construction aggregates in the Chicago area. Stepwise regression on 1970-1984 data, using a 90 percent confidence level and the t-statistic, indicated that total production of sand, gravel, and stone was significantly correlated with the value of construction in place and the real mortgage interest rate ( $R^2 = 73\%$ , standard



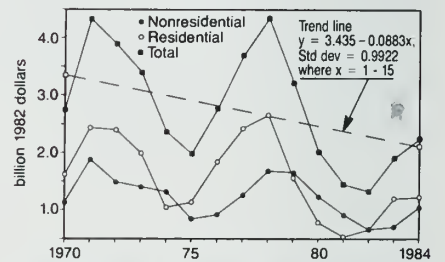
**Figure 7** Employment by county.



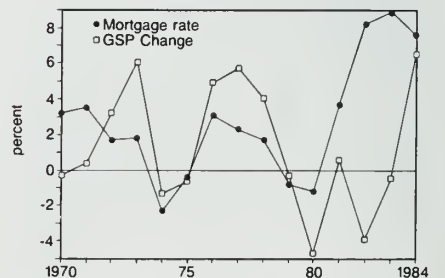
**Figure 8** Cook County employment.



**Figure 9** Chicago-area employment.



**Figure 10** Chicago-area construction value.



**Figure 11** Illinois mortgage rate and GSP changes.

**Table 1** Statistically significant factors affecting aggregates production in the Chicago area, 1970-1984.

Model number	Dependent variable	Intercept	Population (million)	Employment (%/yr)	Change in GSP (%/yr)	Mortgage rate (% real)	Adjusted R <sup>2</sup>	Standard error	Determinant
1	Sand and gravel production (million tons/yr)	377.215	-58.361	15.203	0.302	—	64.907	1.992	0.568
2	Stone production (million tons/yr)	23.041	—	—	0.287	-0.753	63.575	1.902	0.983
3	Total aggregates production (million tons/yr)	40.553	—	—	0.628	-1.294	47.163	4.558	0.983

error = 3.26) as in the following econometric model:

$$\text{Aggregates production} = 28.189 + 4.034 (\text{value of private and IDOT-financed construction}) - 0.903 (\text{real mortgage interest rate})$$

The usefulness of this model is problematic, however, because values for construction in place are available only after the fact, and estimates are available only 6 months in advance. To avoid this difficulty, attempts were made to construct models without using the value of construction in place. The three models resulting from that analysis are presented in table 1.

These models indicate that individual projections for sand and gravel and stone production would be more reliable ( $R^2 = 64$  or  $65$ ) than projections of total aggregates production ( $R^2 = 47$ ); however, model 1 cannot be used for sand and gravel because of the negative correlation between population and production, which cannot be explained without more research. Models 2 and 3 were used to project for 1985 and 1986 stone and total aggregates production in the Chicago area (table 2). As table 2 indicates, the 1985 projected figures are 3 to 4 percent above actual production figures. By inference, sand and gravel production for 1985 can also be estimated as the difference between the two projections in table 2. The 1986 projections for stone are 6 percent below U.S. Bureau of Mines (U.S.B.M.) estimates, and projections of total aggregates production are 10 percent below U.S.B.M. estimates. As expected, accuracy of the projections declines as attempts are made to project production farther into the future.

Models 2 and 3 unexpectedly revealed that changes in GSP and mortgage inter-

**Table 2** Projections for 1985 and 1986 stone and total aggregates production in the Chicago area (millions of tons/year)\*.

	1985		1986	
	Projected	Actual*	Projected	Actual*
Stone	18.2	17.5	18.8	20.2
Total aggregates	32.5	31.5	35.0	36.4

\*Actual production figures include estimates for sand and gravel in odd-numbered years and for stone in even-numbered years.

est rates have more effect on the construction industry than do such factors as population and employment. Given the negative correlation with population in model 1 and the generally low  $R^2$  values, extreme caution is warranted in using the models. Researchers in Oregon, Ontario, and California encountered similar difficulties, which probably stem from inaccuracies in available production figures and other data, as well as from the lack of detailed data on public construction projects at all levels of government.

#### FUTURE RESEARCH NEEDS

The results of this pilot study indicate that the available database is not sufficient for developing reliable econometric models for future projections of construction aggregates production. To improve results of future studies the following points should be considered:

- County-by-county data on public spending on highways, roads, and other facilities must be compiled, and projected spending plans must be available.
- Usage factors for sand, gravel, and crushed stone must be developed for various kinds of construction (e.g., homes, office buildings, roads, and highways) in each county. Geologic occurrence (location and depth) and

quality of these aggregate resources affect their use in different types of construction; therefore, different independent variables may have to be considered in each county or region.

- The study regions should be chosen so as to include the centers of economic activity and all the major satellite communities contributing to that activity. In addition, counties supplying construction aggregates to the study regions must also be included.

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# Industrial Sand in Indiana\*

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## ABSTRACT

About 1900, as the result of rapidly growing foundry and glass industries, Indiana ranked second among the states in industrial-sand consumption. Only about a third of the industrial sand consumed was produced from in-state sources, but these Indiana sources were diverse. They consisted of Silurian sandstone near Logansport, Devonian sandstone near Pendleton, Mississippian sandstone near the Ohio River in south-central Indiana, Pennsylvanian sandstone from various places in southwestern Indiana, and Quaternary dune sand near Lake Michigan. As specifications for various uses became more stringent, only those operators with high-quality sand or sandstone could compete with out-of-state sources. Today, industrial sand is produced in Warren, Porter, and LaPorte Counties for foundry use and in Harrison County for glass, chemical, and miscellaneous uses.

Sand with a high-silica content from the Bethel Formation (Mississippian) in southeastern Indiana has been used intermittently since the late 19th century for glass, foundry, filter, and other uses. In 1985, the CARD Industrial Sand Corporation built a new processing plant near New Albany at the site formerly occupied by the Indiana Glass Sand Company and the Ohio River Silica Corporation. This new operation mines hydraulically and pumps the sand slurry about a quarter of a mile to a processing area with a deslimmer, attrition mill, and hydrosizer. After air drying, the sized sand is further dried in a rotary kiln. The attrition mill removes a large part of the iron oxide coating on the sand grains and provides a silica product with a  $Fe_2O_3$  content of about 0.025 percent. The main industrial-sand markets for this product are within 200 miles of New Albany.

## INTRODUCTION

Indiana has had a flourishing industrial-sand industry. If we could plot production of the state's industrial sand through time (which is impossible because of inadequate records), our curve would probably approximate the curve traditionally shown for the production of a nonferrous-metal deposit (fig. 1). In this hypothetical example, the number of mines, or production, of a nonferrous metal goes through a youthful phase of rapid development following initial discovery. Production stabilizes during the mature stage and then declines slowly through an old-age phase until the deposit is depleted or until the grade of the ore is too low to be economically produced.

This hypothetical example applies when price, demand, and other variables are constant; however, if either the price or demand changes—for instance, because of a war or the discovery of new

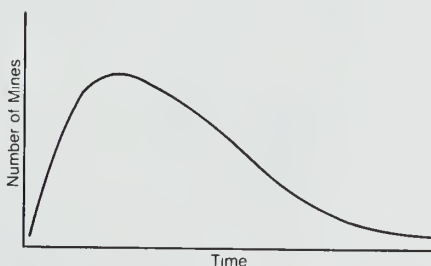


Figure 1 The life cycle of a nonferrous-metal deposit (modified from Peters, 1966, and Skinner, 1979).

deposits—the curve will be modified. The decline in production of a nonferrous-metal deposit is largely due to a depletion of ore, but the decline in production of an industrial minerals deposit such as industrial sand is more likely to result from other causes.

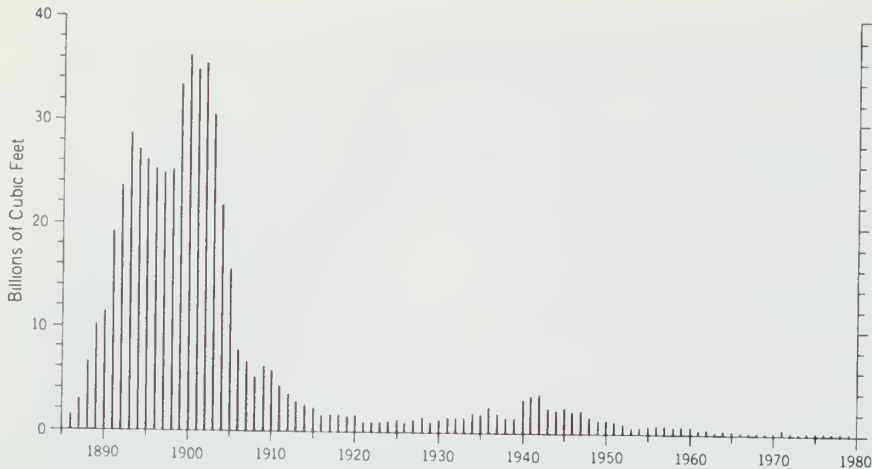
Production of industrial sand in Indiana probably followed the typical curve: rapid increase in production and then a slow decline. The reason for the rapid increase was not a new discovery of industrial sand but the discovery of a cheap source of locally available fuel that permitted the growth of industries using industrial sand. And the reason for the decline was not the depletion of deposits but the new, rigid specifications being set by the glass manufacturers and foundries that put producers of inferior industrial sands out of business.

This paper includes a brief history of industrial-sand production in Indiana, a review of the geology of sands suitable for industrial purposes, and a discussion of the new CARD Industrial Sand Corporation plant near Elizabeth, which seems destined to change the industrial-sand production curve for Indiana.

## PRODUCTION HISTORY

The most abrupt sociologic and economic change that has occurred in the state began in 1886, when gas was discovered in the Trenton Limestone (Ordovician) in northeastern Indiana. This discovery of oil and gas in the Trenton Field—two years after oil had been discovered in the Lima field in Ohio—was the beginning of the petroleum industry in Indiana. The Trenton Field was among the largest fields of its time, and even by today's standards its areal extent ranks it as one of the giant fields in the world. The shallow depth of the gas-producing zone (less than 1000 feet) and the low drilling and completion costs brought a rapid in-

\* This paper includes information pertaining to CARD Industrial Sand Corporation and industrial sand in Indiana in 1987 at the time the 23rd Forum on the Geology of Industrial Minerals was held. The CARD plant has since closed its operation. The reason for its closing is unknown.



**Figure 2** Estimated gas production in Indiana, 1886-1973; all production for 1886-1915 is from the Trenton Field (from Carpenter and others, 1975).

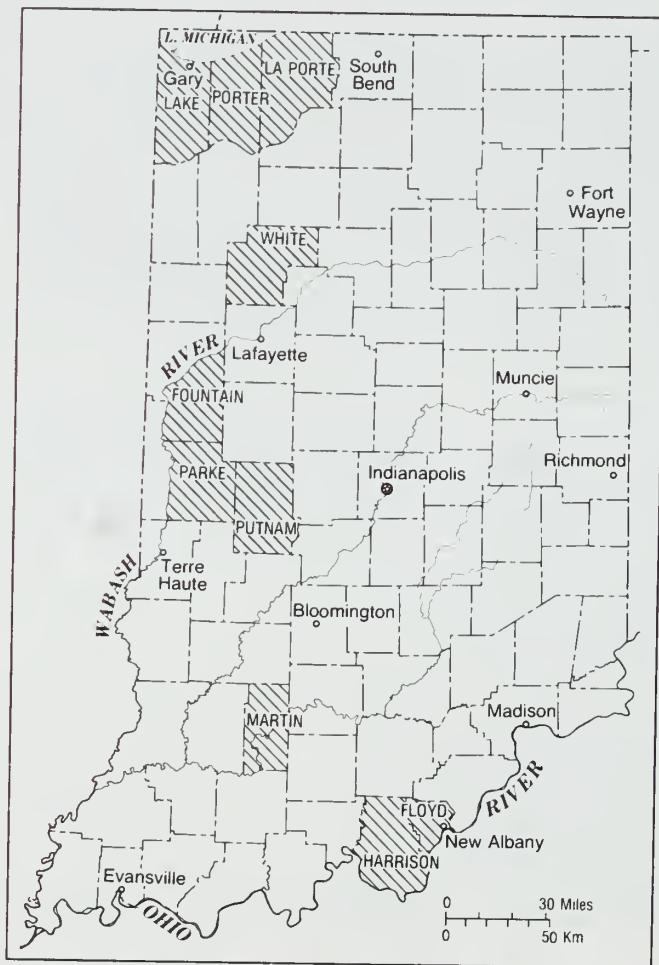
crease in production (fig. 2). But with cheap costs came rapid and uncontrolled development.

Most people in Indiana seemed to believe that gas supplies were unlimited, and it was not uncommon for wells to go uncapped or be burned in giant flambeaus. Some towns used gas for street lights, and some cities, to entice industry from the east to Indiana, advertised in newspapers that gas was free. These inducements led to an influx of diverse industries, and the glass and foundries industries created a large demand for industrial sand.

The lack of understanding of reservoir properties and the absence of a centralized regulatory body to control drilling and production practices led to widespread resource waste and depletion. The production of Trenton gas declined rapidly after 1902. The production curve for Trenton gas and the production curve for industrial sand in Indiana have a similar configuration, which indicates a close relationship between them. As the source of low-cost fuel became rapidly depleted and foundries and glassworks closed, the demand for sand declined. The rigid specifications imposed by the foundries and glassworks that remained in business further reduced the demand for sand.

At the turn of the century, Indiana ranked second in the nation in glass production with 71 glassworks in operation (Burchard, 1907). With such a demand for glass sand, many local sources were tried and many, although inferior by today's standards, were used. Before 1922, sand for glass manufacture was produced in ten counties (fig. 3), but

despite these diverse sources, Indiana producers supplied only one-third of the state's needs. The heyday of glass-sand production was short lived. By 1987 only Harrison County in southern Indiana was producing glass sand.



**Figure 3** Indiana counties that produced glass sand before 1922 (data from Burchard, 1907; Barrett, 1914; and Logan, 1922).

Foundry sands were also in demand at the turn of the century. Specifications for foundry sands were less stringent than those for glass manufacture, and foundry sands were produced in as many as 36 counties (Logan, 1922); however, by 1918 only 27 counties were producing foundry sands (fig. 4). In 1926, Indiana ranked second among states in molding-sand production (Logan, 1930). Just as happened in the glass industry, the loss of an inexpensive source of fuel and an increase in restrictive specifications for foundry sand caused a decline in the number of producers. In 1987, only Fountain, Harrison, Porter, and LaPorte Counties had foundry-type sand production.

The U.S. Bureau of Mines reported that in 1986 about 190,000 tons of industrial sand, valued at \$1.3 million, was produced in Indiana. Current sand production comes from Manley Brothers of Indiana, Crisman Sand Company, Harrison Steel Castings Company, and

CARD Industrial Sand Corporation (fig. 5). Manley Brothers and Crisman are mining Quaternary dune sands, and Harrison Steel Castings and CARD are producing from Paleozoic sandstones of fluvial and submarine-channel origins.

## GEOLOGY AND CURRENT PRODUCTION OF DEPOSITS

### Quaternary sand

Although unconsolidated deposits of sand and gravel are abundant throughout northern Indiana, sands suitable for industrial purposes are less well distributed. The most abundant accumulations occur in northwestern Indiana as dune deposits in several ancestral beach ridges along Lake Michigan. The two most lakeward complexes of dune and beach sediments, the Calumet and Toleston Beaches, have been mined for industrial purposes and are currently being mined by Crisman Sand Company (Calumet Beach) and

Manley Brothers of Indiana (Toleston Beach) (fig. 6).

The Calumet Beach began to form about 11,800 years BP (Hansel et al., 1985) at about 30 feet above current lake level (580 feet). This lake phase was established at an altitude of 610 feet above mean sea level following transgression of the southern shore of ancestral Lake Michigan. The supply of sediment to the southern shore of Lake Michigan was great enough to cause progradation of the shoreline and stacking of nearshore sediments over deeper water deposits (Thompson, 1987). Therefore, the dune sands are in the upper and landward part of the dune and beach complex and commonly overlie beach sand and gravel.

The Toleston Beach began to form about 7000 years ago at the end of the next large-scale rise in lake level (Hansel et al., 1985), when the lake level stabilized at about 600 feet. Throughout its history, the Toleston shoreline has

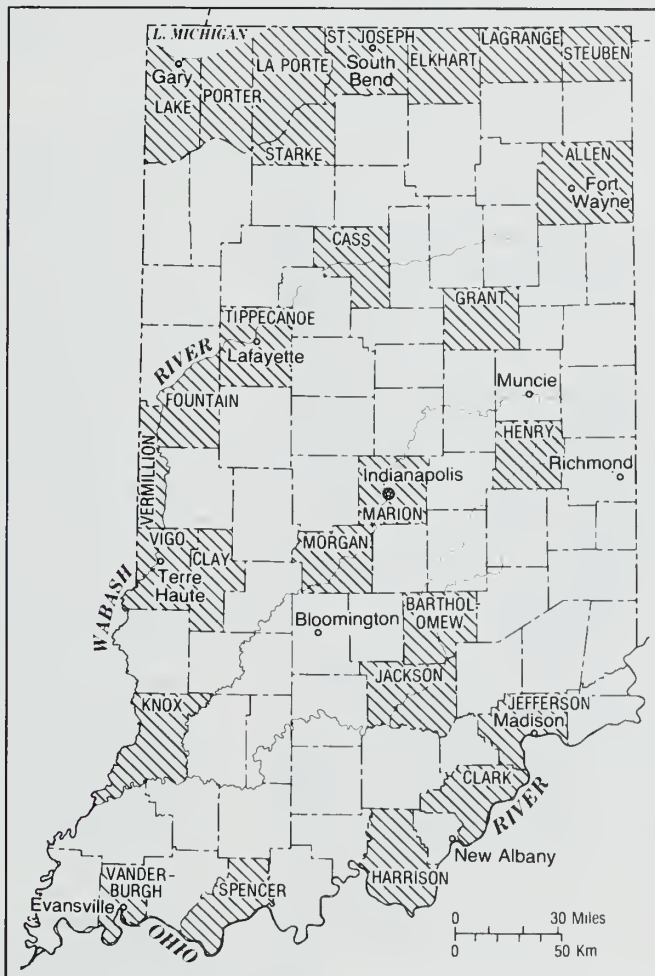


Figure 4 Indiana counties that produced foundry sand in 1918 (data from Logan, 1922).

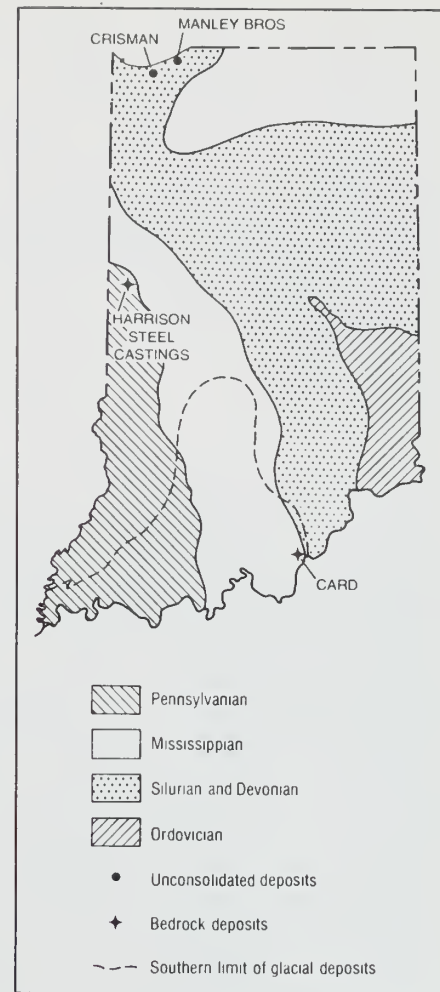
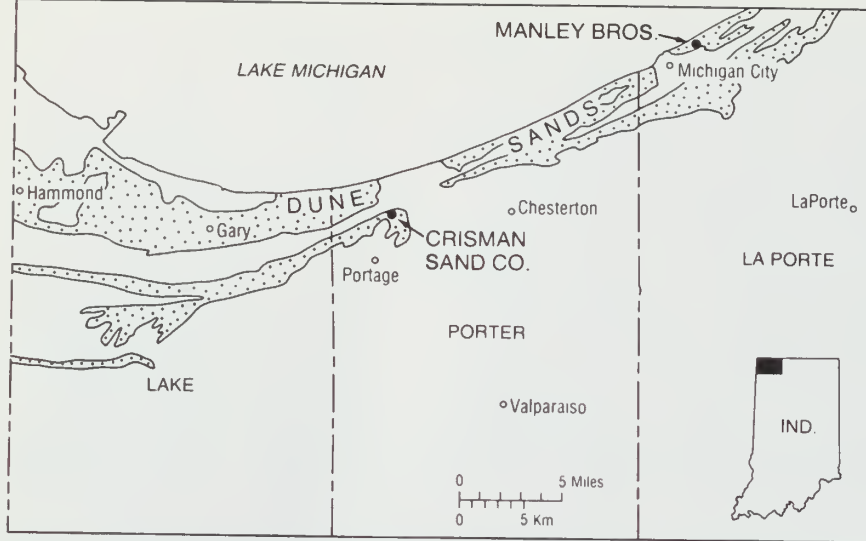


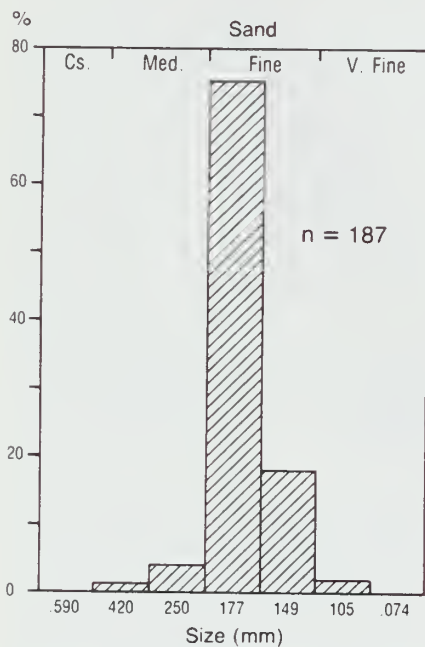
Figure 5 Locations of plants producing industrial sand in 1987, and major bedrock units in Indiana.

prograded in the western part of the Indiana Dunes and has produced more than 100 low-relief beach ridges in the Gary and Hammond areas. The Toleston shoreline in the eastern part of the Indiana Dunes has remained relatively stable, and therefore a thick sequence of dune deposits has formed on top of the early Toleston Beach deposits in the Michigan City area (Thompson, 1987) (fig. 6). In the eastern dune area, the elevation of the tops of the Toleston and Calumet dunes is about the same, although the Toleston dunes are more extensive.

The dune sands are fine grained, well rounded, and moderately to very well sorted (fig. 7). Commonly, a sequence of dune sands fines slightly upward. In general, the Toleston dune sands are slightly finer grained than the Calumet dune sands.



**Figure 6** Major sand deposits in northwestern Indiana and locations of operations of the Manley Brothers of Indiana (Toleston Beach) and the Crisman Sand Company (Calumet Beach).



**Figure 7** Histogram showing size distribution of Quaternary sands in northwestern Indiana and number of samples analyzed.

Dune sands are composed of more than 90 percent quartz in many places (table 1), but in some places 25 percent of the sand may consist of feldspar and heavy minerals. The compositional variations of the sand fraction among beach complexes is not great. Sand produced by the Crisman Sand Company has a relatively high clay content, which gives it natural molding-sand characteristics. This sand is older than the material produced by Manley Brothers, which may explain its higher silt and clay content. The Calumet sand produced by Crisman has had a longer time to develop a soil profile, and the soil probably contributes to the clay content of the dune.

The Crisman Sand Company is a small corporation that mines by front-end loader and ships both bulk and bagged products by truck. The product is used mainly as a blast-furnace runner sand. Manley Brothers is owned by British Industrial Sand. The sand is hydraulically mined and pumped to large towers, where

it is discharged to large conical piles for air drying; further drying is by kiln. The sand is transported by both truck and rail. The product is used for various foundry purposes.

### Pennsylvanian sandstones

About one-third of the Pennsylvanian rocks in the Illinois Basin are sandstones, and several of these sandstones have potential for use as industrial sand (Murray and Patton, 1953; Carr, 1971). Those most widely mined in Indiana are sandstones of the Mansfield Formation—the lowermost Pennsylvanian formation in Indiana—which crop out at the surface from Warren County to the Ohio River. They have been used for refractory brick, glass, and foundry purposes, but are currently being used only for foundry sand.

The thickest Mansfield sandstones are fine to medium grained and are fairly well sorted (fig. 8), but relatively pure coarse-grained sandstones and pebble conglomerates are found in places. The crossbeds are thick and dip unimodally toward the southwest, indicating the alluvial origin of the sandstones (Potter, 1963).

The composition of 48 samples of Mansfield sandstones averaged 96 percent silica (table 1). Carbonate minerals are few to nonexistent in the Mansfield. Silica content can be increased by running the sand through a log washer to remove the clay and silt.

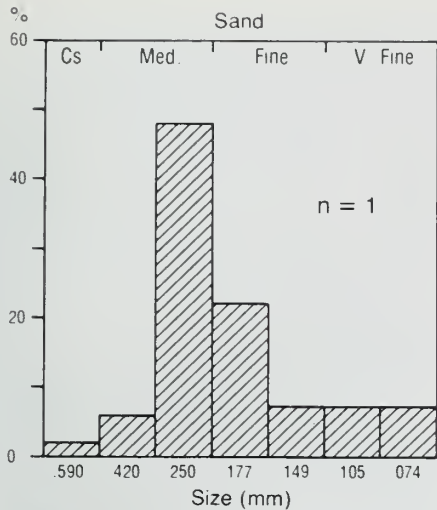
The Harrison Steel Castings Company quarries Mansfield sandstone for its own uses as foundry sand. The sandstone is mined in two benches, each about 12 feet high, by drilling and blasting. The sand is carried by front-end loader to the plant, where it is crushed and sieved to pass a 3/16-inch screen. The oversize is discarded. The sand is sized by a wet classifier and blended with appropriate binders for use in the foundry.

### Mississippian sandstones

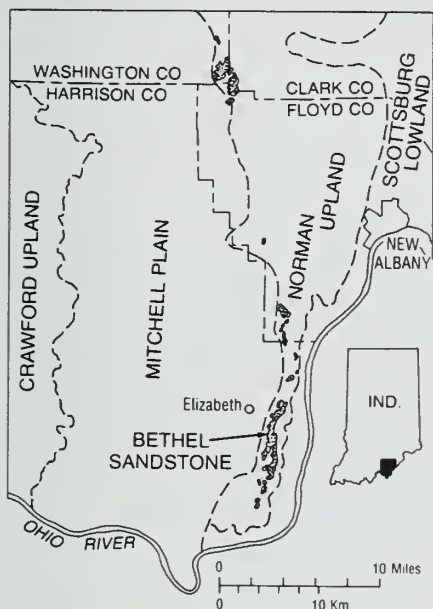
Mississippian sandstones used for industrial sand are restricted to those of the Chesterian Series. Sandstones of the Chesterian constitute about 25 percent of the total Chesterian section (Potter, 1963). Although several of these sandstones have potential for use as industrial sand, sandstones of the Bethel Formation in south-central Indiana have been most extensively used for this purpose.

**Table 1** Chemical analyses of Quaternary sands and Mansfield and Bethel Sandstones.

Deposits	Average chemical analyses content (%)				
	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaCO <sub>3</sub>	MgCO <sub>3</sub>
Dune Sand, Manley Brothers	91.9	3.80	0.53	2.5	0.9
Dune Sand, Crisman Sand Co.	86.7	5.00	1.40	5.3	1.7
Mansfield Sandstones	96.3	2.60	0.66	0.1	0.1
Bethel Sandstones	97.8	1.42	0.39	0.1	0.1



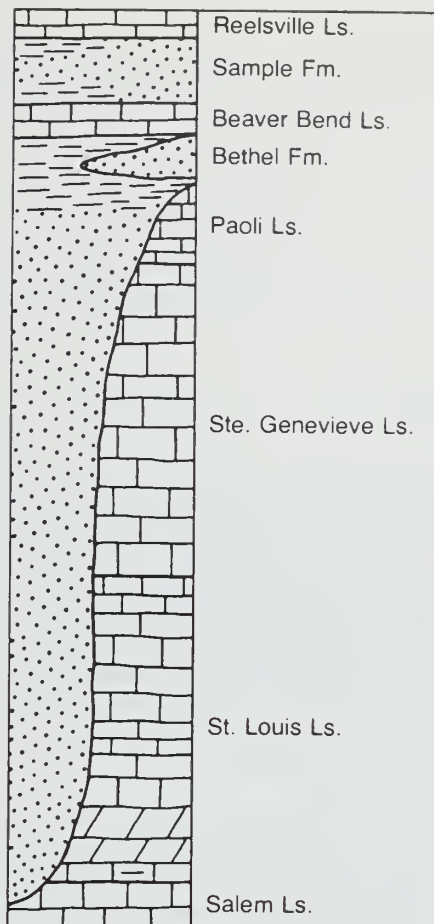
**Figure 8** Histogram showing size distribution of a sample of Mansfield sandstone collected from the quarry of the Harrison Steel Castings Company and number of samples analyzed.



**Figure 9** Distribution of Bethel sandstones along the Norman Upland.

Scattered deposits of poorly consolidated sands forming a linear pattern in areas of the Norman Upland of Washington, Clark, Floyd, and Harrison Counties (fig. 9) were called the Ohio River Formation in 1903 (Ashley and Kindle, 1903). Historical evidence indicates that the deposits have been mined for some time; they were being used to supply a plate-glass operation in New Albany in 1874.

Various theories have been proposed to explain the origin of the sand deposits (Wayne, 1960). A study published in 1969 (Sedimentation Seminar) interpreted the sands as remnants of an extensive sub-



**Figure 10** Stratigraphic section of rocks of Mississippian age showing relationships of the Bethel Formation to underlying and overlying rocks in south-central and western Kentucky. After Sedimentation Seminar, 1969.

marine-channel fill system that extended southward from Indiana into Kentucky. In Kentucky this sand has been called the Tip Top Sand and the Bethel Formation. The term "Bethel" is now used mostly for this sand body in both Indiana and Kentucky. The sand has been mined for many industrial uses in several places, mainly Washington, Harrison, and Floyd Counties; it is mined now in only one location, near Elizabeth in Harrison County.

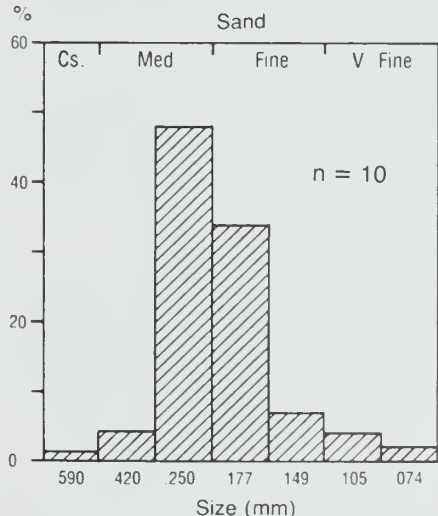
In southern Indiana, the Bethel sand deposits lie on the Salem and St. Louis Limestones (Mississippian), but as the sandstone body is traced south into Kentucky the sand occupies positions higher in the section (fig. 10). About 25 miles south of the Ohio River in Kentucky, the normal stratigraphic sequence is found where the Bethel overlies the Paoli Limestone.

The sandstone is white to brownish red, and because of poor consolidation it

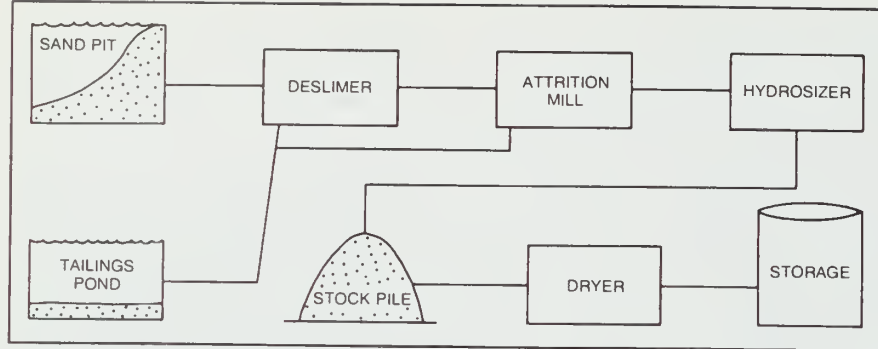
is subject to rapid erosion (in places a miniature "badlands" topography develops). The sandstone is generally 40 to 50 feet thick but is 80 feet thick in places in Harrison County. It is fine to medium grained and moderately well sorted (fig. 11), and the sand grains are round to angular and poorly cemented. Most of the iron oxide in the sand occurs as a film around individual grains. Analyses of more than 30 samples indicate an average silica content of 97.8 percent (table 1) and a clay content of 1 to 2 percent. There are few carbonate minerals.

In 1985, CARD Industrial Sand built a new processing plant near Elizabeth at the site of a former operation (built in 1964) that was known as the Indiana Glass Sand Company. Indiana Glass Sand operated until the early 1970s, when it was sold to the Ottawa Silica Company. The name of the plant was changed to the Ohio River Silica Company and the plant was operated until about 1976, when it became inactive until rebuilt by CARD.

At the CARD operation the sand is mined hydraulically, pumped to temporary storage, and then processed through a deslimmer (log washer) to remove the clay, silt, and very fine sand fraction (fig. 12). The sand is next processed through an attrition mill to remove iron oxide coatings from the sand grains and then through a hydrosizer to separate out the very coarse sand from the medium- and fine-sand fraction. The waste from the deslimmer and attrition mill



**Figure 11** Histogram showing size distribution of samples of Bethel sandstones from Washington, Harrison, and Floyd Counties and number of samples analyzed.



**Figure 12** Steps used by the CARD Industrial Sand Company to process Bethel sandstone near Elizabeth, Harrison County.

is carried to a tailings pond for disposal, and the processed sand is taken to stockpiles for air drying. In the final step of processing, the sand is dried in a rotary kiln and stored in silos.

Although the sand processing is simple, it is effective. By removing the fine clay and silt fraction in the log washers and the iron oxide coating on grains in the attrition mills, a final product can be obtained. The sand has characteristics that make it suitable for many industrial uses; it is currently being produced for foundry, glass, stone sawing, abrasives, and chemical uses.

Industrial-sand deposits are typical of industrial-minerals deposits whose market value depends largely on the location of the deposit. Although specifications for industrial sand are very precise, sand is still bulky, heavy, and expensive to transport. The CARD operation faces competition from companies producing silica sand in the surrounding states of Illinois, Michigan, and Ohio, but the quality of the sand, location of the deposit, and favorable production costs allow the company to be competitive within about 200 miles of its plant near New Albany.

#### SUMMARY

Production of Quaternary dune sands in northern Indiana is limited because of restrictive land uses and strong competition from nearby producers in Illinois and Michigan. Although production of Penn-

sylvanian sandstones in southwestern Indiana should continue to serve local markets, production costs will make these sands less attractive in larger markets. The Bethel sandstones in south-central Indiana are high in quality, are located close to markets, and can be produced competitively. The CARD Industrial Sand operation has added a youthful phase to an otherwise old-age industry in Indiana.

#### ACKNOWLEDGMENTS

We appreciate the help and friendly cooperation of Frederick C. Ward, CARD Industrial Sand Corporation; Edwin Nicholson, Crisman Sand Company; and Butch Swift, Harrison Steel Castings Company.

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# Depositional Environments of Natural-Aggregate Reserves, Osseo District, Twin Cities Metropolitan Area, Minnesota

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## ABSTRACT

Gravel-bearing sand (natural aggregate) has been mined from the Osseo district for about 45 years, and it seems likely that production from the district will continue to the year 2000 and beyond.

In map view, the natural-aggregate bodies occur as regularly spaced, single to compound pods ranging from 5 to 80 acres in area. In vertical section the bodies are from 35 to 80 feet thick, changing downgradient from rod shaped to lens shaped. They are composed of a series of tabular, gravel-rich sand layers 1 to 10 feet thick, containing gravel particles in imbricate structures. They are underlain by sandy to silty clay lacustrine sediments, have steep, saw-toothed interfaces with either sandy or silty clay lacustrine sediments or sand or sandy loam diamict sediments and are overlain by 5- to 25-foot-thick layers of sand to sandy loam diamict and/or silty sand lacustrine sediments.

During its meltback in late Wisconsinan time, the Des Moines Lobe, St. Croixan Association glacier ice reached a series of stillstands at high points within the Osseo district. During ice melting at each of the stillstands, meltwater streams carried freed rock materials from upgradient regimes within tunnels and open channels to the highly fractured, lobate ice margin where, depending upon the amount of head loss, they were deposited as stream-channel and as fan and lake-floor facies within an ice-front, glacial lake. Episodically, for considerable lengths of time, high-volume meltwater flows (from superglacial lakes) carried, as bed-load and by saltation, pebble-, cobble-, and minor boulder-size rock particles and deposited them as tunnel/channel and proximal deltaic eskerine facies. In later stages of meltback, meltwater streams eroded deep channels in the natural-aggregate bodies and filled them with distal, gravel-poor channel-facies sands.

Shallowly underlying the St. Croixan natural-aggregate bodies are pre-late Wisconsinan natural-aggregate bodies that were probably formed within depositional environments similar to those described for the St. Croixan bodies.

## INTRODUCTION

### Location and extent

The Osseo natural-aggregate mining district encompasses about 1800 acres in the northwest part of the Twin Cities metropolitan area in Minnesota (fig. 1). The district is readily accessible from the metropolitan freeway network.

### Production history

Natural aggregates have been produced from 33 pits, ranging from 5 to 65 acres in size, within the Osseo district (fig. 2). Production began before World War II but accelerated during the con-

struction boom that followed the War. As many as 30 operators were active in the district in the late 1940s; currently there are five commercial operations in the district and two other operations managed by state and local government agencies.

### Quality of the aggregate

The physicochemical quality of the aggregate materials from the Osseo district is very high. The coarse-aggregate particles (+2 mm particles) have a lithologic composition (Meyer and Jirsa, 1984) of about 60 percent crystalline rocks, 17 percent carbonate rocks, and 22

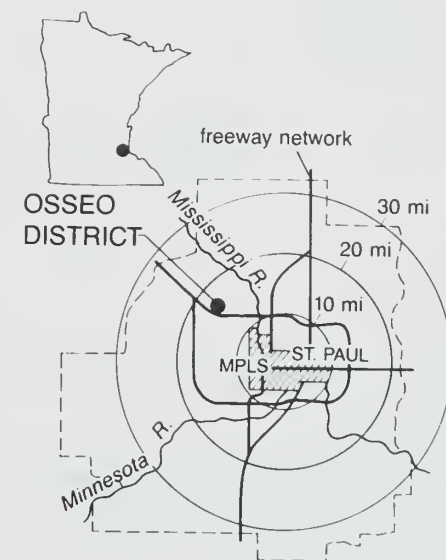


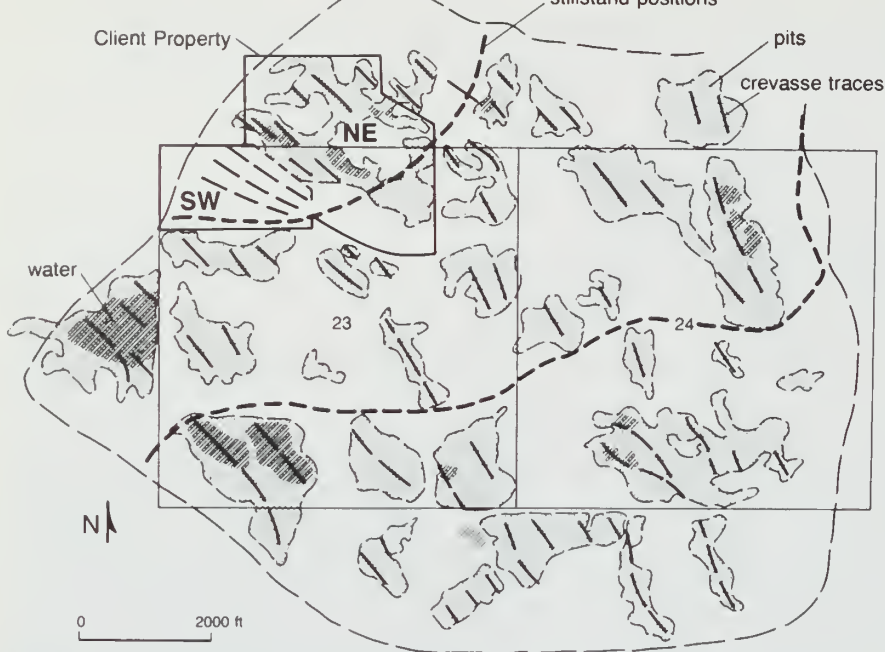
Figure 1 Location map, Osseo district, Minneapolis-St. Paul (Twin Cities) metropolitan area.

percent other sedimentary rocks. The aggregate materials contain less than 1.0 percent shale and less than 0.2 percent chert; their mean Los Angeles Rattler (LAR) value is 14.8.

### Procedures

The stratigraphy of the natural-aggregate and bounding glacial sediments was determined from pit-bank examinations and drilling information on the 150-acre client property (fig. 2). Drilling information consisted of descriptions of the sediment returns from, and rate of bit penetration within, 13 auger borings (fig. 3); each was 4 inches in diameter and 50 to 80 feet deep.

We utilized previously gathered pit-bank descriptions and subsurface data for a reconnaissance study of the district. In addition, vertical, black and white aerial photographs (scale, 1:20,000) were viewed stereoscopically to determine the limits of the active, active-on-demand and abandoned pits. Trends of topo-



**Figure 2** Location of natural-aggregate pits and the client property, Osseo district, and stillstand zones of tunnel esker depositional environment.

graphic depressions from the 10-foot contour-interval Osseo quadrangle map (U.S. Geological Survey, 1967) were used in the interpretation of traces of glacier-ice fissures.

## STRATIGRAPHY

### Overview

The natural-aggregate resources occur as rod- to lens-shaped bodies, 5 to 80

acres in areal extent and 35 to 80 feet thick, underlain, bounded, and overlain by lacustrine and diamicton sediments and crosscut by meltwater sediments.

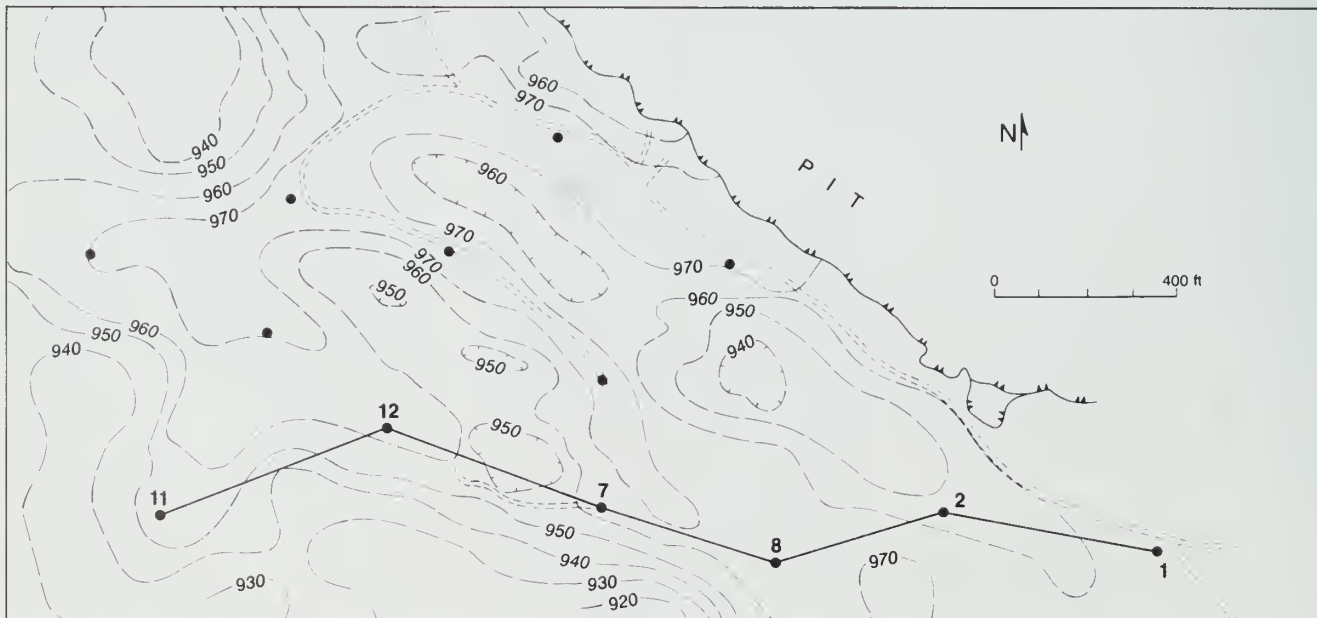
Figure 3, a preliminary map of the topography of the 8.3-acre southwest part of the client property, shows the location of 13 auger borings drilled there. Figure 4 depicts a longitudinal vertical section through a series of southeast-trending

ridges of the southwest part of the client property, and shows the sequence and type of natural-aggregate materials and their relationships to bounding glacial sediments.

The sediments depicted in figure 4 were deposited from two late-Wisconsinan Stage (Hobbs and Goebel, 1982) glaciers. All but the capping sediments are of the St. Croixan moraine association of the Superior lobe (less than 20,500 years BP, Wright, 1972). The capping diamicton and lacustrine sediments are of the Pine Citian moraine association of the Des Moines lobe, the youngest of the Wisconsinan glacial lobes to advance into the Twin Cities lowland (Wright, 1972). About 200 to 300 feet of pre-Superior lobe glacial sediments underlie the St. Croixan sediments and overlie an erosional surface developed upon the Late Cambrian Jordan Sandstone (Jirsa, Olsen, and Bloomgren, 1986). Bedrock valleys as deep as 100 feet (Bonestroo, Rosene, Anderlik, and Associates, 1985) are reported immediately northeast of the client property (fig. 2).

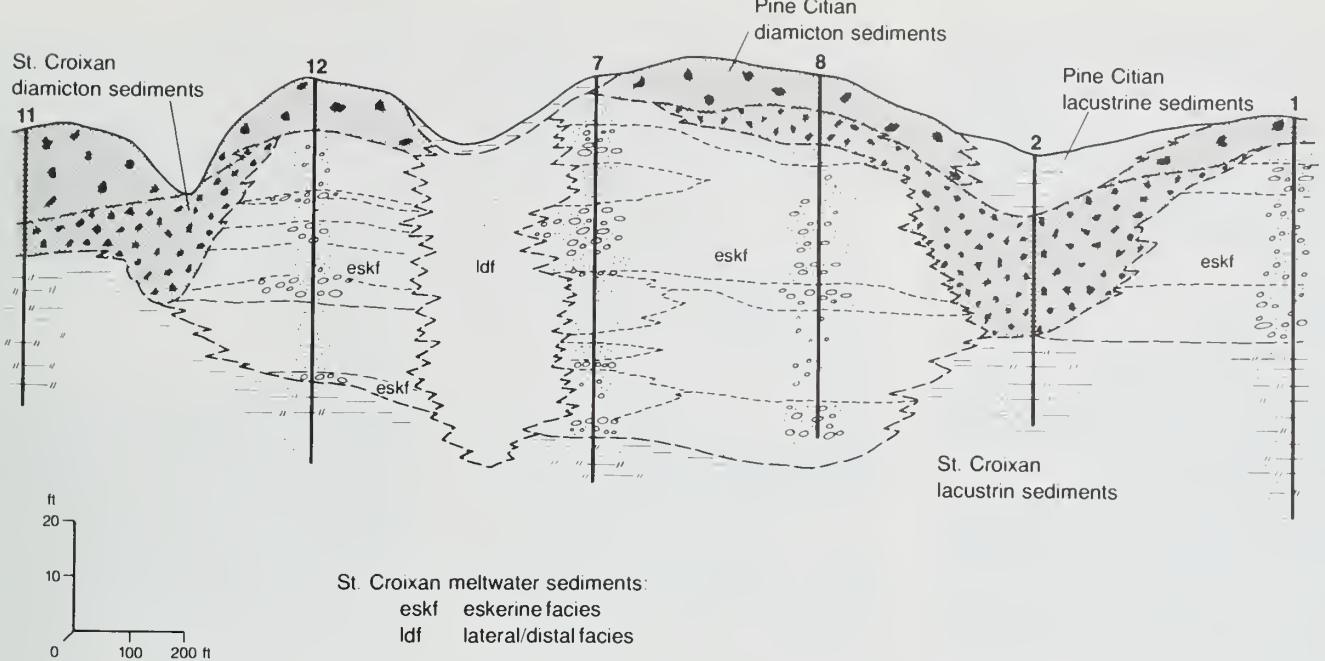
### St. Croixan sediments

**Lacustrine** Up to 33 feet of St. Croixan lacustrine sediments underlie and bound the natural-aggregate bodies (fig. 4). The upper surface of the lacustrine unit has a maximum local relief of about 30 feet beneath the southwest part of the client property; maximum slopes are about 3°. These sediments, in-



**Figure 3** Topographic and boring-location map of southwest part of client property, Osseo district.





**Figure 4** Longitudinal vertical stratigraphic section through southeast-trending natural-aggregate ridges, southwest part of client property, Osseo district.

terpreted to be of fan- and lake-floor facies, range from sandy loam to silty loam (after USDA, in Hunt, 1972, p. 226)—i.e., mostly of medium-size sand within a matrix of 5 to 15 percent silt- to clay-size mineral and rock particles. Pebble-bearing dropstone layers interrupt the lacustrine sequences in some localities.

**Meltwater** The St. Croixan natural-aggregate bodies (meltwater sediments) beneath the southwest client property (figs. 3, 4) are from 23 to 75 feet thick and from 300 to 1500 feet wide and extend from 500 to 1750 feet in a southeasterly direction. In vertical section, the bodies display 10° to 25° sawtoothed inter-fingerings with the bounding St. Croixan glacial sediments, mostly diamictons. In map view, the bodies consist of either a single layer or a complex of teardrop-shaped, pebble- to cobble- to rarely boulder-bearing sand layers, herein termed eskerine-facies sediments. Although most of the gravel-rich layers are matrix supported, clast-supported layers also occur. The layers dip and trend from N60°E to N70°E.

Sparsely pebble-bearing valley-fill sand bodies cross-cut the natural-aggregate bodies (fig. 4). These sandy materials (lateral/distal facies) extend generally southwesterly for distances greater than 1500 feet.

**Diamicton** The St. Croixan diamicton sediments bound the sides and thinly overlie the natural-aggregate bodies (fig. 4). The diamicton sediment sequence, commonly overlain by a thick residuum of loamy sand and sandy loam, ranges in thickness from 2 to 25 feet and is composed of pebble-bearing, loamy sand to sandy-clay loam. The diamicton sediments near land surface (less than 7.5 feet deep) are flow till (Drewery, 1986)—i.e., formed by the direct accumulations of sediments from the glacier ice and somewhat modified by gravitational and/or water transportation. The lower part of the sequence is of melt-out till commonly having pebble layers from 5 to 7.5 feet apart.

**Pine-Citian** A cover of Pine Citian moraine association diamicton and lacustrine sediments overlies the St. Croixan sediments (fig. 4). The thickness of the Pine Citian sediments of the southwest part of the client property ranges from about 5 feet in the eastern part to more than 15 feet in the western part. The diamicton sediments (melt-out till) consist mostly of sandy loam to loam with subangular to well-rounded, pebble- to cobble-size rock fragments. Lacustrine sediments up to 12 feet thick are found within lowlands between the natural-aggregate ridges. They consist mostly of

subangular to well-rounded sand and minor amounts of silty sand and sandy loam.

## DEPOSITIONAL ENVIRONMENT Overview

The following description of the depositional environments of the natural aggregate bodies is based on detailed examinations of the 8.3-acre client tract and reconnaissance examinations of the rest of the Osseo district. A series of stillstand zones formed during the retreat of the St. Croixan glacier-ice margin. (A stillstand may be defined as an equilibrium condition in the flow of a glacier-ice lobe during which the rate of ice melting approximately equals the rate of advance.) The stillstand zones that developed during the retreat of the St. Croixan glacier ice are interpreted, on the basis of land-surface exposures, to have been similar to those that developed during the retreat of the Des Moines lobe along the St. Croix moraine (Hoagberg, 1980) in an area about 15 miles to the southeast of the Osseo district.

Three lobate stillstand zones (southeast, medial, and northwest) formed during the retreat of the St. Croixan glacier ice from the Osseo district (fig. 2). As indicated by the fissure traces or crevasses shown on figure 2, the natural-

aggregate deposits are found within bodies that radiate out from a series of stillstands that occurred within each zone. Meltwater streams having widely varying rates of flow carried and deposited the natural-aggregate materials within ice-tunnel or open channels developed within the ice fissures just up the hydraulic gradient from the glacial ice margins.

### Stillstand zones

The three stillstand zones shown on figure 2 were delineated by differences in the general pattern of the aligned depressions (fissure traces) on the Osseo quadrangle topographic map.

Within the southeast stillstand zone the fissure traces are somewhat radial in plan, but nevertheless oriented southeasterly. The fissure traces of the southeast and medial zones are more widely spaced than those of the northeast zone (see fig. 2).

On the basis of research findings on the hydrology and sedimentation of present-day glaciers we can hypothesize the depositional environments that existed within stillstand zones during the retreat of the late St. Croixan glacier ice from what is now called the Osseo district.

Two general types of eskerine, natural-aggregate bodies, broad crested and steep crested, are found within the district. Both types reflect the shapes of the conduits (tunnels or open channels) that developed along ice fissures near glacial ice margin. Most bodies were deposited within fissures perpendicular to the local flow direction of the glacier ice; some crosscutting bodies, such as the lateral/distal valley-fill sands described earlier, were deposited within meltwater channelways.

**Broad-crested eskers** Broad-crested eskerine sediment bodies are believed to have formed mostly within tunnels developed within the basal beds of the glacier ice just upgradient from the ice margin. The relatively broad form of the basal-ice tunnels is attributed to a series of freezing/thawing cycles of short duration that resulted in low rates of meltwater flow within the channelways (Shreve, 1985). Sediments of the individual broad-crested eskers and coalesced eskers (fig. 5) are fairly well sorted and well bedded; they contain few large cobble to boulder-size clasts.

**Steep-crested eskers** The steep-crested eskerine sediment bodies (fig. 6), such as those in the southeast client property within the northwest stillstand zone, were probably formed in steep-sided, cathedral-shaped ice channelways

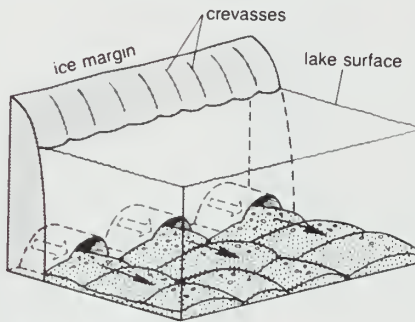


Figure 5 Diagram of a multiple, coalescing tunnel-esker depositional environment (modified from Rust and Romanelli, 1975).

(fig. 6) at positions more distal than the broad-crested bodies; they were probably formed during large-volume flows over relatively short time intervals (LeB. Hook, Wold, and Hagen, 1985). The rocks that had been entrained in the glacier ice adjacent to the channelways were carried mostly as bed load, by the relatively high stream flows. Deposition within relatively short, steep-sided channelways probably resulted in generally poorly sorted, poorly bedded eskerine sediments, whereas deposition within the longer channelways resulted in well-sorted, well-bedded, eskerine sediments. As would be expected, the sediments deposited in either of the depositional environments mentioned reflect the coarse and fine gravel (i.e., less than and greater than 2.0 mm) contents of the adjacent diamicton sediments (Shreve, 1985).

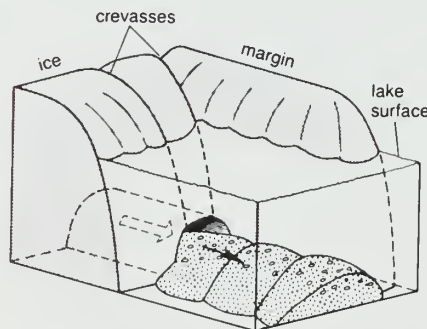


Figure 6 Diagram of a tunnel-esker depositional environment (modified from Rust and Romanelli, 1975).

### Lacustrine sediments

Upon entering the ice-front glacial lakes, the coarse-sediment load of the meltwater flows was deposited in the proximal reaches of open channel/deltas because of sudden dissipation in flow. The fine-sediment load was carried by overflows, interflows, and underflows until deposited in lacustrine fans and in a variety of other basin-margin and basin-floor facies.

### SUMMARY

On the basis of the study findings, we suggest that the depositional controls for the eskerine and natural-aggregate lacustrine-fan sediments of the southeast stillstand zone were deposited periodically within the broad ice-tunnel channelways that developed within fissures perpendicular to the glacier-ice margin. The brief periods of stream flows within these channelways were generally moderate to low because of the cyclic freezing and thawing of the glacier ice.

Within the medial stillstand zone, cyclic freezing of meltwater stream networks within the adjacent glacier ice resulted in moderate to low stream-flows and the development of broad-tunnel channelways. Consequently, the major depositional mode was that of broad-crested, gravel-bearing eskerine deposits.

Within the northwest stillstand zone, the occurrence of closely spaced southeast-trending fissures in the glacier-ice margin is reflected in the abundant sharp-crested eskerine, natural-aggregate-rich ridges. The occurrence of abundant gravel-size rock particles reflects the periodic, high-rate, meltwater flows within steep-walled ice tunnels; the few broad-crested eskerine bodies probably reflect local, broad, ice-tunnel deposition in channelways.

The lithology of the natural-aggregate bodies corresponds with the coarse and fine materials that had been entrained within the adjacent glacier ice.

### ACKNOWLEDGMENTS

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District for the use of their aerial photographs.

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# Evaluation of the Economic Usefulness of Earth Materials by X-Ray Diffraction

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## ABSTRACT

*X-ray diffraction (XRD) analysis makes use of a monochromatic beam of radiation, commonly CuK $\alpha$  to identify and quantify mineral composition. Recent advances in XRD instruments have made it possible to rapidly characterize large numbers of samples at relatively low cost. New instruments have sufficiently high resolution for analyzing bulk materials, unlike earlier XRD units that required separate analyses of fine, poorly crystalline clay minerals and coarse nonclay minerals such as quartz, orthoclase, plagioclase, calcite, and dolomite. However, since fine clay minerals often control colloidal properties and may affect other material characteristics, a separate sample of the fine fraction (<2.0  $\mu\text{m}$  to <0.1  $\mu\text{m}$ ) is usually analyzed.*

*A major shortcoming of earlier XRD instruments was a lack of resolution of amorphous or poorly crystalline constituents. This shortcoming can largely be overcome by defining the broad diffraction band of these components as a diffraction peak. This technique makes it possible to calculate an index of, for example, the intensity of constituents such as volcanic glass and coaly material, and form a ratio of their intensity to the total adjusted intensity of all mineral peaks.*

*This report describes sample preparation, analytical methods, and quantification methods used at the Illinois State Geological Survey (ISGS), and application of XRD analysis to the evaluation of several earth materials—aggregate, fired-clay raw materials, and absorbent clays. XRD results can also be advantageously used to correlate stratigraphic units, determine the orientation (fabric) of platy or fibrous minerals, evaluate the progress of diagenesis and weathering, determine the cause and cure of geotechnical problems, monitor mineral processes, and evaluate waste disposal sites. Chemical analyses and computer programs help to refine the analysis and increase the accuracy and precision of the evaluation.*

## INTRODUCTION

X-ray diffraction (XRD) is an analytical method in which a monochromatic beam of radiation is used to identify and quantify the mineral composition of earth materials and other crystalline substances. When X-rays strike a sample and are diffracted by the crystallites in the sample, the characteristic diffraction pattern produced can be recorded and shown graphically on a diffractogram. The heights, areas, shapes, and positions of the peaks on the diffractogram (fig. 1) provide information not only about the mineral composition of the sample as a whole but about each of the minerals present in the sample.

XRD has a wide range of applications to the industrial minerals industry as well as to the geological and material sciences. It is particularly useful in geological studies related to exploration, controlled mining, and processing of mineral resources; investigations of weathering processes and diagenetic, hydrothermal, and metamorphic changes in geologic materials; and monitoring of mixing and process reactions of materials during their conversion or use. In this report we (1) describe how ISGS geologists and other earth scientists and engineers are using XRD to solve a wide range of geologic problems, and (2) summarize sample preparation and quantitative

methods appropriate for these types of investigations.

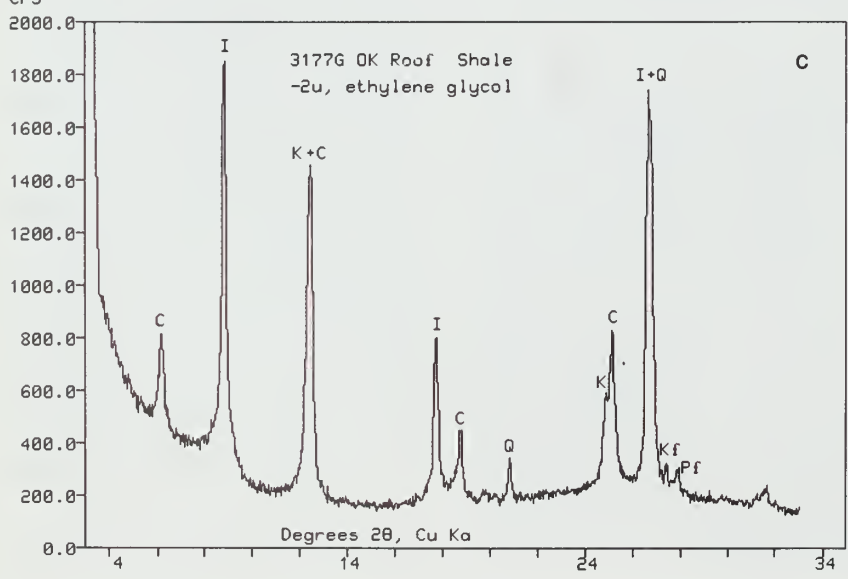
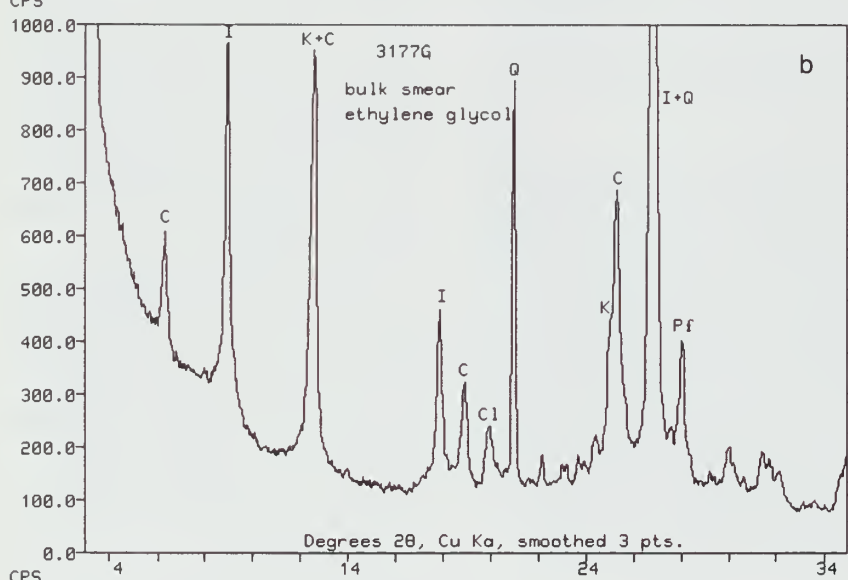
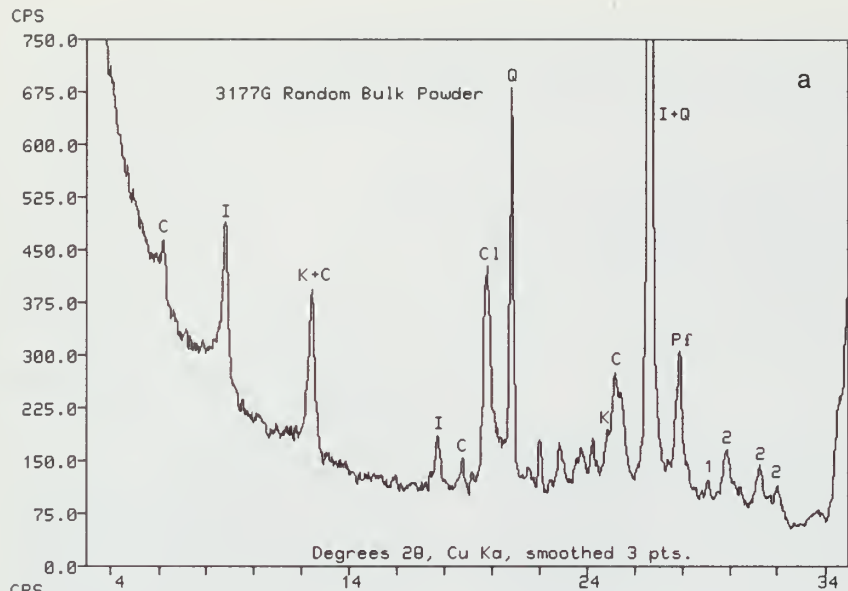
## XRD APPLICATIONS

The Survey's clay mineral database includes information on more than 100,000 samples analyzed by X-ray diffraction techniques. Most of the samples were analyzed to study stratigraphic relationships in Quaternary glacial deposits, investigate depositional and diagenetic patterns associated with Pennsylvanian coal strata, and estimate the resource potential of argillaceous (clayey) materials of all geological ages.

### Resource evaluation

An accurate XRD analysis yields a rapid estimate of the suitability of a material for virtually any use—from landfill covers, terra cotta, refractory ware, and pelletizing agents to building foundations and earth fill. Although analyses of clay mineral content of a material usually correlate with particle-size analyses of the clay-size fraction, XRD analysis directly reveals the mineralogical composition—and indirectly an approximate chemical composition of the sample—regardless of the size of mineral grains. In addition, mineralogical analyses often clarify aspects of material properties that chemical analyses fail to explain. For example, differences in the distribution of chemical constituents among minerals in different materials result in different firing properties for samples having identical bulk chemical compositions. We have found that, in general, XRD data on mineralogical composition (including data on the presence of amorphous material such as coaly or glassy phases) are useful in evaluating almost any questions involving the discovery, development, and use of earth materials.

Figure 1 shows XRD patterns for a shale that could be used for common and



face brick and red-fired ware. In evaluating a clay material for these or other clay product applications, four questions are paramount:

- Does it contain enough kaolinite to give extended firing range and/or refractory properties?
- Does it contain enough expandable clay minerals to qualify as an absorbent and barrier material?
- Are its quartz and feldspar contents optimum for a particular product or use planned on the basis of clay mineral content? (For example, if the material is kaolin, is the quartz content low enough that it can be used for paper filler applications? If the material is illitic common clay, is the quartz content high enough to permit rapid and thorough firing?)
- Does it contain any deleterious minerals such as calcite, dolomite, pyrite or marcasite, siderite, jarosite, or gypsum?

Questions such as these can often be answered in less than 30 minutes of instrument operation and pattern analysis. Because the new, automated diffractometers can operate almost unattended on a 24-hour, 7-day schedule, the time required for sample preparation and interpretation of results has become the principal factor limiting productivity in many laboratories.

Aggregate materials can be rapidly evaluated by XRD analysis (fig. 2). The amount of calcite, dolomite, and other phases can be determined with high precision. The magnesium content of calcite and the iron content of dolomite can also be measured by XRD. Contaminants in limestone and dolomite aggregates such as quartz and illite are easily detected by XRD and can be studied in detail by dissolving the carbonate fraction with acid before diffraction to concentrate the insolubles. Compositional trends in an aggregate deposit can be quickly estimated by XRD. Figure 2 illustrates the XRD pattern of two aggregates from Illinois—one calcite rich, the other containing mainly dolomite.

**Figure 1** XRD traces of a typical roof shale (Energy Shale) of the Herrin (No. 6) Coal. Top (a) is a random bulk mount; middle (b) is a bulk smear mount; and bottom (c) illustrates the diffractogram of an oriented, -2 μm slide. C, chlorite; I, illite; K, kaolinite; C1, peak common to clay minerals; Q, quartz; Kf, K-feldspar; Pf, plagioclase; 1 and 2, denote peaks for 1M (diagenetic) and 2M<sub>1</sub> (detrital) illite polytypes.

## Stratigraphic analysis

XRD analysis of mineral composition has provided critical data for stratigraphic interpretations at the ISGS for more than three decades. The recognition of individual units and delineation of their boundaries within Quaternary, Tertiary, and Cretaceous deposits have often simply been a matter of sorting the XRD patterns into types (e.g., patterns dominated by illite-chlorite, soil kaolinite, weathered expandable clay minerals, or any recurring association of minerals) (Frye, Willman, and Glass, 1964). Mineralogical trends in Pennsylvanian underclays can be similarly described by type designations (Parham, 1964; Hughes, DeMaris, et al., 1987). Most of the Survey's Quaternary studies have been based on the percentage of illite in unaltered diamictons (tills). However, Glass and Killey (1986) have reviewed studies in which distinctions have been made on the basis of calcite-dolomite ratios, vermiculite content, lepidocrocite or goethite presence, and the illite to kaolinite + chlorite ratio. Ongoing ISGS research on Quaternary materials includes recalculating raw data from weathered or altered materials to unaltered equivalents (personal communication, H.D. Glass, ISGS) (see TCI, table 1, method I) and extending and testing the hypothesis that all products of a major ice event have similar mineral compositions (Glass and Killey, 1986). If this hypothesis were correct, it would be possible to relate even isolated outcrops of till or outwash, lacustrine or fluvial deposits, and perhaps loess to a particular ice advance.

XRD mineralogical analysis data can indicate the relative stability of minerals during weathering and soil formation. For example, Hughes and Glass (1984) found that smectite in loessial paleosols (ancient soils) weathers to mixed-layered, kaolinite/smectite (K/S) at a higher rate than would have been expected from the rate of feldspar-to-kaolinite weathering. Hughes, DeMaris, et al. (1987) similarly demonstrated that in certain underclays (paleosols under coal beds), chlorite—normally the least stable mineral during weathering—was preserved, while illite and orthoclase were heavily altered by plant-induced removal of  $K^+$ . Hughes et al. interpreted this clay assemblage to indicate that the paleosol was similar to a gley. The rela-

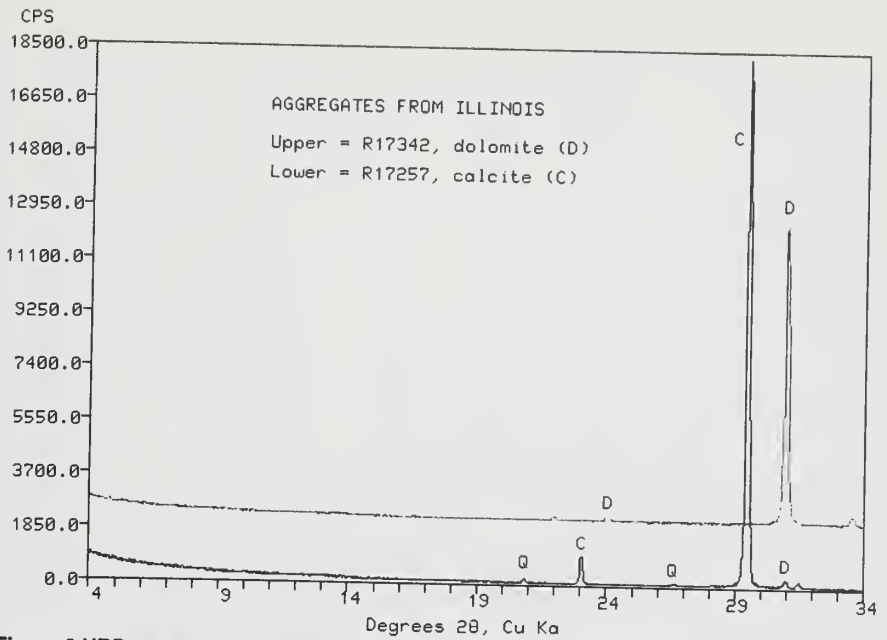


Figure 2 XRD traces of dolomite aggregate (upper) and limestone (lower). Q, quartz; C, calcite; D, dolomite.

tive stability of minerals is also controlled by the size of the mineral grains. Much remains to be studied about the mineral alteration in different particle-size fractions of modern and ancient soils.

The possibility that a till may weather in a unique way and that the environment during weathering may produce distinctive alteration patterns suggests that many profiles that were previously impossible to interpret may eventually yield useful information. Hughes, DeMaris, et al. (1987) noted that the type of mineral alteration in clays associated with coal measures may indicate the dominant plant group(s) that caused the alteration. As a result, studies of paleosols may provide new information about the environment and the floral population during poorly understood periods of pedogenesis, nondeposition, and sub-aerial exposure.

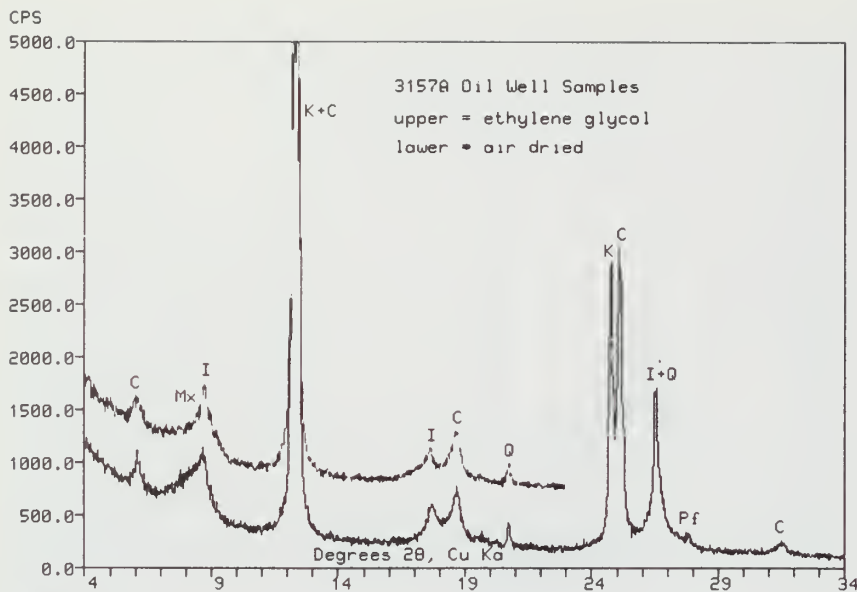
### Mineral orientation studies

Webb (1961) and Odom (1963) demonstrated that XRD techniques can be used to determine the orientation of clay minerals within undisturbed slabs of rock samples. Generally, samples mounted parallel and perpendicular to bedding are analysed, and the ratio of 001 peaks to 0k0, h00, or hkl peaks is calculated to quantify the degree of orientation. In this method, 001 peak intensity measures the number of platy particles that are oriented parallel to the sample surface or fissility, for example; 0k0 and h00 peaks are

measures of platy particles oriented perpendicular to the sample surface; and hkl peaks describe platy particles at some intermediate angle with the surface. In normal shales 001 peaks dominate slabs cut parallel to bedding and 0k0 peaks are most prominent in slabs taken perpendicular to bedding. Webb and Odom's results suggested that increases in preferred orientation and salinity during deposition were negatively correlated. Epoxy-penetrated, oriented slabs have been used at the Survey to study orientational changes induced by compaction and shear of earth materials. Preferred orientation of minerals other than clays can be studied by this technique, as can minerals and compounds in manufactured products.

### Diagenesis research

Investigations of changes that occur in sediments after deposition (diagenesis) and mass balances between sediment source areas and associated sedimentary deposits are among the most interesting and potentially valuable geologic studies being conducted currently. The discovery and recovery of oil and gas, metallic and nonmetallic mineral deposits, and potable water all may be functions of the nature of associated sedimentary deposits and the postdepositional changes that have affected them. The petroleum industry has been particularly active in research on diagenesis; we suggest that other mineral industries could improve



**Figure 3** Diffractograms of <2  $\mu\text{m}$  slides from an oil well in the Cypress Sandstone. Upper trace is after ethylene glycol solvation, lower trace after drying in air. C, chlorite; Mx, mixed-layered illite/smectite; I, illite; K, kaolinite; Q, quartz, Pf, plagioclase.

their exploration and recovery programs by increasing research efforts in this area.

The two most important analytical techniques for studies of diagenesis are X-ray diffraction and scanning electron microscopy/energy dispersive X-ray analysis (SEM/EDX). XRD reveals the mineralogical composition of a sample, and SEM/EDX yields information on petrographic relationships, grain size, microfossils, and chemical zonation. Clay mineral XRD analysis has been widely used to estimate burial and thermal history (Hower, 1981) and resulting petroleum accumulations (Burtner and Warner, 1986); differentiate authigenic versus "soil-type" kaolinite (Hughes et al., 1987a); and measure detrital versus diagenetic illite content in Paleozoic strata (Hughes, Glass, et al., 1987; Austin, Glass, and Hughes, 1989). Furthermore, XRD and SEM/EDX give practical information on the quantity of clay minerals such as chlorite, smectite, and fibrous illite that react uniquely with fluids used in the completion, stimulation, and enhanced recovery of oil and solution mining wells. Figure 3 shows the XRD trace of the <2 $\mu\text{m}$  fraction of a reservoir sandstone in which oil wells are often completed or stimulated with acid. Chlorite and perhaps illite in this sample (identified on the diffractogram) will rapidly react by hydroxyl layer and  $\text{K}^+$  layer dissolution with acid, and the result-

ant swelling clay will plug or damage the formation.

Geographic and stratigraphic variations in mineral content of geologic materials can provide important clues that aid in the discovery, evaluation, and production of mineral resources. The location and orientation of ancient shorelines and similar paleogeographic features are often revealed by trends in mineral content. The movement of fluids associated with petroleum migration and with certain types of mineralization produces patterns of diagenetic mineralization (principally clay minerals) that can be used to find these resources. Other types of mineralization often can be detected by halos of particular minerals associated with the deposits.

#### Mineral process monitoring

A wide range of mineral processes can be rapidly and efficiently monitored by XRD. Figure 4 shows the XRD trace of a coal char treated with carbon monoxide and ethanol to remove sulfur in a precombustion cleaning process (Webster et al., 1986). In this process, a number of chemical and mineralogical changes occur. Troilite ( $\text{FeS}$ ), reduced by CO from pyrite/marcasite, catalyzes the removal of organic sulfur. During organic sulfur removal with ethanol, oldhamite ( $\text{CaS}$ ) occasionally forms. The catalyst is removed after the ethanol step by slight

oxidation of the nonmagnetic troilite to a magnetic variety of pyrrhotite and magnetite—which is then removed by magnetic separation. The magnetic pyrrhotite content and the mineralogical changes resulting from changes of experimental conditions can all be accurately quantified and monitored by XRD. Similarly, it is often possible to measure flotation selectivity with XRD. Continuous online monitoring of mineral processes by XRD and X-ray fluorescence from the same X-ray source is a promising new application.

#### Geotechnical problems

Expandable clay minerals such as smectite can cause serious geotechnical problems, as noted in the keynote paper (Grim, this volume). The variety, amount, exchangeable cation, and moisture content of clays within a deposit have a major effect on the properties of geologic materials. When the presence of hydrated (10  $\text{\AA}$ ) halloysite is suspected in a deposit, samples should be collected, processed, and analysed by XRD before and after drying to confirm its occurrence. Hydrated halloysite, which loses structural water irreversibly, often gives confusing results in engineering tests when the samples are dried and rewet. Similarly, materials containing smectite, vermiculite, illite/smectite, or other expandable clays should be collected, mounted, and analysed by XRD at field moisture content whenever possible. Changes induced in hydrated clay minerals by mining, earth moving, compaction, and wetting/drying can be detected and the magnitude of a variety of resulting geotechnical problems can be estimated by the use of proper sampling techniques and XRD analysis.

#### Waste disposal studies

Most hazardous waste landfill designs involve argillaceous materials because most earth materials that attenuate, absorb or adsorb, or otherwise immobilize wastes are in the clay mineral and related zeolite mineral groups. XRD can be used to study the attenuation process and the reactions between wastes and clays. For example, exchange of interlayer  $\text{Na}^+$  or  $\text{Ca}^{++}$  in smectite by metal ions in wastes often results in a change in basal spacing of the smectite and the volume it occupies by 2 to 100 times. This change can cause shrinkage of clay barriers and leakage, or expansion and destruction of the integrity



of other parts of the containment structure. The primary use of XRD in waste disposal problems is as a screening method; small 001 spacing changes that may result from the dilute solutions common in the natural setting generally cannot be detected by XRD.

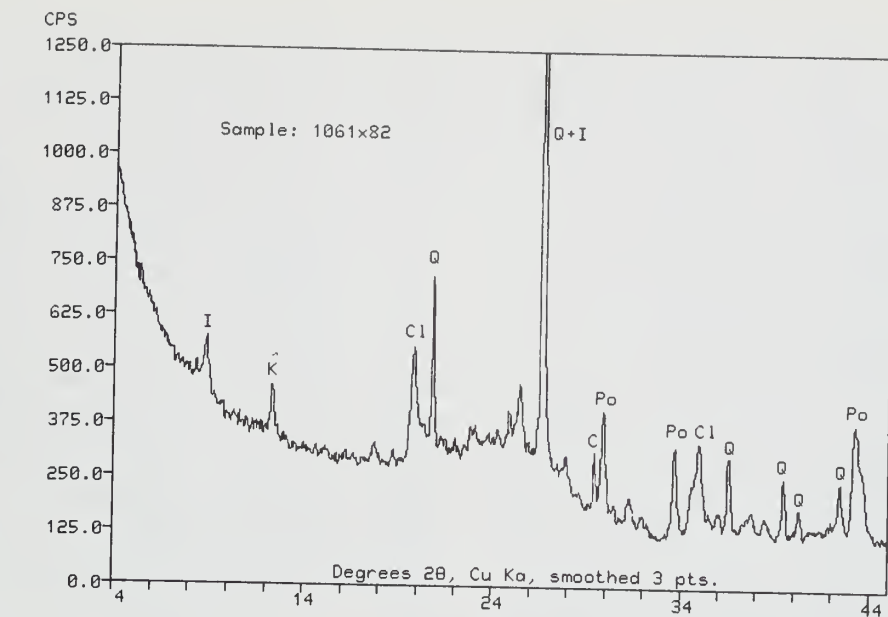
### Uses of XRD analysis: summary

Almost any geological problem requiring qualitative or quantitative mineralogic analysis can be advantageously attacked first by employing X-ray diffraction techniques. For example, XRD analysis can be helpful in

- locating, mining, processing, and manufacturing industrial minerals.
- evaluating clay and shale resources, aggregate materials (sand and gravel, limestone, dolomite and lightweight materials), coals, industrial sands, gypsum deposits, and metallic and non-metallic ores.
- monitoring all types of mineral processing.
- correlating strata and estimating the degree of weathering, diagenesis, or metamorphism that has affected rock materials.
- providing critical data on the degree of orientation of platy or fibrous minerals for geological and engineering compaction studies.
- evaluating the potential for contamination of groundwater at existing or potential waste disposal sites by determining whether the earth materials would tend to attenuate or transmit wastes. The amount and variety of clays and the presence of calcite or dolomite determine the degree of seal and buffering capacity of waste repository materials.
- characterizing the mineral content of earth materials for geotechnical evaluations of potential sites, evaluating the hydration and colloidal state of geotechnically sensitive minerals such as clays, and assessing the effects of possible disturbances of the site.
- solving a wide range of other geological problems.

### XRD INSTRUMENTS

At the Illinois State Geological Survey we currently operate three diffraction units: a 36-year-old General Electric model XRD5 with a horizontal goniometer (an instrument that measures the diffraction angles), a 20-year-old Norelco unit with two vertical goniometers,



**Figure 4** XRD trace of coal char from Co/Ethanol desulfurization process. Triolite or pyrrhotite from pyrite/marcasite is used to remove organic sulfur. I, illite; K, kaolinite; Cl, peaks common to clay minerals; Q, quartz; C, calcite; Po, troilite.

and a 2-year-old Scintag diffractometer with a vertical  $\theta/\theta$  goniometer. This newest unit has a nitrogen-cooled germanium detector and a 12-position automatic sample changer. The Norelco has a sealed proportional and a scintillation detector, and both goniometers have graphite monochromators. The General Electric unit employs a geiger counter with a nickel  $K\beta$  filter. All three units are operated with radiation tubes that produce a  $Cu K\alpha$  beam, although sources of  $Fe K\alpha$  radiation are available in the laboratory.

The types of X-ray diffraction instruments available to the researcher partly determine what types of samples can be analyzed and how the samples should be prepared for analysis. The sample preparation techniques discussed in the next section are generic; most can be carried out in any modern, well-equipped XRD laboratory.

### SAMPLE PREPARATION

A number of different methods can be used to prepare samples for XRD analysis, but by far the most common method is to grind the sample into a fine powder, which produces a characteristic powder diffraction pattern on the diffractogram (fig. 1a) when X-rays are diffracted by its crystals. The purpose of the analysis, the characteristics of the material being analyzed, and the type of

instrument used determine how the powder should be ground and mounted in the sample holder.

For routine analyses, powders should be ground to about  $<100 \mu m$ . If high levels of accuracy and precision are required, the sample must be ground to  $10 \mu m$  or finer. Careful grinding is essential: overgrinding of some minerals can alter their structure, cause a loss of XRD peak intensity and peak resolution, and broaden the peak, resulting in a diffractogram that is difficult to interpret. Optimum grinding is especially important in analysis of heavy minerals because large grains of heavy minerals have high X-ray absorption coefficients and are particularly opaque to X-radiation. If the inner and outer zones of a grain differ, grinding or chemical removal of the outer layer may be an essential part of the analysis.

Preferred orientation occurs when anisotropically shaped crystallites are oriented nonrandomly with respect to the surface plane of the sample. For example, acicular grains and platy clay minerals often have much higher intensities for some XRD peaks from the preferred orientation, and much weakened intensities for all other peaks. The extreme example of preferred orientation is the nearly perfect orientation of clay particles that results from sedimenting, vacuum deposition, or smearing of clay particles

on slides (see the following discussion). For identifying unknowns or quantifying known phases, it is desirable to achieve as random an orientation as possible. Similarly, oriented slides of clay minerals should be as oriented as possible.

Powder sample holders are designed for front-loading, back-loading, and side-loading. Because the best approximation to perfectly random particle orientation (and therefore the greatest quantitative precision) can be achieved with the side loader, this design is generally preferred. However, when only a small sample of a material is available, a shallow-well front loader is generally used, or an alcohol slurry is made with the powdered sample and deposited on a glass slide. Any method of grinding and mounting will produce fairly random orientation when used with materials having equant grains (grains having the same diameter in all directions); however, grains with fibrous, platy, or other habits, or cleavages that cause preferred orientation require fine grinding and a side-loader mount to reduce preferred orientation and achieve reproducible quantitative results. Increased randomization of clay minerals can also be obtained by spray drying a size-fractionated slurry of clay (Hughes and Bohor, 1970). In a new technique being developed in our ISGS laboratory, coarse equant grains such as quartz or fluorite are added to fine fractions of fibrous or platy clays. The finer particles tend to be randomized by the equant grains. Furthermore, partial randomization of clays can be achieved by sedimenting part of the silt fraction with the clay fraction on a glass slide. The coarser grains often randomize the clay minerals sufficiently that additional details about the clays can be observed.

Thin-section mounts (without the cover slip), polished sections, oriented rock slabs, and a wide variety of "as received" samples can also be investigated with XRD. The main requirement is that the sample be properly aligned in the X-ray beam.

Early investigators, working with more primitive X-ray diffractometers than those used today, were only able to study poorly crystallized clay minerals by preparing oriented aggregates (fig. 1c). Oriented samples, still used for most analyses of clays, are usually prepared by sedimenting the sample on a glass slide. The clay-bearing sample is dispersed in

water with a dispersing agent such as sodium hexametaphosphate and sometimes a pH modifier (pH 7 is about optimum). The content of clay-sized particles in the dispersion should be about 3 percent solids by weight for best results. (This requirement is approximate; at this percentage, smectite may gel and kaolinite may be too thin for best results.) Samples containing large amounts of silt and sand may require greater concentrations in the dispersion than clay-rich samples would. The better the disaggregation, the less sample required. Dissolution of carbonates and organic material with acid and oxidizing agents increases the clay-size content of a sample. Ultrasonic disaggregation or intense mixing or grinding in a blender also increases the amount of clay-size particles.

A fine-size cut of the dispersed slurry can be made by settling the sample according to Stokes Law (a formula expressing the settling rates of spherical particles in a fluid) or by increasing the settling rate with centrifugal force. The fine slurry, usually made with water, can then be sedimented on a glass or porous slide, pulled onto a Millipore filter or porous slide by vacuum, or smeared on a slide. In any case, the dried sample has a high degree of preferred orientation. When few equant grains are present and the clay grains are strongly platy or fibrous, nearly perfect preferred orientation results.

Three methods of preparing samples with preferred orientation are currently used in our laboratory.

- A sample is dispersed in water, stirred, and then settled to a selected size (usually  $<2\mu\text{m}$ ); an aliquot of the sized fraction is then pipetted onto a glass slide, and the slide is allowed to dry (this is the oldest technique).
- For some applications, the sized aliquot prepared by the first method is placed on a porous disk or slide and water is allowed to drain through the slide or is extracted by vacuum.
- A paste made of the whole material (fig. 1b) or a size fraction of the sample is smeared on a disk or slide. Whole-sample smears yield reasonably high degrees of orientation if the material is rich in clay. If the sample contains abundant equant particles, little preferred orientation results, and the sample may

approach a random powder in degree of orientation.

In approaching certain problems, such as a detailed description of the particle size of contaminant or valuable particles in an earth material, it may be helpful to analyze several narrow particle-size intervals (such as  $<0.10\ \mu\text{m}$ ;  $0.10$  to  $0.20\ \mu\text{m}$ ) from a sample. To accomplish this, the sample should be resuspended and sized three or four times for each size interval to remove most of the particles in that interval. A series of size separations based on a doubling of the diameter of the particle is a common choice of size interval. However, size interval fractions made on the  $\sqrt{2}$  times  $\sqrt{nd}$  (where  $nd$  is a doubled diameter series (i.e., 1, 2, 4, 8) yield more detailed information. Each interval can also be weighed to obtain a cumulative particle-size analysis.

Wet mounts can be quite useful for some problems. For example, gels of smectite or other expandable clay minerals can be mixed with liquid hazardous wastes to determine the likelihood that a clay barrier will fail and crack; seal, absorb, and/or attenuate the wastes; or expand and damage other parts of the containment structure. Furthermore, detection of the hydrated ( $10\ \text{\AA}$ ) form of halloysite is improved when the sample is run wet. In this case, a sample must be maintained at field moisture, processed wet, and analysed as a wet smear on a slide or as a gel in a powder holder.

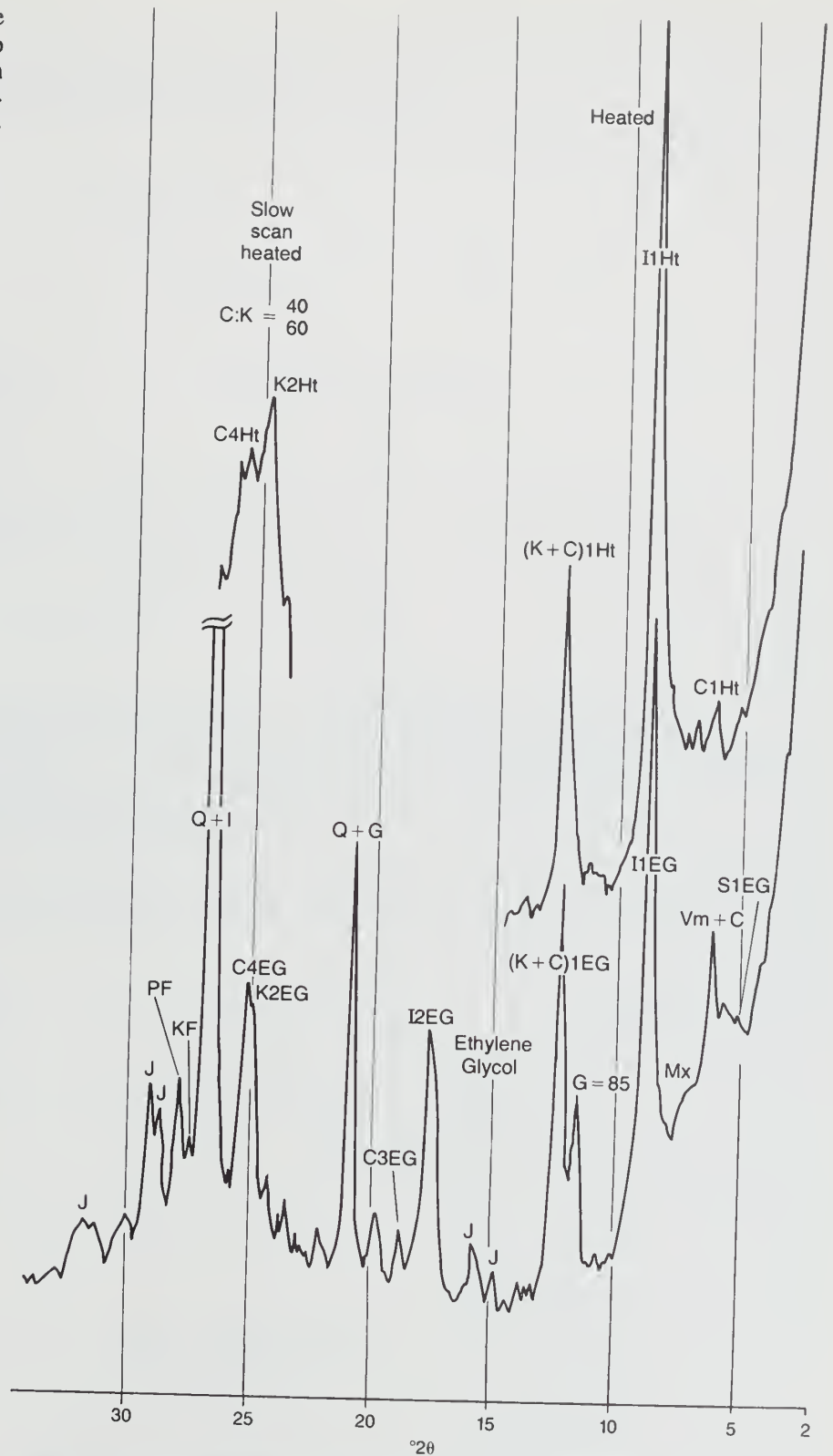
### Quantification methods

XRD estimates of abundances of minerals in samples range from simple descriptions and ratios of peak intensities to complex calculations involving additions of standard minerals, measurements of sample absorbance, determinations of chemical content, and computer programs employing chemical, diffraction, and other constraints to produce high-precision determinations. The choice of method is usually determined by the requirements of the problem to be solved and the number of samples to be analyzed. Rapid sample preparation and quantification techniques can be used for most geological problems; however, the description of a new mineral variety, for example, can take days or weeks. Most of the samples studied in the ISGS laboratory can be prepared and analyzed in a few minutes to a few hours.

An approach we have found valuable in investigating certain problems is to define the appearance of the diffraction trace as a series of *types* (see stratigraphic analysis section). Underclays and similar materials were evaluated by this method by Parham (1964) and Odom and Parham (1968). Use of the type method avoids several numerical anomalies that occur in quantitative analyses. For example, two samples may contain equal amounts of a particular clay mineral, but the degree of crystallinity may indicate two different origins. Furthermore, as illite is weathered to illite/smectite (I/S) or detrital I/S is diagenetically altered toward illite, the bulk quantity of I/S is often constant; the distinguishing change is the variation in the I:S ratio.

An early method of quantification still used in our laboratory is to compare the intensity of mineral peaks in a sample to intensities for pure standards of the same mineral. This method assumes standard sample preparation and an equal coefficient of absorbance between previously analyzed (external) standards and analytical samples. A modification of this method applicable to clay minerals is to use external standards to calculate clay mineral content on a 100 percent basis, even though nonclay minerals are present in the samples. This procedure often reveals stratigraphic, weathering, or diagenetic trends that are missed when only absolute mineral content is calculated, because variations in nonclay mineral content obscure changes in clay mineral content.

Table I describes the three methods of clay mineral analysis most commonly used in the ISGS laboratory. Method I, in use for more than 30 years, has undergone some changes during that time (Glass and Killey, 1986). This method uses peak height ratios among expandable (smectite or low-charge vermiculite), illite, and kaolinite + chlorite (fig. 5) from a diffractogram of an ethylene glycol-solvated, <math><2\mu\text{m}</math>, oriented sample to calculate the clay mineral content on a 100 percent basis. The quantitative factors are based on pure clay mineral peak intensities; the factors for XRD instruments using a logarithmic intensity scale (GE) are different from those using linear intensity scale (Norelco). Other clay mineral ratios (table 1) such as vermiculite index (V.I.), diffraction index (D.I.), type composition index (T.C.I.), heterogeneous swelling



**Figure 5** XRD traces of weathered clay wastes from coal mining illustrating peaks used in three methods of quantitative analysis described in Table 1. S1EG, Smectite 001 after solvation with ethylene glycol; Vm+C, vermiculite + chlorite; C1Ht, chlorite 001 after heating to 375°C for 1 hour; I1EG, I1Ht, illite 001 after glycol and heating; G, gypsum; (K+C)1EG, (K+C)1Ht, kaolinite 001 and chlorite 002 doublet after glycol and 375°C; J, jarosite; I2EG, illite 002 glycol; C3EG, chlorite 003 EG; Q, quartz; K2EG, kaolinite shoulder, glycol; C4EG, chlorite shoulder, glycol; K2HT, kaolinite 002 after 375°C; C4Ht, chlorite 004 after 375°C; Kf, K-feldspar, orthoclase, Pf, plagioclase.

**Table 1.** Quantitative Methods of XRD Analysis

**METHOD I.** Clay Percentages, Nonclay Peak heights, and Clay Indices-Ethylene Glycol Solvation (E.G.) (used primarily for stratigraphic and soil profile studies).

**Raw Data:** (See figure 5 for peak identifications.) Peak heights of expandable-S1EG, illite-I1EG, kaolinite + chlorite (K + C)1EG, counts for non-clay peaks, mm height of smectite, illite, and vermiculite (Vm-14A), color of XRD slide.

**Calculations:**

A. Adjusted clay intensity,  $\Sigma cl = S1EG + 3I1EG + 2(K+C)1EG$  for log scale, or

$$\Sigma cl = 1.4 S1EG + 4I1EG + 1.8 (K + C) 1EG \text{ for linear scale}^*$$

$$B. \% E = \frac{S1EG \text{ or } 1.4 S1EG^*}{\Sigma cl} \quad \% I = \frac{3I1EG \text{ or } 4I1EG^*}{\Sigma cl} \quad \% K + C = \frac{2(K + C)1EG \text{ or } 1.8(K + C)1EG^*}{\Sigma cl}$$

Vermiculite Index (V.I.) = a. If  $Vm > I = +$  mm height of  $Vm - I$

b. If  $Vm < I = -$  mm height of  $I - Vm$ ;

$$\text{Diffraction Intensity Ratio (D.I.)} = \frac{I1EG}{(K + C)1EG}$$

Heterogeneous Swelling Index (H.S.I.) = mm height of expandable peak above low angle background minimum

$$\text{Type Composition Index (T.C.I.)} = \frac{\%I1 + \%(K + C)^1}{\frac{\%I2}{\%(K + C)^2} + 1} = \text{corrected \% K + C}$$

1 From type composition, unweathered sample.

2 From unknown, weathered or altered sample.

NOTE: If TCI exceeds type composition, kaolinite is forming and/or illite is being weathered.

If TCI is less than type composition, chlorite is probably being lost by weathering.

**Results:** % E, % I, % K + C, V.I., D.I., H.S.I., T.C.I., presence or enrichment of kaolinite or chlorite, presence of mixed-layered kaolinite/smectite, peak height of calcite and dolomite, and presence of lepidocrocite, goethite, and any other uncommon phases.

**METHOD II.** Clay Mineral Parts-In-Ten (percentages) and Nonclay Heights—EG and 300°C (used for resource evaluations and general studies).

**Raw Data:** Peak heights of S1EG, I1EG, I1Ht, (K + C)1Ht, I2Ht (002 illite heated), C3Ht (003 chlorite heated), K2Ht, C4Ht, Q, Kf, Pf, Cc, D (fig. 5).

**Calculations:**

A. Adjust Peak Ratios,  $I_y = 1.0$

$$S_y = \frac{S1EG}{4I1EG}$$

$$E_y = \frac{I1Ht - (S1EG + I1EG)}{I1EG}$$

$$C_y = \frac{C3Ht}{I2EG}$$

$$K_y = \frac{K2Ht}{2C4Ht} \times C_y$$

NOTE: If chlorite is absent,  $K_y$  and  $C_y$  replaced by  $K_y = \frac{K1Ht}{I1EG}$

Total clay intensity,  $\Sigma cl = I_y + S_y + E_y + C_y + K_y$

$$B. \text{ Parts-in-ten, PIT, S (\%)} = \frac{S_y \times 10 (100)}{\Sigma cl} ; \text{ PITI (\%)} = \frac{1.0 \times 10 (100)}{\Sigma cl} ;$$

$$\text{PITE (\%)} = \frac{E_y \times 10 (100)}{\Sigma cl} ; \text{ PITK (\%)} = \frac{K_y \times 10 (100)}{\Sigma cl} ;$$

$$\text{PITC (\%)} = \frac{C_y \times 10 (100)}{\Sigma cl}$$

**RESULTS:** PIT or %-S, Vm (if all expandable = vermiculite  $\pm$  smectite), E, I, K, and C; Peak heights for Q, Kf, Pf, Cc, D, and other nonclays.

**METHOD III.** Clay Mineral Percentages, Corrected Illite/Smectite Percentages, Nonclay Peak Heights, Clay Indices, and Colloidal Index—E. G. and 375°C (used for resource, diagenetic, and general investigations).

**Raw Data:** Peak areas (height x width at 1/2 height or integrated area) of I1EG, I1Ht, (K + C)1Ht, and S1EG; expandable variety(s); smectite content of I/S and vermiculite expandability; K2Ht:C4Ht(y:z); nonclay peak heights in counts from ethylene glycol solvated trace (fig. 5).

**Calculations:**

A. Adjust clay intensity,  $\Sigma_{cl} = I1Ht + \frac{(K+C)1Ht}{2.5}$

B.  $\%I = \frac{I1EG}{\Sigma_{cl}}$                        $\%E = \frac{I1Ht - I1EG}{\Sigma_{cl}}$

$\%K = \frac{(K+C)1Ht/2.5}{\Sigma_{cl}} \times \frac{y}{y+z}$  ;               $\%C = \frac{(K+C)1Ht/2.5}{\Sigma_{cl}} \times \frac{z}{y+z}$

Corrected illite and smectite (E)

1) if only I/S is present,  $\%E_{corr} = \%E \times \text{smectite content of I/S}$ ;

$\%I_{corr} = \%I1 + (\%E - \%E_{corr})$

2) if only smectite is present,  $\%E_{corr} = \%E$

if only vermiculite is present,

$\%E_{corr} = \frac{\%E1(7 - (17 - Vm Pk \text{ in } \text{Å}))}{7}$

$\%I_{corr} = \%I1 + (\%E - \%E_{corr})$

3) If more than one expandable clay mineral is present, only an estimated expandability can be made, or if smectite and another phase are present, it may be possible to calculate discrete smectite (see part c, next) and then proportion the other phase according to (1) or (2) above.

C. Alternate method for samples with discrete smectite.

1. adjust clay intensity

$\Sigma_{cl} = I1Ht + \frac{(K+C)1Ht}{2.5}$

2.  $\% \text{ Smectite (S)} = \frac{S1EG/6}{\Sigma_{cl}}$  ;               $\%E = \frac{I1Ht - (I1EG + S1EG/6)}{\Sigma_{cl}}$

$\%I$ ,  $\%K$ ,  $\%C$ , and corrected  $I$  and  $E$  are calculated as in B1 and B2 above.

D. Clay Index

1. Adjusted nonclay intensity

$\Sigma_{nc} = .15G + Q + \frac{(Kf + Pf)}{3} + .47Cc + .25D + .83PM$

2. Clay Index (C.I.) =  $\frac{\Sigma_{cl}}{\Sigma_{cl} + \Sigma_{nc}}$               C.I. = 1.0 = all clay minerals  
C.I. = 0.0 = no clay minerals

E. Colloidal Index (CL.I.) = C.I. x (%E<sub>corr</sub>); CL.I. = 100.0 = all smectite  
CL.I. = 0.0 = no smectite

**Legend:** E = expandable, E1EG = peak at 17Å on EG trace, S = smectite, S1EG = smectite peak at 17Å on EG trace, Vm = vermiculite, I/S = mixed-layered illite-smectite, I1EG and I1HT = illite peak at 10Å on ethylene glycol-solvated and heated scans, (K + C) = kaolinite and chlorite peak at about 7Å, (K + C)1Ht = kaolinite + chlorite peak at 7.1Å on heated trace, I2Ht = second order illite peak at 5.0Å on heated trace, C3Ht = third order chlorite peak at about 4.6Å on heated trace, K2Ht = kaolinite peak at about 3.6Å on heated trace, C4Ht = chlorite peak at 3.55Å on heated trace, G = gypsum peak at 7.61Å, Q = quartz peak at 4.26Å, Kf = K - feldspar peak at 3.3Å, Pf = plagioclase peak at 3.2Å, Cc = calcite peak at 3.04Å, D = dolomite peak at 2.89Å, PM = pyrite and marcasite peak at 2.71Å.  
EG = ethylene glycol solvated.

Ht = heated at 300°C (Method 2) or 375°C (Method 3). 375°C partly destroys K/S; use 325°C for K/S.

Pk H = peak height in counts above background.

Pk A = peak area by integrated area or Pk H x width at 1/2 Pk H.

index (H.S.I) and chlorite presence or kaolinite peak changes are also determined, as are the presence and intensities of nonclay mineral peaks such as calcite, dolomite, goethite, and lepidocrocite. The color of the XRD slide is also described, because color often reveals subtle indications of zonation and oxidation state.

Method II (table 1) is similar to method I (peak height ratios are also calculated), except that the ethylene glycol-solvated sample is analyzed after being heated to 300°C for at least 1 hour. This method yields parts-in-ten results: smectite, expandables other than smectite (vermiculite and I/S); illite; kaolinite; and chlorite. The parts-in-ten results can be easily converted to percentages by multiplying each by 10. If only one expandable mineral is present, the expandable can be recorded as the actual mineral variety. This method is used with sedimented slides and <2 $\mu$ m smears.

Method III (table 1) makes use of peak areas, which until recently were calculated as the width of the peak at half height times the peak height. (New instruments now make it possible to calculate the integrated area of a peak by computer.) This method requires sample analysis after ethylene glycol solution and after heating (fig. 5); it gives a percentage of the four common clay minerals (E, I, K, and C) on a 100 percent basis (much like methods I and II).

However, for samples containing illite/smectite, a further step is included: the smectite in I/S is portioned off as "equivalent smectite," and similarly the illitic fraction of I/S is added to the percent of discrete illite. Vermiculite is not only recorded as vermiculite but also apportioned as though it were I/S with a smectite content of 4/7 (vermiculite expands to 14 Å and smectite to 17 Å in ethylene glycol; subtracting 10 Å as the nonexpanding layer leaves 4 and 7 Å, respectively). These further calculations are made because the degree of weathering of nonexpanding clays, the degree of diagenesis of expanding clays, and the colloidal properties of a sample are all proportional to the amount of equivalent smectite in that sample. New instruments and computer programs now make it possible to analyse complex mixtures of smectite, vermiculite, and I/S. Method III is used on analyses of <2 $\mu$ m sedimented or smear slides and whole-sample

smears. It can also be applied in a modified way to analyses of random bulk samples.

Two additional steps are normally taken in method III (table 1) to determine the clay mineral content and equivalent smectite content of whole-sample smears (or random-powder mounts). The first index calculation, the Clay Index, involves a ratio of the adjusted sum of the clay mineral peak area intensities to adjusted intensities for the nonclay minerals. We interpret this ratio to obtain information on weathering, diagenesis, and colloidal properties. Because colloidal properties of rocks are closely correlated with equivalent smectite content, a colloidal index is calculated for many samples.

Detection, identification, and quantification of certain minerals require that the samples be treated or modified in some way during the analysis. For example, all methods of clay mineral analysis in our laboratory use ethylene glycol solvation to expand I/S, vermiculite, smectite, and similar minerals to reproducible spacings. Method I (table 1) requires only one XRD analysis of a <2 $\mu$ m sedimented slide that has been solvated with ethylene glycol. Methods II and III require a second XRD analysis after the sample has been heated to 300°C to 375°C for at least 1 hour. Other treatments that can help resolve complicated diffractograms are analysis of air-dried and ion-exchanged samples; expansion of kaolinite with an intercalation agent; heating of gypsum to anhydrite; dissolution of carbonates, chlorite, and iron sulfides with acid; removal of organic matter by low-temperature ashing, H<sub>2</sub>O<sub>2</sub>, or NaOCl treatment; and dissolution of glass, smectite, I/S, and dehydroxylated kaolinite with caustic.

Samples that contain several minerals produce complex diffractogram patterns in which key peaks for different minerals are superimposed; these samples may require extra treatment to identify all the minerals on the diffractogram. In general, we prefer to obtain three or more isolated peaks for a confident identification of an unknown phase. However, small amounts of common minerals (such as quartz and calcite) expected in a particular sample are often identified and quantified on the basis of only one or two peaks.

Some XRD methods of quantifying mineral content are based on calculations

of peak height intensities, but the most precise methods are based on peak areas. The choice between calculating peak heights or peak areas usually is determined by the precision with which peak areas can be measured, by the degree of variation in peak sharpness of phases in the sample, and the extra time required to calculate areas. Methods I and II are based on calculations of peak heights, and method III is based on measurements of peak areas.

Software improvements on new or upgraded computerized diffraction systems have greatly increased the precision of XRD analysis. The current trend is to develop interactive, expert systems that enable the analyst to use several types of analytical data to constrain the result and use one or more methods of quantification. The learning aspect (artificial intelligence) of the new systems promises to allow retrieval of results with special characteristics, quantity, association, or variety of a particular mineral, from very large databases. It may also be possible to take advantage of stored "experience" to improve analyses of new sample sets. Another trend in quantitative XRD analysis is away from specially prepared fine-sample fractions (such as <2 $\mu$ m) and toward whole-sample analysis by bulk powder or smear methods. This last trend is the result of the higher resolution available on newer instruments.

#### **Factors affecting choice of XRD method and instrument**

It is a common misconception that the newest instruments and quantitative methods give the best results. In fact, the opposite is often true, at least in our laboratory. The choice of instrument and method is strongly dependent on the purpose of the analysis. For example, using our 36-year-old GE diffractometer and method I (table 1), we obtain precisions of 2 percent, even though the resulting data are often much less accurate than data obtained with newer instruments and methods. High precision results based on replicate analyses are particularly valuable for investigations of soil profile development and stratigraphic affinities between units. In such studies, the lack of accuracy of method I can be an advantage. The kaolinite + chlorite percentage calculated from method I is commonly greater than the amount actually present—generally by about 2.5 times.

But stratigraphic units and soil zones are much easier to detect when the apparent differences are magnified.

Studies of natural resources or material mass balances usually require the highest possible accuracy, even at the expense of precision. For instance, the built-in error of 2.5 times mentioned above, which could be useful in a stratigraphic study, would be unacceptable in an analysis to determine resource recovery or the processing and firing characteristics of a shale deposit. An error could also occur when using methods I and II to analyze a fine, smectite-rich material such as loess mixed with a coarse illitic material such as till. A sample of this mixture would appear to have more loess than till, because the fine, lower density smectitic particles preferentially settle more slowly in the beaker and on the slide. The correct material balance for this mixture can be obtained by preparing a smear or random powder of the whole sample or clay mineral fraction.

Another factor affecting choice of instrument is the ease of acquiring data. When the type method of analysis is chosen, any instrument is equally useful. The choice then is based on whether treatments such as ethylene glycol solvation or heating are required. For instruments with automated sample holders, an environmental chamber may be needed to keep treated samples from drying out or otherwise changing before they arrive in the X-ray beam to be analyzed. This factor can favor older instruments—in which samples are run individually before changes occur—even when the newer unit would give increased accuracy and precision.

Choice of method or instrument may also be affected by the need to compare new data with previous results. If critically important publications or data in a certain field have been based on a certain approach, it is best to use the same instrument, if still available. If type methods are used, results from a particular instrument may simply make type identification easier or make more direct comparisons possible. To summarize, the information required to solve a certain problem, the

time available, and the resources that will be devoted to the problem all should be considered when selecting the analytical method to be used.

#### ACKNOWLEDGMENTS

We are grateful to H.D. Glass, J.H. Goodwin, R.D. Harvey, D.M. Moore, and W.A. White for their helpful criticisms. We also appreciate the support of our research by the State of Illinois.

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# The History, Geology, and Future of Industrial Clays in Illinois

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## ABSTRACT

*Economically useful clay products in Illinois are made from raw materials ranging from Quaternary to Paleozoic in age and from claystone to siltstone in texture. The materials can be used for five types of products: (1) common ware and refractory fired-clay products, (2) absorbent clays, (3) impermeable barriers, (4) flux clays for cement manufacture, and (5) miscellaneous uses. Fired-clay products include bricks, field drain tile, stoneware and pottery, structural and decorative wall tile, sanitary tile, lightweight aggregate, and refractories. Absorbent clays have traditionally been used to absorb oil (in floor-sweep compounds, for instance) and as pet litter absorbent. New uses for absorbent clays include binders and pelletizing agents for everything from iron ore to coal to animal feeds; slurry stabilizers and suspension aids for agrichemicals and similar materials; absorbents for hazardous wastes; and impermeable barriers for hazardous waste impoundments and similar structures. Flux clays are used in cement manufacture to control melting point (energy use), trace element and sulfur contents, and strength. Miscellaneous potential uses include filling, stabilizing, pelletizing, and suspending various agrichemicals; producing synthetic zeolites; providing sources of alumina and other elements. Light-weight aggregate has been produced in Illinois in the past, and excellent raw materials for this use are still available.*

*The fired-clay industry has declined steadily in Illinois since the turn of the century. This decline can be attributed to a number of factors, including replacement of structural brick and tile with steel and concrete, substitution of brick veneer by cheaper products, increasing use of plastic drain pipe to replace clay field tile, competition from southern states due to low labor and freight costs, a failure to modernize plants, increased energy costs or shortages, tighter environmental regulations, urban encroachment, and the poor business practices common among small, family-owned businesses. New trends that tend to sustain the industry are nearness to markets, preferential treatment for in-state producers, automation, potential co-mining, captive sources of natural gas, abundant resources, and coal-fired plants.*

*Absorbent clays continue to be a strong and growing segment of the industrial mineral industry in Illinois. Increased local demand has resulted in a search for new resources, and this search identified several deposits for further testing and evaluation. Some new interest has also developed for fired-clay products.*

vanian strata contain shales and claystones that are ideal raw materials for red-fired ware; kaolinitic underclays (low-grade refractory clays) that can be blended to lighten red-fired ware; and underclays that contain sufficient expandable clay minerals to have potential as absorbent or barrier clays. Pre-Pennsylvanian clays and shales generally are not preferred materials for most uses because they contain carbonates and have too little kaolinite. However, these older strata are usable in regions lacking higher quality raw materials. The kaolinitic clay that is a minor component in the St. Peter Sandstone in La Salle County could be used in many markets currently using Georgia kaolin and Kentucky/Tennessee ballclay.

In this report we discuss the distribution of clay reserves, their geologic setting, and their current or future potential for development. We also review the industry's past, discuss co-mining as an alternative source of materials, and emphasize strategies that may increase the vitality of the clay products industry in Illinois.

## INTRODUCTION

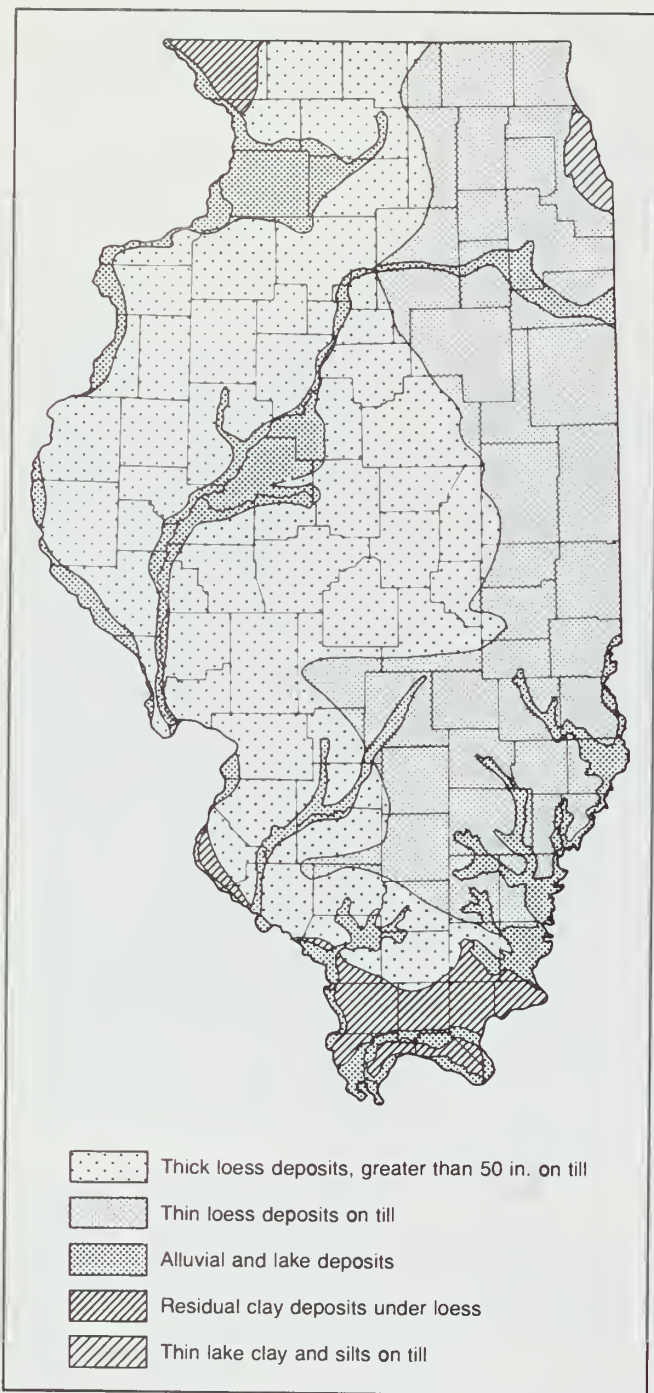
The clay products industry in Illinois began with Indian pottery dating from about 1000 B.C. Following European settlement, early pottery, brick, and tile works expanded rapidly in the 19th Century. The industry benefited from pioneer research at the Illinois State Geological Survey (ISGS) directed by J.E. Lamar and R.E. Grim, early leaders in the developing field of industrial minerals. A key element in the success of the clay industry has been the large investment in basic and applied research at agencies such as the ISGS. This research produced one of the most extensive databases on the composition of clay-rich strata in the nation.

Knowledge of the geology of clay-rich raw materials expanded greatly in the

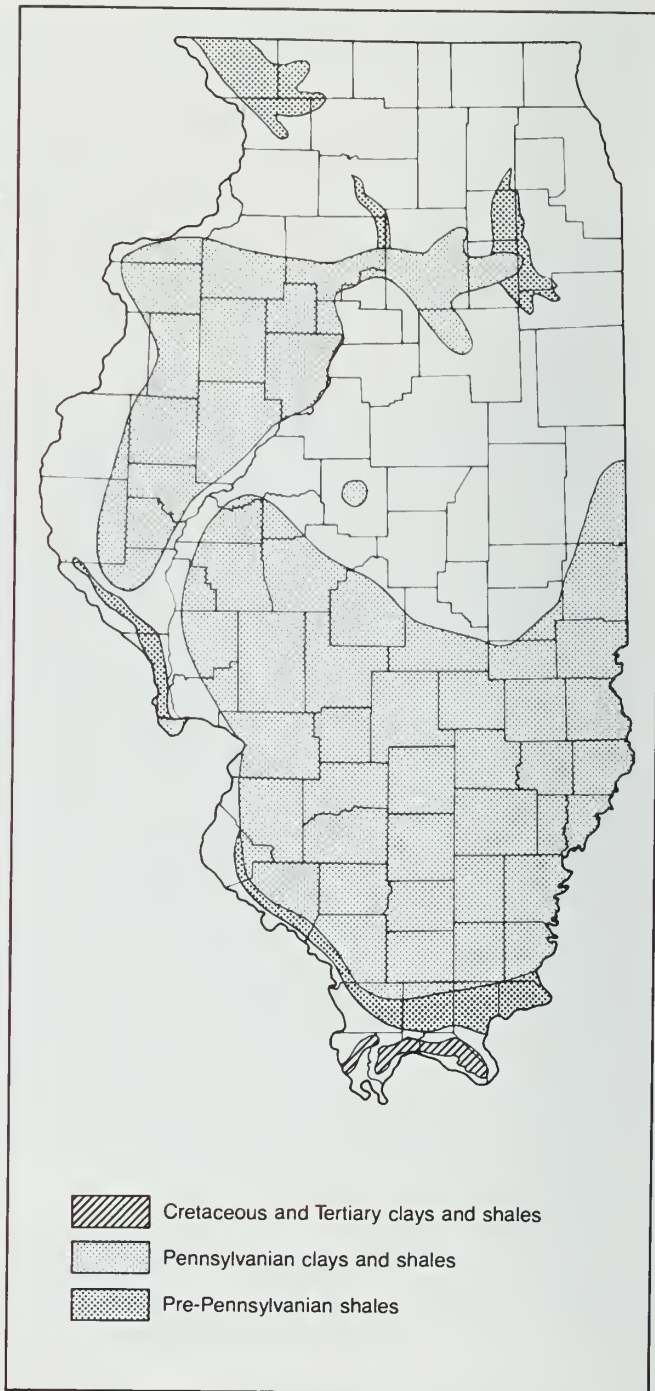
## GEOLOGY OF CLAY RESOURCES

Clay products are currently made from Quaternary, Paleocene, and Pennsylvanian strata. Fired-clay products are produced from Pennsylvanian shales and Quaternary till and lacustrine deposits. Quaternary deposits with potential value include the tills associated with Wisconsinan, Illinoian, and pre-Illinoian ice advances, Sangamonian and Yarmouthian paleosols and gleys, lacustrine deposits, and Wisconsinan loess deposits. The

Paleocene Porters Creek Formation occurs in Alexander and Pulaski Counties, the southernmost counties in the state. The Porters Creek is a smectite-rich clay that is mined for absorbent clay products. The Cretaceous McNairy Formation, which occurs in southern Illinois, consists of an upper and lower fine, micaceous sand with a middle lignitic to silty clay called the Levings Member (Willman et al., 1975). The Levings Member may have use for fired-clay products. Pennsyl-



**Figure 1** Surficial clay deposits of Illinois (after Jonas, 1957).



**Figure 2** Bedrock clay deposits of Illinois (after Jonas, 1957).

past few decades, partly because of improvements in automation of instruments such as X-ray diffractometers. These advances made it possible to investigate clays from individual localities in greater detail and survey large numbers of samples in a relatively short time.

Ceramic products have been manufactured from shale, clay, diamicton (till),

loess, lacustrine clay, and stream alluvium. The distribution of surface materials—till, loess, lacustrine deposits and alluvium—is shown in figure 1; the distribution of shales and claystones beneath the surface materials is shown in figure 2.

Shales have been used in the manufacture of common, facing, paving, and patio

brick; wall, floor, quarry, roofing, and field tile; partition tile and block; sewer pipe and flue liners; earthenware; stoneware; art objects; lightweight aggregate; and cement. Nonrefractory clays have the same uses as shales. Till is used to make some of the same products that are made from shale; loess has been used for brick, field tile, and light-weight aggregate

manufacture, and has also been mixed with shale for pottery and similar products. Alluvium is used for pottery by amateur potters; it can be used in the manufacture of cement if the magnesium content is not too high.

Except for light-weight aggregate, refractory clays have been used for the same products as shales, for refractory bricks and shapes, refractory filler, caulking compounds, cements, and in stove and boiler liners. Refractory clays are added to other nonrefractory raw materials to extend the range of melting temperature, modify color, and increase the strength of the product.

Economic trends at least partly beyond the control of local producers caused a slump in the Illinois fired-clay products industry, but the industry may have bottomed out; investment in new, highly automated plants has begun. In contrast, the absorbent clay industry in the southernmost Illinois counties has prospered in the last 20 years.

The future of clay products in Illinois hinges on several economic trends. If concerns with hazardous waste disposal result in increased use of naturally impermeable and absorbent materials, all usable absorbent or barrier clays will realize enhanced economic potential. If the transportation anomalies discussed in the next section are eliminated, the fired-clay products industry will benefit immediately. Even without changes in outside factors, the need for fired-clay products and the availability of energy and raw materials near major markets will probably result in growth in this area. In addition, several promising new directions—such as products for hazardous waste disposal—could increase the markets for clay materials. Finally, it is clear that the clay industry, like all basic industries, would benefit from a return to investment strategies of the past—more in concert with attitudes of foreign producers—in which long-term strategies are considered as important as short-term profit-taking.

## **HISTORICAL DEVELOPMENT**

### **Fired-clay products\***

Native Americans in Illinois used materials outside their living quarters for their pottery and fired it in wood fires to

about 500°C to 600°C (900°F to 1100°F) for several hundred years before European settlers arrived. The first European settlers in Illinois brought their pottery with them, and shortly thereafter, bricks and pottery were being shipped into the state from Europe, Louisiana, and down the Ohio River from states to the east.

In 1806, Robert Harrison set up the first commercial pottery along Cahokia Creek, 4 miles north of Edwardsville in Madison County, and began to produce earthenware. A second pottery was established in Union County about 1820, and a third was founded in Madison County in Upper Alton a short time later.

The first bricks made in Illinois probably were produced at the end of the 18th or the beginning of the 19th century in St. Clair and Madison Counties. A brick church in Cahokia is said to date from 1699, but the bricks may have been imported. The first brick house in Illinois was built for Major Nicholas Jarrot in Cahokia in 1801. The bricks were most likely made by the soft mud method, dried in the sun or in open sheds, and fired in stone-type kilns with wood and later with coal. At first, bricks were made from the surface material at the place they were to be used.

As villages increased in population and size, brick plants were erected, and bricks were hauled from the manufacturing yards for building purposes. In 1835 the first horse-powered brick machine was invented, and by 1840 the machine was adapted to steam power. After the brick machine was invented, organized brick yards began to spring up in every little village, town, and hamlet. Some of these early plants produced bricks only a few months of the year; others operated longer, but few plants stayed open during winter. Many plants were run by just two or three people, whose pay ranged from 50 cents to a dollar for a 10- to 16-hour day. If two plants existed near a city, the plant that produced the least paid the least per day for labor. Many of the plants that operated for the longest days paid the least and produced the least.

Ceramics industries in Illinois remained small and localized until after the Civil War. Until the railroads began to appear in the 1850s, all industrial growth was hampered by poor transportation, but

by 1870 the manufacture of ceramic products was increasing rapidly.

In 1866, the first field tiles were manufactured by A. Horrock at Colchester, who converted his fire brick plant into a tile plant. He had difficulty selling the first tile because the farmers didn't believe man could improve on "God's handiwork." But when farmers saw the increased crop yields in tiled fields, the field tile industry expanded rapidly. Many brick plants began to produce both brick and field tile for local consumption.

In 1875, a man named Hoefler made the first paving brick at Bloomington, Illinois. Although the Pleistocene tills used for raw materials were poor in quality, the bricks lasted for 20 years. By 1884, a plant in Galesburg had begun making paving brick of such high quality that the term "Galesburg brick" became the standard for paving contracts west of Indiana. Other companies also made paving bricks from Pleistocene tills at Bloomington, Urbana, and other cities.

Around 1868, the Galesburg Press Brick and Tile Company was the first to commercially produce brick from shale (Pennsylvanian-age Purington Shale). Before 1868, bricks had been made only from Pleistocene surface materials, but the shales also made high-quality paving brick and field tile.

D.V. Purington came to Chicago from Maine in 1869 and established a lumber yard. After the Chicago fire in 1871 he recognized the potential for rebuilding the city. As an associate of Straus, Hahne and Company, brick manufacturers, he was responsible for that company's use of the Chambers Brick Machine, the artificial drying of bricks, and the steam shovel to mine clay. He later became president of Purington-Kimbell Brick Company, which at the time was claimed to be the largest brick manufacturer under one management. He was also president of Purington Paving Brick Company of Galesburg, a major paving brick manufacturer. His brother, Charles, introduced a method to use crude oil in firing brick.

The modern building-tile industry in Illinois dates from about 1880, when H.B. Williams and C.H. Bess established the Standard Brick Company plant about a mile east of Ottawa. The Standard Brick plant, which operated until about 1960,

\*This section on fired-clay products was adapted from an unpublished manuscript written by Robert L. Major around 1970 (on file at the ISGS).

made building tiles from Paleozoic shales and Pleistocene till.

In 1882, S.E. King, another local capitalist, organized and built the Ottawa Fire Clay and Brick Company plant near the Standard plant. Chicago Retort and Fire-brick Company acquired King's company about 1900.

The Illinois Terra Cotta Lumber Company, organized in 1885, made hollow flat-arch tile for iron construction, flooring tile for wooden joists, ceiling tile, partition tile, wall furring and defining, column, girder, and beam castings. In 1892 the Chicago Terra Cotta Company commenced manufacturing terra cotta roofing tile in a plant 3 miles northwest of Ottawa.

According to Risser and Major (1968), 697 plants in Illinois produced clay products valued at \$8.4 million in 1894, and the value of clay products grew continuously until 1957, when it reached \$60.7 million. At the beginning of their study of moisture expansion of brick in 1959, Hoskins, White, and Parham (1966) could choose test materials from about 60 plants making ceramic materials in Illinois. In spite of the growth in value up to 1957, Illinois dropped from second rank among the states just before 1900 to fifth in 1905, and has been too low to be ranked since 1905. At present, only three or four plants are producing fired-clay products and only about six plants are producing clay products of any kind.

### **Kaolin**

Kaolin was produced near Anna from early in this century until a few years after the end of World War I. The kaolin was mined from sinkholes in Mississippian and Devonian strata. One of the authors was told that some of the kaolin that was mined underground was plastic enough that it could be rolled and loaded on mules and hauled out of the mine. Production of refractory clay (rich in kaolinite) from the Cheltenham Clay and underclay below the Colchester (No. 2) Coal has continued from before World War II to the present. The Cheltenham Clay and underclay were mined together at most localities. Until about 1980 this clay was added to refractory products; at present it is used only in ironstone pottery and high-strength bricks.

### **Light-weight aggregate**

The method for expanding clay and shale to produce light-weight aggregate

was invented by Hydraulic Press Brick Company in Kansas City just after World War I. This company opened a plant with a rotary kiln in St. Clair County, Illinois, a short time later. When their patent expired after World War II, Alton Brick Company, Postam Brick Company of Springfield, and Western Brick Company of Danville all opened competing plants. Alton Brick used a traveling-grate kiln and a mixture of Quaternary raw materials, coal, and coke. Postam and Western both used rotary kilns, Pennsylvanian-age raw materials, and some coal and coke as additional bloating agents. Material Service Company started production from two or three large rotary kilns in La Salle County, using the entire local section from Quaternary to below the Canton Shale (Pennsylvanian), excluding gravels. This large increase in production and the availability of cheaper raw materials led to the closure of competing plants. Alton Brick closed in 1963, Hydraulic Press Brick ended production in 1970, Postam went out of business in 1971, Western stopped production in 1973, and Material Service closed its plant in 1978. River sand replaced Hydraulic Press Brick's product, while Alton, Postam, and Western all were displaced by Material Service's large output, and Material Service's product finally lost out to slag, a waste product.

### **Absorbent clay**

Porters Creek clay (Paleocene) was used by Standard Oil of Indiana and Sinclair Oil companies because of its natural absorbent qualities for removing contaminants and clarifying, "bleaching," and decolorizing motor oils. Its absorbent properties also made it useful for sweeping compounds used to remove grease and water from garage floors in service stations. At some time around World War II, acid-treated bentonites from Mississippi were shown to have a higher bleaching capacity than the Porters Creek Clay. When oil companies acid-treated the Porters Creek Clay, they added too much acid and obtained little or no decolorizing improvement. As a result, the two oil companies changed to Mississippi clay.

Although Porters Creek clay had been sold occasionally as a litter material for livestock barns, its main use was still for oil decolorizing. About 1960, Ed Lowe opened a plant in Olmstead to produce pet

litter material from the Porters Creek Clay; this use is the principal market for absorbent clay in Illinois today. The clay has also been used in the last decade as animal feed binder to increase the flowability of animal feed. It can also be used as a carrier, stabilizer, and suspending agent for various agrichemicals.

### **ECONOMIC TRENDS**

Figures 3, 4, and 5 show total production, unit value, and total value for clay products for the years for which reliable statistics are available. Some of the peaks and valleys clearly reflect major wars, economic panics, and the Depression. The peak in value during World War I (fig. 4) was largely the effect of shipping blockades that decreased imports of clay products and raw materials, and from extensive mining of Anna kaolin to replace previously imported products. (Anna kaolin, suddenly much in demand, was being sold at cents per pound rather than the cents per ton being paid for common clays.) Figure 3 shows the decline in production that has occurred in the last 25 years. It is important to note that the two companies that produce absorbent clays are not included in the figures. Although the picture would seem considerably brighter if these figures had been added, the overall loss to the state of Illinois is more clearly reflected by the values for fired products because this industry is more uniformly distributed in the state.

Many factors contributed to the decline of this fired-clay industry, but the main causes were (1) the change from solid, load-bearing brick walls to steel and concrete construction with brick veneer; (2) replacement of brick veneers by less expensive wood, concrete block, vinyl, and aluminum; (3) substitution of clay field tile by plastic pipe; (4) inexpensive northward (back haul) rail and truck transportation for products from southern states; (5) poor management by small, family-owned businesses; (6) pollution-control regulations; (7) labor costs and strikes; and (8) low-quality raw materials.

Rail haulage economics played a particularly important role. Production costs for clay products are normally lower in southern states, and this price advantage is amplified when heavy southward flow of freight traffic creates subsidies for northward haulage in cars that would otherwise return empty. Given the nearly

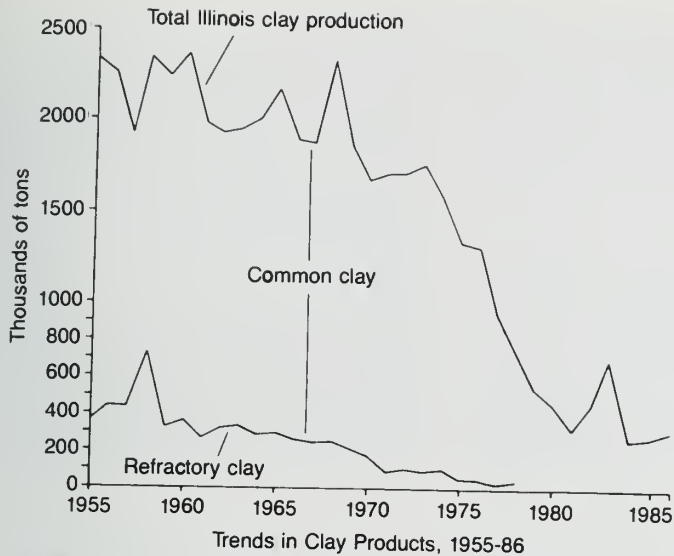


Figure 3 Illinois clay production from 1955 to 1986.

700 small, locally owned clay businesses in Illinois at the end of the last century (Risser and Major, 1968), it was to be expected that extensive consolidations would occur. However, several producers also went out of business because of family entanglements, especially the retirement or death of the founder, often the driving force of the business; a failure to invest in modern equipment; lack of concern about market shifts; and the recent investment strategy prevailing in the United States that emphasizes short-term returns.

There are also some hopeful signs. Illinois and surrounding states are among the largest markets in the nation, and Illinois is the nearest producer to several markets in nearby states. The state has an excellent transportation system, several major investment centers, and a progressive and helpful attitude towards new business. Illinois and the federal government have several programs to promote the industrial use of coal as a source of inexpensive energy. The state is known for its skilled labor force, but as a major industrial state, Illinois has higher-than-average labor costs. Therefore, new plants will have to be highly automated to be competitive.

### GEOLOGY OF CLAY RESOURCES

Raw materials for clay products are found in Holocene and Pleistocene, Tertiary, Cretaceous, Pennsylvanian, Mississippian, Devonian, and Ordovician strata. In order of importance, the Tertiary (Paleocene), Pennsylvanian, and Quaternary are the sources of all active operations. Table 1 contains an outline of units that are or could be important as clay resources. The following review provides a geologic framework to aid in the exploration for new clay resources and the evaluation of existing or previously mined deposits.

#### Holocene and Pleistocene

Holocene and Pleistocene deposits, the earliest sources of ceramic materials, continue to be used in small amounts. Because most of Illinois is covered by these deposits, their potential for future uses is high. Quaternary history includes at least three major ice advances and at least three interglacial stages, including the Holocene. From youngest to oldest, the order of events is: Holocene (interglacial), Wisconsinan (ice advance), San-

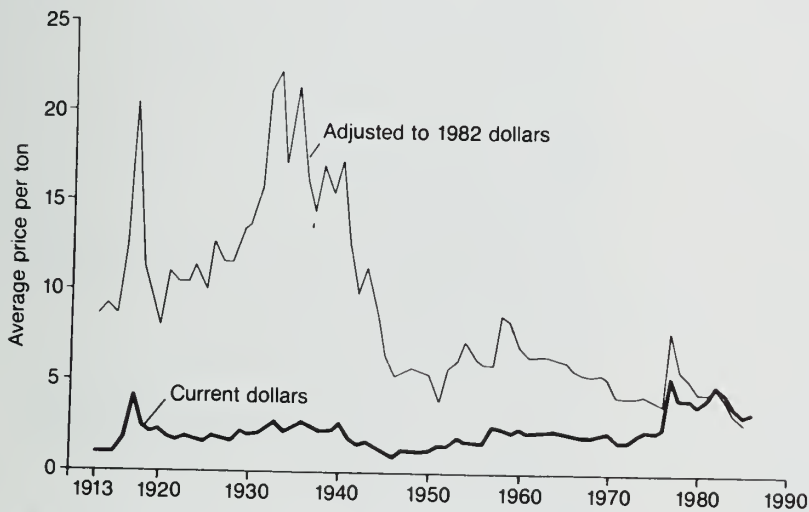


Figure 4 Unit value of Illinois clay production from 1917 to 1986.

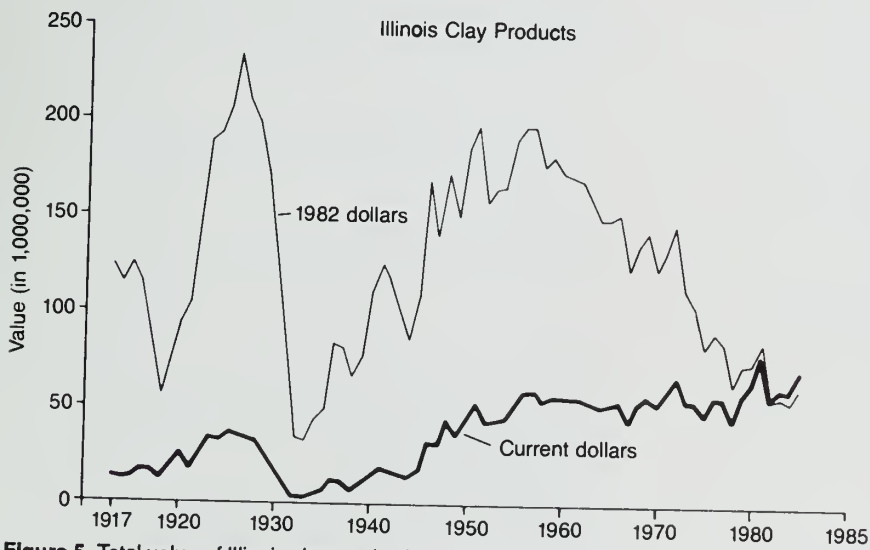


Figure 5 Total value of Illinois clay production from 1917 to 1986.

gamonian (interglacial), Illinoian (ice advance), Yarmouthian (interglacial), and pre-Illinoian (formerly Kansan and Nebraskan ice advances). The Illinoian glacial advance penetrated farther south in this state than any other continental glacier in the United States. The Quaternary deposits of Illinois contain excellent stratigraphic records of glacial and interglacial deposition, and interglacial soil development.

The Holocene is represented mainly by alluvium along and near rivers and streams. Holocene peat, lacustrine deposits, and sand dunes also formed on a small scale. The modern soils, developed

mostly on Wisconsinan loess, are weakly developed in comparison with their Sangamonian and Yarmouthian predecessors. Carbon 14 dating indicates that the last ice at Chicago probably melted about 14,000 years B.P. (Hansel et al., 1985); the youthful nature of the recent soils supports this date.

Wisconsinan ice moved into Illinois from the Lake Michigan Basin and covered much of the northern half of the state. Fluvial and lacustrine deposits, varying from gravels to clays, occur both within the area of ice-associated deposits and beyond the ice margin. Wisconsinan diamictons (tills) can often be traced

stratigraphically by their clay mineral composition (Glass and Killey, 1986). The Wisconsinan deposits consist principally of diamictons in a series of recessional moraines that become successively younger in the up-ice direction closer to Lake Michigan (Willman and Frye, 1970) (fig. 6). The lower part of the Wisconsinan usually consists of silt and alluvial diamicton.

Wisconsinan-age loess deposits are thickest along the eastern bluffs of the Mississippi and Illinois Rivers; they thin to the east-southeast. Older loesses exist, but they have not been important for clay products. The clay mineral composition

**Table 1** Available materials by geologic age (after Willman et al., 1975).

<b>CENOZOIC</b>	
<b>Quaternary</b>	Bond Formation
<b>Holocene</b>	"New Douglas" Shale Member
<i>Alluvium, soil, loess, and lake sediments</i>	<i>Blue-gray, grading upward into yellow-gray, silty laminated shale</i>
<b>Pleistocene</b>	"Grand Avenue of Springfield" Shale Member
Wisconsinan	<i>Blue-gray below, brown near top; well laminated and silty</i>
Till, loess, alluvium, and lake sediments	
Sangamonian	Modesto Formation
Loess and soil	"East St. Louis" Shale Member
Illinoian	<i>Blue below, poorly laminated, gray above, massive to well bedded, sandy</i>
Till, alluvium, loess, and lake sediments	
Yarmouthian	Farmington Shale Member
Loess, soil	<i>Blue-gray to grayish brown, poorly laminated, many siderite concretions in some areas, weathers brown; grades laterally from well laminated to massive</i>
Pre-Illinoian	Carbondale Formation
Loess	Underclay below Danville (No. 7) Coal Member
Till, alluvium, and lake sediments	<i>Clay is soft, light gray, slickensided (in Vermilion County, clay is red); much limonite present on most outcrops; lower part calcareous in La Salle County; in Grundy and Vermilion Counties, it is more kaolinitic than other underclays of the Carbondale Formation</i>
<b>Tertiary</b>	Lawson Shale Member
<b>Eocene</b>	Mottled green and gray, silty, massive shale
Wilcox Formation	Energy Shale Member
	Gray, clayey to sandy, some areas containing concretions of calcareous concretions; plastic, poorly laminated
<b>Paleocene</b>	Underclay of the Herrin (No. 6) Coal Member
Porters Creek Formation	Expandable-rich in most areas; illite and kaolinite increase on east and northwest side of basin
Buff clay with deep desiccation cracks; below oxidized upper surface buff zone, clay is dark blue to black and variable in composition	Canton Shale Member
Clayton Formation	Blue-gray, weathers brown; well laminated
Sandstone, silty clay	Underclay of the Springfield (No. 5) Coal Member
	Expandable-rich in most areas
<b>MESOZOIC</b>	Purinton Shale Member
<b>Cretaceous</b>	Blue-gray, clayey near base; grades upward into sandstone; laminated in some areas
Baylis Formation (western Illinois)	Francis Creek Shale Member
Thin beds of silty montmorillonite and kaolinitic clay in sandstone	Dark blue-gray in lower few feet, gray and sandier in upper part
Owl Creek Formation (southern Illinois)	Spoon and Abbott Formations
Sandy, silty, glauconitic clay	Unnamed underclay of the Colchester (No. 2) Coal Member
McNairy Formation (southern Illinois)	Massive, gray to cream, slickensided, with occasional root traces; often kaolinitic and sometimes sandy
Lenses of sandstone, lignitic clay (mostly kaolinite with minor illite and smectite), and siltstones	"Greenbush" Shale Member
Tuscaloosa Formation (southern Illinois)	Light gray, slightly bedded, sandy
White clay cementing chert gravel and quartz sand; interstitial clay is kaolinite	"Macedonia" Shale Member
	Gray, weathered, thinly bedded; concretions
<b>PALEOZOIC</b>	
<b>Pennsylvanian</b>	
Mattoon Formation	
"Albion" Shale Member	
Blue-gray, well laminated, grading up into yellow-gray, sandy shale	

of loess reflects its associated tills and fluvial source area, which vary in montmorillonite (smectite):illite content. Therefore, loess in western Illinois typically contains 60 to 90 percent montmorillonite, loess derived from the Wabash River system contains abundant illite, and loess east of the Illinois River has an intermediate composition. Sangamonian interglacial time is represented by either a well-drained or a gleyed paleosol on the uppermost Illinoian tills. One or more Illinoian tills are commonly present, and they are distinguished from one another and from older tills by their illite and dolomite content (Glass and

Killey, 1986). Illinoian deposits cover most of the state beyond the Wisconsinan end moraine and often underlie Wisconsinan deposits as well. At most localities Holocene deposits and Wisconsinan loess, silt, and/or alluvium overlie the Illinoian (fig. 6).

In much of east-central, central, and west-central Illinois, a well-drained or gleyed Yarmouth paleosol that formed on the uppermost pre-Illinoian till underlies Illinoian deposits (Willman and Frye, 1970). One or more pre-Illinoian tills are commonly present below the Yarmouth paleosol. Pre-Illinoian deposits had either eastern or western sources (Willman and

Frye, 1970): high illite and dolomite content characterize material with an eastern source and high smectite and calcite content identify material with a western source.

The texture and mineral composition of glacial deposits that overrode bedrock or pre-existing Quaternary deposits vary, depending on the characteristics of the underlying material. In general, there are three distinctive mineralogical characteristics of Quaternary deposits (Glass and Killey, 1986; Willman, Glass, and Frye, 1963): (1) changes in the ratio of smectite (western source) to illite (northern and northeastern source), and chan-

#### Cheltenham Clay Member

*The Cheltenham Clay is the name given to a clay 3 to 30 ft thick that was deposited during the time cyclothem were being deposited in the more rapidly subsiding parts of the Illinois Basin. This clay, which most likely supported vegetation continuously during its accumulation, covered various intervals during Spoon and Abbott time. These materials, which contain from 50 to 100 percent kaolinite, are semirefractory to refractory clays. They burn from buff to white, depending on their iron content. The clay occurs only along the edge of the Illinois Basin, which was a stable area during Spoon and Abbott time. Equivalent units in the central part of the basin are less kaolinitic, but may have economic value. The origin, properties, and composition of the Cheltenham Clay are essentially the same as those of kaolinitic underclays in other localities and parts of the section*

#### Mississippian

##### Chesterian Series

###### Grove Church Shale

*Gray, fossiliferous, interbedded shale and limestone*

###### Kincaid Limestone

###### Cave Hill Shale Member

*Upper 15 ft consists of calcareous dark gray and greenish gray shale at the top and red and green shale below. The middle is limestone with interlayered dark gray to black shale. The basal shale, about 15 ft thick, consists of dark gray shale (locally black) and includes some silty shale and a little gray to dark gray and green siltstone*

###### Fraileys Shale

*Shale containing beds of limestone, siltstone, and sandstone; dominantly a dark gray shale with a bed of red shale below the top*

###### Ridenhower Formation

*Includes limestone and sandstone strata, greenish gray, but red, green, and brown shales are common*

#### Valmeyeran Series

##### Warsaw Shale

*Blue-gray shale, fossiliferous, containing strata of argillaceous limestone, quartz geodes, and beds of coarse silt and fine sand*

##### Springville Shale

*Greenish gray to dark brownish gray, clayey shale; in places, mottled red and green and called "calico shale"*

##### State Pond Shale Member

*In a bed of soft glauconitic shale at base of the Springville Formation; greenish gray; contains phosphate nodules*

#### Kinderhookian Series

##### Hannibal Shale

*Green to gray, argillaceous siltstone*

##### Nutwood Shale Member

*Dark brown to black, slightly calcareous to noncalcareous; grades laterally and vertically into gray Hannibal Shale*

#### Devonian

##### Saverton Shale

*Bluish to greenish gray, silty, with thin, sandy and calcareous strata*

##### Grassy Creek Shale

*Black shale, hard*

##### Sweetland Creek Shale

*Gray and green shale; contains some dark gray and black strata*

##### Blocher Shale

*Lower 8 to 10 ft black, upper 6 ft gray*

##### Lingle Formation

###### Misenheimer Shale Member

*Dark gray to gray-brown, calcareous*

#### Ordovician

##### Neda Formation

*Red shale interbedded with red-brown or black oolite containing spheroids of goethite or hematite; in a few places, contains layers of gray or green shale*

##### Brainard Shale

*Greenish gray to green shale, dolomitic, silty*

##### Scales Shale

*Orchard Creek Shale Member (in extreme southeastern Illinois only)*

*Blue-gray upper part interbedded with limestone*

##### Claremont Shale Member

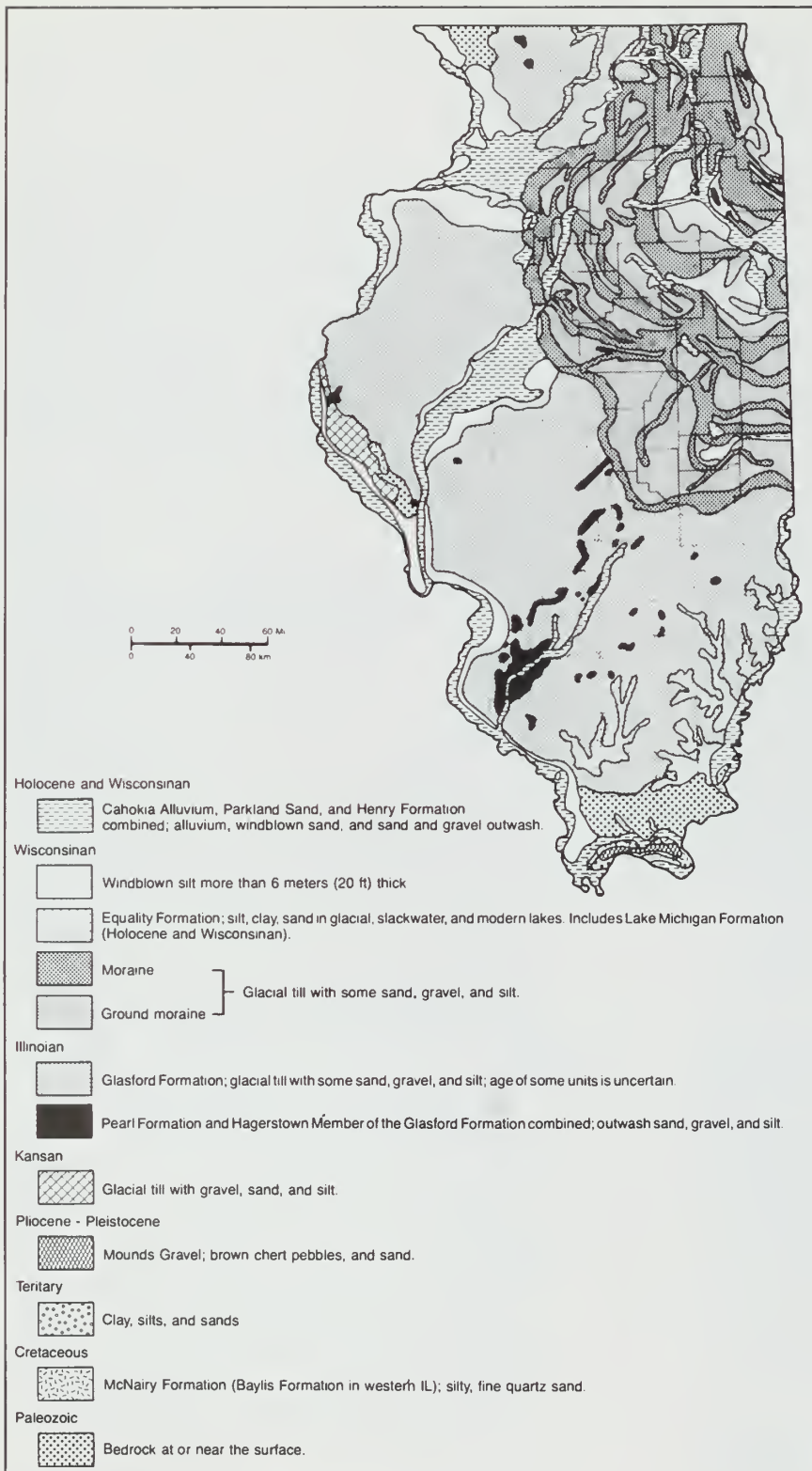
*Dominantly gray shale interbedded with limestone*

##### Elgin Shale Member

*Dominantly dark gray shale; in places, all shale, but usually interbedded with limestone, dolomite, siltstone, and sandstone*

New Albany Shale Group

Maquoketa Shale Group



**Figure 6** Surficial geology of Illinois (after Lineback, 1979).

ges in the ratio of calcite (western source) to dolomite (northern and northeastern source); (2) significant kaolinite content in deposits of glaciers passing over Pennsylvanian bedrock, especially basal or bedrock tills; and (3) the formation of weathering products such as low- and high-charge vermiculite, well-crystallized and "smeared" smectite, and mixed-layered kaolinite/smectite, and the associated alteration of chlorite and illite (Frye, Willman and Glass, 1960; Willman, Glass, and Frye, 1966).

Sangamon and Yarmouth paleosols are extensively developed in Illinois. The paleosols are sometimes more than 3 meters thick; those in low areas of the paleosurface are usually covered by an accretion gley. The expandable clay minerals present in paleosols, pre-Illinoian tills, gleys, and backwater fluvial and lacustrine deposits are an untapped source of absorbent and/or barrier clays.

Pre-Illinoian and gleyed Illinoian diamictons contain larger amounts of expandable clays than do similar Wisconsinan units. However, the pre-Illinoian and Illinoian units contain larger amounts of kaolinite and are similar in quality to Wisconsinan deposits used as ceramic raw materials. Loess, tills, and lake deposits can be used as raw materials for fired-clay products. However, tills and related materials may cause problems because they typically contain calcite and dolomite clasts, pebbles of all rock types, and pyrite/marcasite grains in unoxidized materials. Coarse-grained material can be removed by screening. Calcareous materials cause crumbling or "pops" in the product unless the product is treated with water or steam during cool-down (generally from 400°C downward). The beneficial effects of water treatment of these products during cool-down can be extraordinary. A fired body of Quaternary material that often turns to dust in a few weeks without water treatment develops normal fired hardness, strength, and integrity after water treatment. Water treatment also reduces long-term moisture expansion of brick made from any raw materials (White, 1964). In some cases, unweathered Quaternary materials contain enough pyrite/marcasite to cause SO<sub>2</sub> air-pollution problems upon firing that require the extra expense of pollution abatement.

Generally, Quaternary materials can be used advantageously to create unusual



aesthetic effects; for example, the "Chicago common" bricks (no longer produced) were available in a wide range of colors and color patterns. Although part of the effect was the result of firing in a scove kiln, the bricks also varied because of the heterogeneity of the feed material—a pebbly till from the southwestern suburbs of Chicago. Some Quaternary materials are blended with shales to add silt and sand and lighten shales that would otherwise form heavy, difficult-to-fire bodies. Overall, however, Quaternary materials are usually too low in quality to be a preferred source for raw materials.

### Tertiary

Significant Tertiary deposits are present in Illinois only in the southernmost counties of Alexander and Pulaski. A few Tertiary gravel deposits have been found north of this area, but the Paleocene Porters Creek Formation, 50 to 150 feet thick in these two counties (Kolata, Treworgy, and Masters, 1981), is the primary unit of interest for clay products. The Porters Creek Formation and associated Cretaceous and Tertiary deposits are the northernmost sediments of the Mississippi Embayment. They dip southward: the top of the Porters Creek Formation is about 350 feet above mean sea level where it is mined at Olmsted in Pulaski County, but only about 200 feet below mean sea level at Cairo (Kolata, Treworgy, and Masters, 1981).

The Porters Creek contains sandy and silty zones and varies significantly in mineral composition in the northern part of the Mississippi Embayment (Thomas and Murray, 1989). Near Olmsted the unit contains abundant montmorillonite, significant amounts of cristobalite, quartz, muscovite/illite, kaolinite, and traces of orthoclase, plagioclase, and chlorite. The cristobalite is poorly crystallized, but the X-ray diffractogram of this mineral is clearly more similar to cristobalite than to opal CT or tridymite. Some cristobalite is present as diatom tests, which are found mostly in the upper part of the section and are concentrated in the 1 to 2  $\mu\text{m}$  fraction. The montmorillonite content of the Porters Creek has been estimated to be 50 to 70 percent, although some of the properties of the clay are similar to those of materials having a higher montmorillonite content. It appears that the unexpected absorbent and decoloration prop-

erties of these clays may be partly attributable to the pore spaces within diatoms and partly to interparticle pores. According to Moll and Goss (1987), only interparticle pores are effective in absorbance.

As previously discussed, the Porters Creek Formation currently is used mainly for pet litter absorbent products, but oil absorbent or sweeping compounds, pelletizing binder for animal feeds, binder for foundry sands, oil decolorizing materials, and bulking and suspending agents for agricultural slurries also have been produced. Attempts have been made to add sodium to the clay and use it as a replacement for western bentonites. For some markets, the savings in shipping might offset the cost of additions of sodium.

### Mesozoic

The Cretaceous McNairy Formation crops out across southern Illinois to the north and east of the Porters Creek outcrop area and west of the Cache River. Although the McNairy is predominantly a sandstone, it contains abundant zones of kaolinite-rich clay. The unit ranges from 25 to 450 feet thick (Kolata, Treworgy, and Masters, 1981). Where present, the Levings Member in the middle section of the McNairy is a silty to lignitic gray to black clay; the upper and lower parts of the McNairy consist of "fine, white to gray, clayey, cross-bedded, micaceous, fine-grained quartz sand" (Kolata, Treworgy, and Masters, 1981). Potter and Pryor (1961) thought the sediments came from the southern Appalachians, and Pryor (1960) suggested that the succession represented the upper facies of a deltaic succession.

The clay mineral fraction of the McNairy is about 80 percent kaolinite with significant amounts of illite, muscovite, and smectite. Early settlers used these kaolins for pottery, although no pottery production exists at present. The materials could be separated during processing and marketed as quartz, muscovite, and clay; however, the economic viability of clay production from this formation—with or without beneficiation—is questionable. Production for local ceramic or cement plants has been evaluated at various times in the past and has shown economic promise.

Kaolins presumed to be Cretaceous in age have been mined near Anna in south-

western Illinois (Grim, 1934), and halloysites of unknown age have been discovered in southern Illinois (Lamar, 1942). The Anna kaolin, found in sinkholes, seems to be heavily weathered residual clay. The halloysites, on the other hand, are associated with mineralization in the fluorspar district and are assumed to be of hydrothermal origin. Both materials were used in ceramics production, and the kaolins were used for a few specialty markets. Exploration during the last two or three decades has failed to yield any new Anna deposits. The chances of finding halloysite are greater, and at least one new deposit has recently been located.

Cretaceous-age deposits also occur in Adams and Pike Counties in extreme western Illinois (Frye, Willman, and Glass, 1964). The Bayliss Formation contains zones in which the clays are thinner and perhaps coarser grained than the McNairy clays but have abundant kaolinite, montmorillonite, and "soil kaolinite" that is assumed to be mixed-layered kaolinite/smectite (Frye, Willman, and Glass, 1964). The deposits could provide ceramic or absorbent raw materials that would probably vary in value as a function of demand in nearby markets. However, access to the nearby Illinois, Mississippi, and Ohio Rivers could greatly extend the potential markets of clay products produced from the Porters Creek, McNairy, and Bayliss Formations.

### Paleozoic

The Paleozoic in Illinois can be divided, for the purpose of this discussion of clay products, into Pennsylvanian and older strata (fig. 7). Pennsylvanian shales and underclays have furnished the raw material for most of the fired-clay products made in recent years. The lower Pennsylvanian units, sandier and more kaolinitic than the upper ones, are presumed to represent fresh to brackish water deposits; shales, coals, and limestones reflect the increasingly fine, more marine characteristics of the upper part of the section. Alternating layers of sandstones, underclays, coals, shales, and limestones frequently occur in cyclical sequences called cyclothems (Wanless and Weller, 1932), which represent repeated cycles of marine invasion and retreat. The vertical variation of Pennsylvanian strata has an important bearing on the uses of these clays because relatively

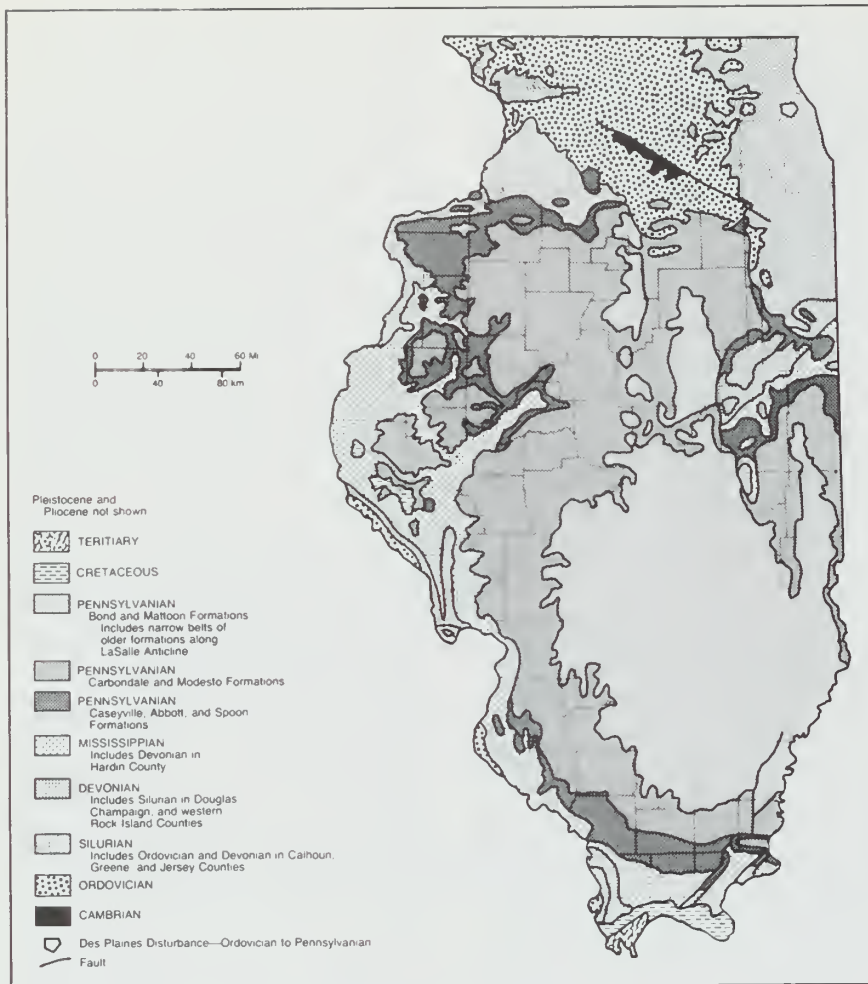


Figure 7 Bedrock geology of Illinois (after Willman et al., 1967).

refractory, kaolinitic underclays are often located near thick, upward-coarsening shales. Thus, products with a range of colors and degrees of refractoriness can be produced at a single location. The variations in clay content of shales make it possible to control the feed clay so that it is clayey enough for extrusion yet silty and sandy enough for rapid and complete firing. The rapid compositional variations in these strata reflect their complex origin.

The clays and shales of the Pennsylvanian vary from flintclays (Hughes and White, 1969), to fireclays such as those in the Cheltenham Clay, to illite-rich units, to underclays with a high mixed-layered illite-smectite content. Different suites of clay minerals or facies tend to be spatially distributed in a regular way (Parham, 1964; Odom and Parham, 1968; and Hughes et al., 1987). Details of the distribution and content of the mineral suites

suggested to Hughes et al. (1987) that illite-rich sediment was the source material and that kaolinitic and illite/smectite-rich clays were formed within the basin, primarily as a result of the growth, decay, and diagenesis of plants.

Pennsylvanian raw materials for fired-clay products tend to be kaolinitic (for refractories) or illitic (for common ware). Fired-clay products are difficult to produce from Pennsylvanian materials such as clays and shales containing siderite, calcite, or dolomite concretions, pyrite or marcasite, or abundant expandable clay minerals. Materials that have (1) a range of mineral constituents providing varying degrees of refractoriness through a wide firing range, (2) a silty texture, and (3) adequate plasticity are superior raw materials for fired ware.

Widespread underclays rich in expandable mixed-layered illite/smectite should be usable as sorbents or imperme-

able barriers. The organic content of these underclays may also increase their absorbent capacity for immiscible organic wastes.

Shales and claystones from strata older than the Pennsylvanian tend to have narrower firing ranges because of the absence of kaolinite, are often calcareous, and do not contain the range of clay minerals required for optimum plasticity and firing. For this reason, older shales have usually been exploited only where a local market is at some distance from Pennsylvanian sources. For example, older shales may be locally valuable for fired products in the Chicago area.

Bohr and Ehrlinger (1968); Kahn, Bhagwat, Baxter, and Berggren, 1986; and Kahn, Bhagwat, and Baxter, 1986, showed that the kaolinite in the pores of sandstone of the St. Peter (Ordovician) in northern Illinois could be isolated from tailings or mined from tailings ponds. There are also a few reports of kaolinitic-rich occurrences, often as sinkhole or crackfill deposits, in Silurian or Ordovician-aged strata (Frye, Willman, and Glass, 1964). These occurrences are small and erratically distributed and are not expected to yield economically minable deposits.

## FUTURE DEVELOPMENT

The potential for improving or expanding existing clay businesses and creating new ones seems bright to us, primarily because of the potential market area in and around Illinois. Many factors have contributed to increases and decreases in various sectors of this business. The successes here and in other states with similar advantages and constraints can be used as a guide for expanding or creating new enterprises in sectors that have declined. Although it is virtually impossible to isolate a single factor as the cause of business declines, investment practices, management philosophy, labor costs, transportation, energy, innovation, product demand, new resources, and new uses all contribute to the success or failure of clay businesses. The following short discussion suggests ways to manipulate these factors advantageously.

In the fired-clay products industry, investment in new, automated plants and a return to a long-term view of returns would greatly improve chances of success. Rather than remaining at or returning to older sites of production, industries

should locate new mines and plants on the basis of the qualities of raw materials, transportation, markets, labor, taxes, energy availability, and cost. For example, a company may receive federal and state aid and reduce costs by using coal rather than natural gas or petroleum. Abandoned underground coal mines are a potential source of cheap gas. Furthermore, new automated plants, which are more energy efficient and more productive, also can reduce another major cost: products that fail to meet specifications. Examples of potential new markets include field tile (Illinois is the northernmost and westernmost source of clay field tile), and "Chicago common" brick, which is no longer manufactured. However, competition from plastic field pipe may make new ventures in field tile economically risky.

Patio, sidewalk, and landscaping brick and tile are increasingly in demand. Some of the clay products imported to Illinois fail under local weather conditions, and a return to patio brick and tile of the quality of the previously mentioned Galesburg paving brick would be a significant selling point for an Illinois producer. In markets such as field tile, roof tile, and housing brick, the clay industry can make a strong case for long-term returns in spite of higher initial costs.

Although difficult to manage, co-mining ventures such as extracting underclay, coal, shale, and perhaps limestone from one mine can greatly increase profitability. Reject materials such as the kaolin-rich tailings from silica sand production may represent an ideal and inexpensive source of raw materials for one party and a reduction of cost or even a small profit from waste disposal for the other. Eventually it may be possible to process certain shales for oil, trace metals, alumina, soluble silica, and products such as glass building panels, fiberglass insulation, synthetic zeolites, and glass or synthetic minerals for isolation of radionuclides. Coal processing wastes may also provide a source of raw materials for clay products.

The study of the relative value of materials for existing and new markets is a research area we believe would produce large returns from relatively small investments. Evaluation of different geological materials such as sorbents or barriers to waste migration is an obvious need. Among the candidate absorbent clays are

those in underexploited zones in the Porters Creek; expandable-rich underclays; and clay zones from loesses, tills, paleosols, accretion gleys, and lacustrine deposits. Fired-clay research should evaluate ceramic raw materials on the basis of whole-sample mineral content, and Odom and Parham's (1968) study of the petrography of underclays should be extended to newer samples and materials other than underclays. In addition, whole-sample characterizations should often include the engineering value of low-cost clay materials. Generally, we have found that collecting precise information about the composition, properties, and relative merits of materials for particular uses yields important long-term benefits, many of which are not apparent during data collection and analysis.

### SUMMARY

Illinois has a rich clay-products history. Although most fired-clay products have declined in value in the recent past, absorbent clays continue to grow in value. Clay materials are variably distributed in Cenozoic, Mesozoic, and Paleozoic strata, and are widespread geographically. Because of their special properties and material concentrations, the Porters Creek Formation, Pennsylvanian shales and underclays, and Quaternary units provide all the materials currently used in Illinois clay production; they vary respectively from most to least significant in terms of value of production. Several avenues exist for new or improved clay businesses. The most important factors in improving strategies for improving profitability are optimizing site, raw material, and product selection; using raw materials in new ways; obtaining more precise data on the composition of raw materials; investing in automated plants that efficiently use low-cost energy sources; and taking advantage of "waste" materials that have unexpected uses.

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# Effects of Rock Types in Illinois Gravels on Freeze-Thaw Test Beams

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## ABSTRACT

*The percentage of chert gravel, especially low-specific-gravity (<2.35) chert, in natural aggregate samples used by the Illinois Department of Transportation (IDOT) to make portland cement test beams correlated strongly with expansion of the beams due to freeze-thaw testing (ASTM C666-77). Not exceeding 3 percent by weight in any sample, low-specific-gravity chert showed a very strong statistical relationship to expansion. Although no sample contained more than 1.5 percent by weight of ironstone, ironstone also correlated strongly with expansion. Silty dolomite probably caused some expansion. Other minor rock types considered suspect were weathered carbonate, cherty carbonate, pyritic dolomite, sandstone-siltstone, conglomerate, shale, and possibly weathered rocks in general. These statements apply only to gravels approved by IDOT for use in portland cement-based highways and to the percentages of rock types found in the study samples.*

*Samples collected from selected gravel-producing areas were studied by two methods. In Phase I, "as received" gravel was freeze-thaw tested and rock types were identified on surfaces of slabbed test beams. Descriptions of polished surfaces of the slabs suggested that cracks formed in and propagated from expanding pebbles were the only visible cause of the measured beam expansion. In Phase II, individual rock types were sorted, grouped, incorporated into beams, and tested. Statistical analyses of these data helped provide the conclusions summarized above; however, the data were often difficult to interpret and less than conclusive. The variability of the expansion data, due partly to the relatively small number and inconsistent positions of highly expansive pebbles in the cores of test beams, was a major problem in the statistical analysis of the effects of aggregate lithology on freeze-thaw expansion.*

## INTRODUCTION

An Illinois Department of Transportation (IDOT) statewide survey of Illinois portland cement concrete highways revealed that "D-cracking" (deterioration cracking) was a major contributor to premature pavement failure, which requires replacement or major repairs to highways in many parts of Illinois (Traylor, 1982). The term D-cracking generally refers to "fine, closely spaced cracks which occur parallel and adjacent to longitudinal and transverse joints, intermediate cracks, and the free edges of pavement slabs" (Stark, 1976). A major cause of D-cracking is the use of certain coarse aggregates that are susceptible to fracturing by frost action (Klieger et al., 1974).

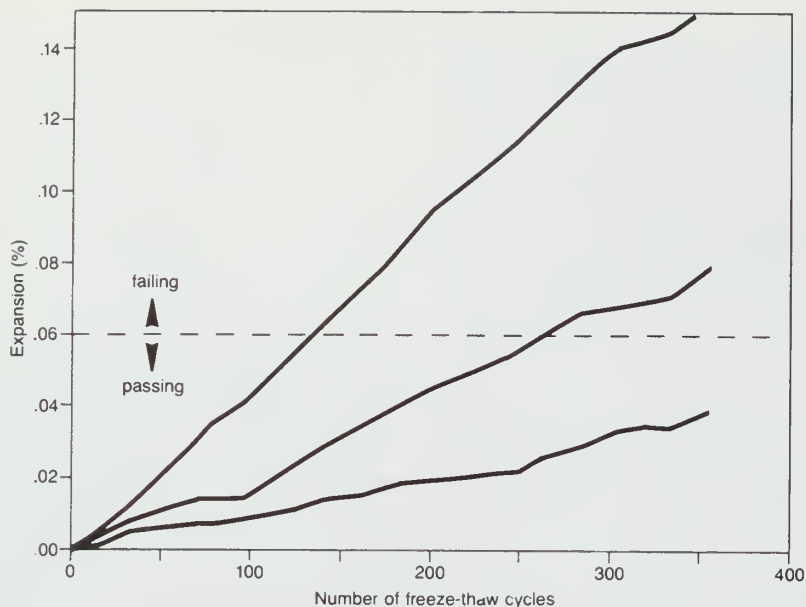
Because existing quality-control tests were not identifying many of these troublesome aggregates, IDOT began freeze-thaw testing all gravel sources previously approved for use in portland cement concrete pavements, using ASTM C666-77 Method B (ASTM, 1979). In this program, three concrete test-beams (3x4x15 inches) are prepared for each aggregate sample. The test beams are subjected to 350 rapid (3-hour) freeze-thaw cycles; they are frozen in air at 100 percent humidity and thawed in water. The sample does not meet IDOT criteria if the average expansion of the three test beams is greater than 0.06 percent.

Freeze-thaw expansion data for replicate test beams made from some gravel sources showed considerable variation

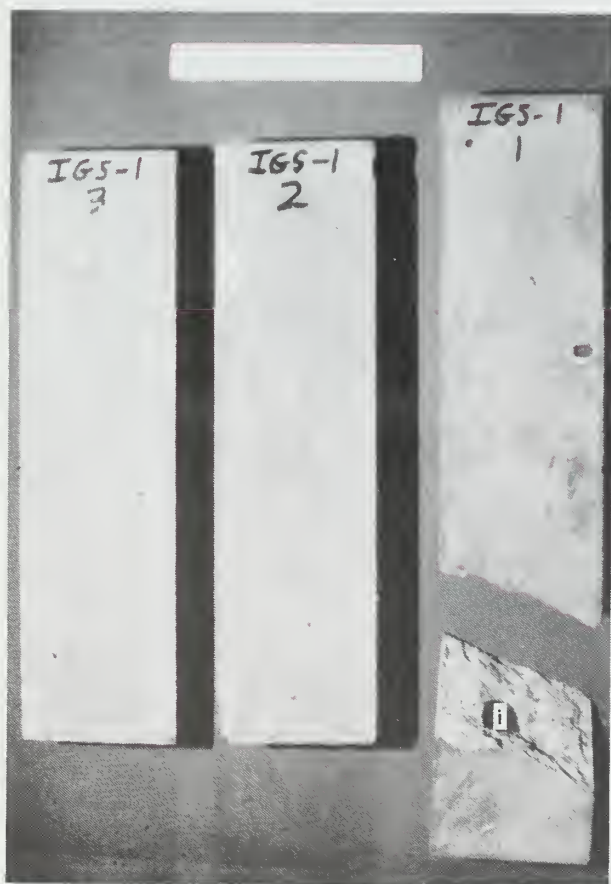
(fig. 1). In one case, expansion of a single piece of ironstone (an oxi-iron-hydroxide concretion) caused a test beam to break (fig. 2) while the other two beams in the set expanded only moderately. Results of repeated tests of aggregate from these gravel sources were also variable, and it was difficult to correlate test results with field performance data. Reduction in the maximum size of the gravel aggregate did not always eliminate these problems.

As a result of the freeze-thaw testing, IDOT rejected 20 previously accepted gravel sources (29% of the state's total); this action greatly reduced competition and altered the statewide distribution of gravel sources for concrete. For example, in the central Illinois (Springfield-Peoria) region, gravel pits had been the only approved local sources of concrete-quality aggregate. Most of these sources were disqualified on the basis of freeze-thaw testing, and aggregates had to be transported from greater distances. As haul distances increase, the delivered price of aggregates increases substantially. For example, at a distance of 50 miles from a quarry, the price of crushed stone can be three times the cost at the quarry (Subhash B. Bhagwat, personal communication, 1987). The obvious effect is a significant increase in the cost of construction projects.

In response to concern about the decreased availability of concrete-quality aggregates and unexplained variations in the freeze-thaw test data, IDOT and the Illinois State Geological Survey (ISGS) began a cooperative research project (Masters and Evans, 1987) to identify the rock types in gravel that cause freeze-thaw expansion and attempt to explain the variability in previous test results. This paper is a condensed version of the final report for that project. The project was suggested by George Dirkes, Executive Director of the Illinois Association of Aggregate Producers.



**Figure 1** Inconsistent expansion curves resulting from the freeze-thaw testing of three replicate beams from gravel sample number 11.



**Figure 2** Beam 1 of this triplicate set of freeze-thaw tested beams from sample 1 was broken by the expansion of a large ironstone pebble (i) near the axial line. The sketch shows the orthorhombic shape of a test beam, the measuring pins protruding from the centers of the top and bottom ends, and the axial line connecting the pins.

### SAMPLE SELECTION

Most of the gravel deposits that yield coarse aggregate for use in concrete are in the northern half of Illinois (fig. 3). To ensure that the study samples would be as representative as possible, the geology of the gravel deposits and the freeze-thaw test results from each source were reviewed.



**Figure 3** Gravel pits from which gravel-aggregate samples were obtained for this study.

Gravel deposits that are current sources of concrete-quality coarse aggregates in Illinois are all of glacial origin. These pits, located within the Holocene and Wisconsinan map unit (fig. 4), recover gravel from the Henry Formation (Willman and Frye, 1970). A few Wisconsinan-age sand and gravel deposits are underlain by older glacial gravel deposits of sufficient quality to be mined along with the Wisconsinan gravel. Gravel deposits in northern (especially northeastern) Illinois contain the greatest amounts of durable and unweathered rock types, mostly dolomite and relatively small amounts of igneous and metamorphic rocks (Masters, 1983, table 1). Henry Formation gravels progressively farther to the south tend to contain greater amounts of weathered pebbles and potentially deleterious rock types

such as chert, ironstone, sandstone, siltstone, shale, and coal.

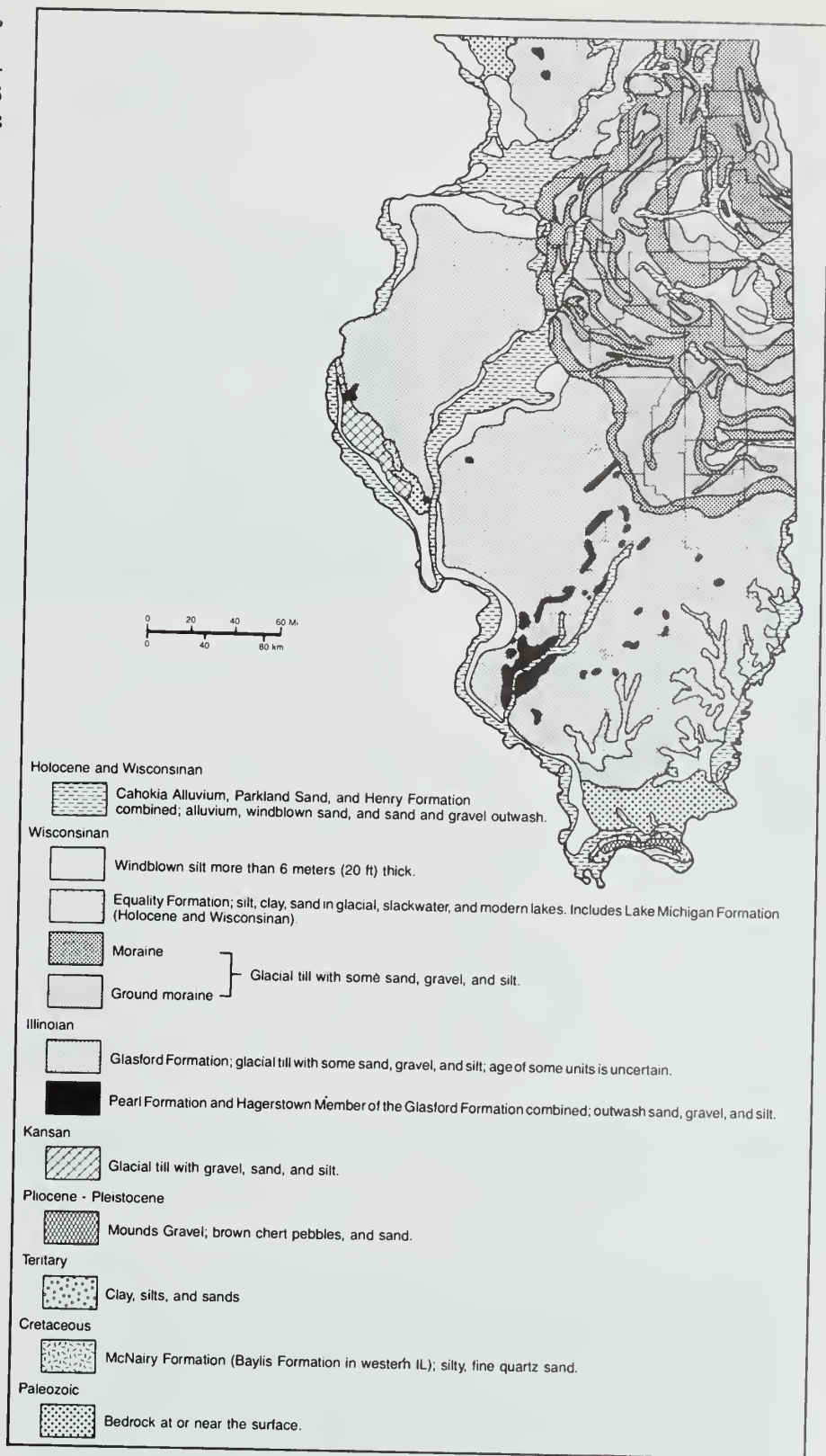
Samples were chosen from outwash-plain deposits in northeastern Illinois (3 samples) and from valley-train deposits in five river valleys: the Rock (4 samples), Fox (2 samples), Illinois (3 samples), Kickapoo (1 sample), and Wabash (3 samples) (fig. 3). All samples were collected by IDOT staff from production stockpiles containing nominal 3/4-inch top-size gravel. Two specially prepared samples, 5a and 5b, both taken from the same source, contained no particles larger than 3/4 inch; they were included in the study to determine whether the physical properties of a gravel aggregate will differ significantly when the gravel is (1) crushed with a jaw crusher or (2) crushed with an impact crusher. An additional sample from the Mississippi River valley source was compared petrographically with the other 16 samples but was not freeze-thaw tested.

### CLASSIFICATION OF ROCK TYPES

The classification scheme used in both Phase I and Phase II of the study to identify the constituents of the gravel samples was designed to (1) allow for the possibility that certain rock types or physical properties of gravel may be more likely than other rock types or properties to cause deleterious behavior of aggregate in portland cement concrete, and (2) measure the rock-type variability of these gravel samples.

The rock-type names used were based on accepted rock and mineral names, and textural and compositional terms (Bates and Jackson, 1980; Pettijohn, 1957; Moorhouse, 1959; and Pirsson and Knopf, 1947). Many rock types were classified by specific rock names, such as dolomite and limestone, or rock names coupled with modifiers, such as laminated dolomite. Other rock-type names were derived by combining certain types of rocks having similar compositions or origins. Each rock type is described and photographed in Masters and Evans (1987, appendix). A 10-power hand lens and a low-power binocular microscope (fig. 5) were used to identify rock types.

Significant textural and mineralogical variations were observed in the dolomite, the most abundant rock type; therefore, it was classified into four dolomite subtypes: (1) dolomite, relatively pure and



**Figure 4** Simplified geologic map of surficial unconsolidated materials, Pleistocene (glacial age) and older, in Illinois (after Lineback, 1979 and 1981).

nonfriable; (2) laminated dolomite, having any type of visible layering; (3) silty dolomite, similar to the first two subtypes but permeable and containing silt, sand, or clay; and (4) pyritic dolomite, similar to any of the other three but containing observable pyrite (Masters and Evans, 1987).

Rock-group names were used as rock types for metamorphic and igneous rocks, which were present in relatively small amounts in the samples. However, some metasedimentary pebbles found in relatively large amounts were given specific rock-type names (i.e., quartzite and metagraywacke) on the basis of their textures and mineral contents. Such distinctions for abundant rock types can be critical in evaluating freeze-thaw expansion; other studies have shown that a specific rock such as metagraywacke causes significant expansion (Stark, 1976).

**TESTING OF PROCESSED GRAVEL SAMPLES: PHASE I**

**Experimental design**

In Phase I, IDOT personnel obtained samples from the selected sources and sieved them into four size-ranges. Sixty pounds of material was split for each

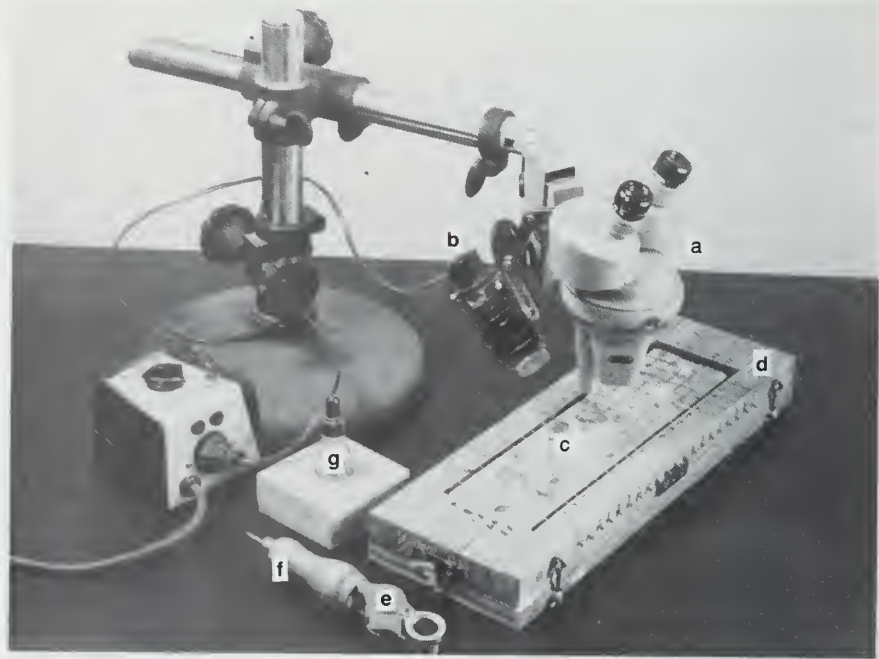


Figure 5 Equipment used to identify, sort, and count rock types: (a) binocular microscope; (b) light; (c) slab from a test beam; (d) point-counting box; (e) combination hand lens, and (f) hardness tester; and (g) acid bottle in holder.

three-beam freeze-thaw test: 3 pounds of 1- to 3/4-inch material, 24 pounds of 3/4- to 1/2-inch material, 12 pounds of 1/2- to 3/8-inch material, and 21 pounds of 3/8-

inch to number 4 mesh-size material. The coarse aggregate, sand, and cement were mixed and cast into test beams, and the beams were freeze-thaw tested.

The beams were then sawed into slabs, and selected slab surfaces (fig. 6) were ground smooth to facilitate point-counting. The pebbles exposed on the slab surfaces were identified and counted; the cracked pebbles in each rock type were also counted, and cracks in the matrix were sketched. As a first attempt to establish which rock types caused expansion, statistical analyses were made relating point-count data to expansion values. Results of other IDOT quality-control tests on splits of the study samples also were compared with Phase I expansion values.

**Beam expansion, popouts, cracks**

Freeze-thaw deterioration features such as cracks and surface ruptures (popouts) were visible on the exterior surfaces of the test beams. A study of the types of rocks involved in popouts revealed that only six rock types were found in the cores of popouts: chert was the most abundant, followed by ironstone and weathered carbonate. Comparison of the total numbers of pebbles in popouts with the average expansion values for the Phase I test beams suggested that popouts

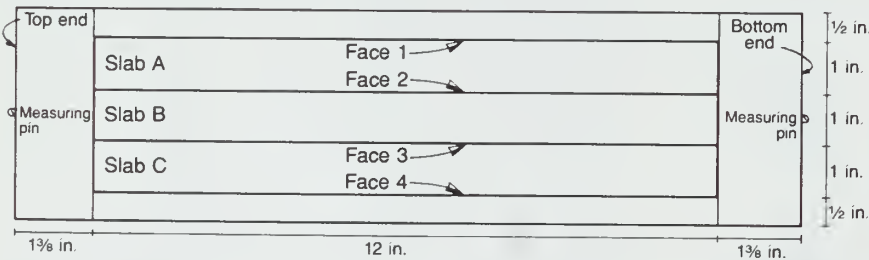


Figure 6 Diagram of the top surface of a freeze-thaw test beam, indicating measurements and lines for rock-saw cuts. Slabs A, B, and C were cut from the central part of the beam. Both cut surfaces of slabs A and C (faces 1, 2, 3, and 4) were studied for rock-type contents and deterioration features. Measuring pins protrude from the top and bottom ends of the beams.

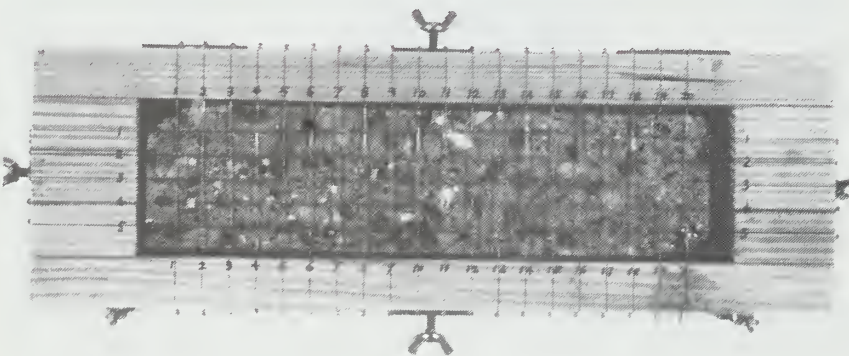


Figure 7 Point-counting box containing a slab (3x12 inches) under the wire grid.



**Table 1** Average point-count percentage and average freeze-thaw expansion percentage of each rock type found in the triplicate test beams of each gravel sample (Phase I).

Sample number	1	2	3	4	5a	5b	6	7	8	9	10	11	12	13	14	15
Average percent expansion	0.028†	0.016	0.030	0.081	0.059	0.041	0.062	0.062	0.030	0.067	0.063	0.089	0.082	0.067	0.066	0.063
<b>Sedimentary rocks</b>																
Dolomite	65.9	63.3	65.5	45.4	54.1	43.8	45.8	55.9	63.0	29.3	25.5	41.9	33.9	21.2	22.2	18.8
Laminated dolomite	1.1	2.7	3.3	2.5	2.7	2.6	1.7	3.1	0.7	3.0	0.6	0.9	2.0	1.1	1.7	0.3
Silty dolomite	1.1	1.0	1.2	1.5	1.3	0.2	1.0	1.9	2.0	5.2	3.0	2.2	2.3	0.9	2.4	0.3
Pyritic dolomite	1.7	0.6	0	0	0	0	0.3	0.1	1.6	0.2	0	0	0	0	0	0
Limestone	3.2	4.4	3.9	4.9	1.8	3.4	2.8	3.9	3.1	11.4	9.6	7.4	8.1	10.5	11.8	7.5
Cherty carbonate	4.6	4.9	1.6	2.9	3.2	3.5	2.9	5.5	5.1	3.7	2.6	4.5	4.4	3.1	3.8	6.3
Chert	7.2	6.3	7.7	22.0	20.3	23.8	23.6	10.2	9.8	15.4	24.5	16.0	19.2	22.4	18.0	24.9
Weathered carbonate	5.0	3.8	3.7	3.6	3.2	4.0	3.4	7.4	4.7	14.1	2.7	4.8	5.7	5.4	2.8	5.4
Ironstone	0.3	0.2	0.4	0	0.3	0	0	0	0.1	0.6	0	1.0	0.8	0.4	0.2	0
Shale	0	0	0	0	0	0	0	0	0	0.2	0	0.5	0.3	0.5	1.4	0
Sandstone-siltstone	1.1	1.7	1.6	3.7	2.0	3.4	2.7	1.7	1.3	4.6	2.4	5.5	3.7	4.2	4.7	9.0
<b>Igneous rocks</b>																
Mafic	2.1	2.7	2.7	1.2	1.2	3.2	1.8	1.7	2.1	0.9	2.0	2.4	1.5	2.9	10.1	3.1
Weathered mafic	0.6	0.2	0	0.3	0.2	0.3	0.6	0.3	0.3	0	0.2	0.3	0.7	0.5	1.4	1.0
Coarse felsic	0.3	0.3	0	0.2	0	0	2.4	0	0.4	0	4.5	0.2	0.2	0.9	0.4	0.6
Weathered coarse felsic	0	0	0.2	0	0.2	0	0	0	0	0	0	0	0	0	0	0.2
Fine felsic	0.3	0.2	0.7	0.3	0	0.5	0.4	0	0.1	0.2	2.3	1.4	1.5	0	0	0.5
Massive quartz	0	0.5	0.6	1.0	1.0	0.2	2.1	0.9	0.3	2.5	3.9	1.2	1.3	2.5	1.7	3.6
<b>Metamorphic rocks</b>																
Gneissic	2.3	3.0	3.0	3.2	3.8	3.7	2.7	2.6	4.1	2.1	3.6	3.3	4.1	3.4	4.7	5.2
Weathered gneissic	0.5	0.2	0.3	0	0	0	0	0	0	0.2	0.8	0.2	0.2	0.5	0	0.5
Schistose	0.5	0	0	0	0	0.5	0	0	0.3	0.2	0.2	0	0	0.2	0.2	0.6
Weathered schistose	0.2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Metasedimentary	1.5	2.2	2.2	4.6	2.5	3.2	3.6	3.0	0.3	3.7	8.0	3.1	5.7	11.9	6.6	7.0
Weathered metasedimentary	0.2	0	0	0	0	0	0	0.1	0	0	0.3	0.3	0.3	0.4	0.2	0.3
Metagraywacke	0.5	1.7	1.5	2.0	2.3	3.2	2.0	0.9	0.6	1.5	2.0	2.2	2.8	2.2	0.9	2.4
Tillite	0	0.2	0	0	0.2	0.3	0.1	0.3	0	0	0	0.3	0.2	3.3	1.9	1.2
Quartzite	0.2	0	0	0.7	0	0.2	0.3	0.5	0.1	1.1	1.5	0.3	1.5	1.6	3.1	1.5

\*Two rock types, conglomerate and weathered fine felsic, are omitted because none were found.

†Includes one projected expansion value for a broken beam.

probably relieve expansion pressure and are not directly related to measured expansion values. Examination of the slab surfaces cut from the test beams indicated that the most critical factor in determining whether or not an expansive pebble would cause a popout was its proximity to the surface of the beam. Expansive pebbles within the core (centered around the axial line between the measuring pins of the test beams) (fig. 2, sketch)—especially those pebbles near the heads of the measuring pins—probably contributed the most to expansion. These pebbles cracked, and the cracks often propagated

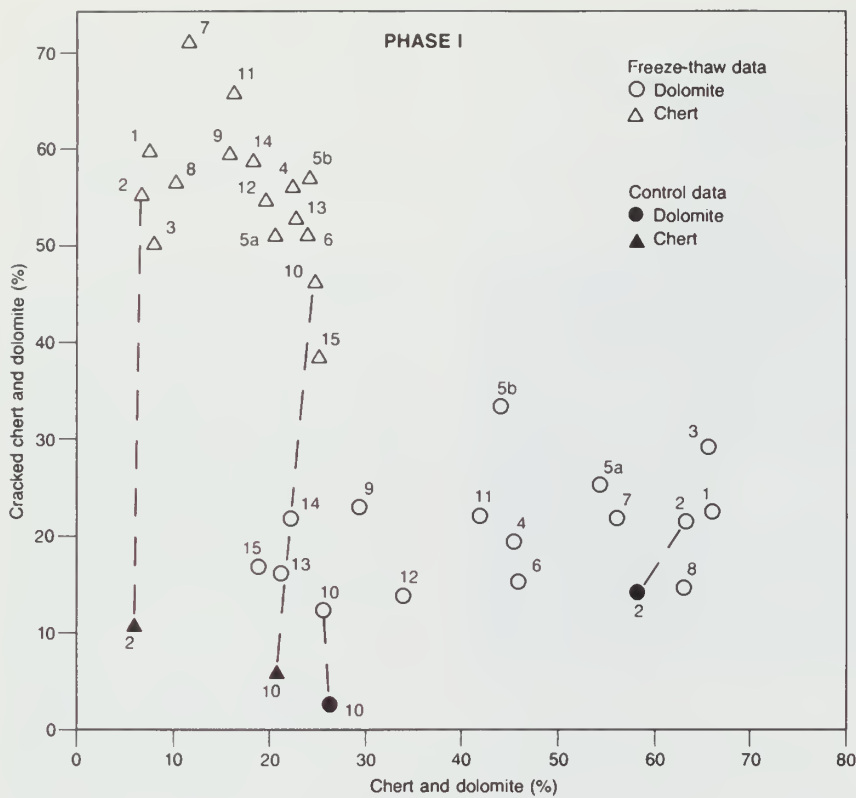
into the matrix material, forming three-dimensional networks that often extended to the beam surfaces, producing polygonal patterns.

#### Point-count data

Point counts of the rock types were made on slabs sawed from Phase I test beams overlain by a wire grid (fig. 7). Almost no reaction rims were found; reaction rims are evidence of a chemical reaction between the aggregate and the cement (Dolar-Mantuani, 1983).

The average percentage of each rock type found in each sample and the

average expansion value for each sample are given in table 1. These averages do not show the variability of the data within the 3-beam sets. For example, the chert contents of the three beams for sample 6 (27.9%, 24.6%, and 18.7%) varied directly with expansion; however, this relationship was not consistent. For sample 10, just the reverse was true; the highest chert content (27.5%) was in the beam that expanded the least (0.035%) and the lowest chert content (24.8%) was from the beam that expanded the most (0.082%) (Masters and Evans, 1987, tables 4, 5, and 6).



**Figure 8** Comparison of the percentage of chert and dolomite pebbles in the test beams with the percentage of cracked chert and dolomite pebbles in the test beams (point-count data). Each point is identified by its sample number. Dashed lines connect values for the percentage of cracked chert and dolomite pebbles in control samples not freeze-thaw tested with equivalent values for freeze-thaw tested samples.

The average percentage of chert found in samples from the same area was also variable; the greatest variations were in samples from the Illinois River valley (16.0% to 24.5%, samples 10 and 11, table 1). More samples must be studied to evaluate such relationships more fully.

The major deterioration feature found on the slab surfaces was the abundance of cracked chert pebbles. The striking tendency of chert pebbles to crack and dolomite pebbles not to crack is illustrated by the clustering of points in figure 8. As a test of how much of the observed cracking actually occurred during

freeze-thaw testing, two samples (2 and 10) were cast into beams and prepared for point-counting without being freeze-thaw tested. The amount of cracked chert in sample 2 rose from 11 percent without freeze-thaw testing to 55 percent with freeze-thaw testing, whereas the amount of cracked dolomite increased from 14 to 21 percent under the same conditions. The cracked chert content in sample 10 increased from 6 to 46 percent, and the cracked dolomite content increased from 3 to 12 percent. Thus, freeze-thaw testing cracked 40 to 44 percent of the chert but only 7 to 9 percent of the dolomite in

**Table 2** Classification of rock-type groups for the Phase II series of freeze-thaw tests.

Group 1: chert
Group 2: dolomite
Group 3: laminated dolomite, pyritic dolomite, limestone, cherty carbonate
Group 4: coarse felsic, fine felsic, mafic, quartzite, metasedimentary, metagraywacke, tillite, gneissic, schistose, quartz
Group 5: ironstone, silty dolomite, sandstone-siltstone, shale, conglomerate, weathered coarse felsic, weathered fine felsic, weathered mafic, weathered metasedimentary, weathered gneissic, weathered schistose, weathered carbonate

samples 2 and 10. The similarity of the results from the two control samples leads to the conclusion that chert and dolomite in the other samples would probably react in much the same way.

## TESTING OF SELECTED ROCK-TYPE GROUPS: PHASE II

### Experimental design

In Phase II, freeze-thaw testing was conducted on individual rock types and groups of rock types to check and refine Phase I data. An additional split (60 lbs) of gravel in the designated weights of each size-fraction was taken from each bulk study sample. Each pebble in these splits was identified and sorted into the 28 rock-type categories used in Phase I (for each of the 16 samples about 12,500 pebbles were identified and sorted).

After the samples were separated by size and rock types, the beams were made. The amount of each rock type obtained was augmented by enough standard, nonexpansive crushed dolomite to make a new set of three test beams. For example, if a 60-pound sample split contained 10 pounds of chert pebbles, that 10 pounds of chert was mixed with 50 pounds of the standard dolomite to obtain the 60 pounds of coarse aggregate in the correct size-range for test beams. Any resulting beam expansion thus could be attributed only to chert.

For the first five samples studied in Phase II, every rock type except those present in trace amounts (<0.05%) was individually cast into a set of test beams. Freeze-thaw testing of these beams provided expansion data for dolomite and chert; however, the less abundant rock types generally did not cause detectable expansion, probably because there were too few of them in the test beams. Therefore, the procedure was modified for the last 11 samples; the same 28 rock types were separated, but for testing they were recombined into five rock-type groups (table 2).

These groupings were chosen to provide (a) independent testing for chert (group 1) and dolomite (group 2), the only rock types abundant enough to be individually tested in all samples; (b) a minimum of 6 pounds combined weight of rock types in the remaining groups (10% of the 60-lb sample); (c) a separate group of sedimentary rocks (group 3); (d) a separate group of unweathered igneous

**Table 3** Total percentage (by weight) of each rock type identified in each of the 60-pound gravel samples studied in Phase II.

Sample number	1	2	3	4	5a	5b	6	7	8	9	10	11	12	13	14	15	16*
<b>Sedimentary rocks</b>																	
Dolomite	59.7	61.1	56.3	41.5	52.1	47.1	49.1	57.6	62.2	32.0	21.3	34.7	28.6	15.2	19.6	20.2	1.6
Laminated dolomite	3.5	2.8	2.5	2.8	2.1	1.2	1.4	1.8	3.0	1.3	1.8	1.6	1.8	1.9	2.5	1.3	TR
Silty dolomite	1.6	2.1	3.3	2.7	1.9	1.6	0.4	2.1	0.8	6.2	0.9	3.0	3.0	2.6	2.3	0.7	0.1
Pyritic dolomite	1.3	1.1	2.7	0.3	0.7	0.8	0	1.5	0.3	0.4	0	0.3	0.5	0.1	0.2	0	TR
Limestone	5.1	3.9	4.4	3.6	3.1	2.3	3.2	2.7	4.3	7.7	6.1	8.1	9.7	9.4	10.9	9.4	1.8
Cherty carbonate	4.5	4.6	7.4	7.2	4.8	4.7	5.3	10.1	6.2	6.1	3.9	8.2	7.2	9.4	8.6	6.7	2.3
Chert	5.2	5.9	7.5	20.3	17.2	19.6	18.3	9.7	6.5	15.0	21.1	14.4	15.6	19.0	15.0	21.4	21.7
Weathered carbonate	5.1	2.9	3.2	4.4	3.2	2.1	7.0	4.0	4.6	8.5	6.6	4.9	5.7	2.0	2.3	2.1	0.5
Ironstone	0.5	0.6	0.3	0.1	TR	TR	TR	0.1	0.1	1.5	0.2	1.4	0.8	0.4	0.4	0.2	1.4
Shale	0.1	0.3	0.3	0.1	0.1	0.1	TR	0.1	TR	0.7	TR	0.5	0.4	0.6	1.3	0.1	TR
Sandstone-siltstone	2.9	2.1	2.7	2.8	2.0	2.4	3.2	1.6	2.9	5.7	7.1	4.2	4.0	5.9	5.2	8.2	1.3
Conglomerate	TR	0.1	TR	0	TR	TR	TR	0	TR	0.5	0.2	0.1	0.3	0.3	0.3	0.1	TR
<b>Igneous rocks</b>																	
Mafic	3.9	5.3	2.5	2.2	1.8	4.2	1.7	1.9	1.6	3.1	2.4	4.0	4.0	5.8	6.1	4.1	4.7
Weathered mafic	0.4	0.5	0.3	0.2	0.1	0.1	0.3	0.2	0.6	0.4	0.6	0.2	0.2	0.3	0.2	0.5	0.5
Coarse felsic	0.3	0.4	0.3	0.3	0.3	0.2	0.2	0.2	0.3	0.2	0.5	0.4	0.4	0.3	0.2	0.2	2.0
Weathered coarse felsic	TR	TR	TR	TR	TR	0	0	TR	TR	TR	0.1	TR	TR	TR	0	0.1	0.1
Fine felsic	0.4	0.4	0.3	0.5	0.5	0.8	0.6	0.4	0.3	0.2	5.2	1.0	2.1	0.1	0.1	0.3	9.6
Weathered fine felsic	0	0	0	0	TR	0	0	TR	0	0	0	0	TR	TR	0.2	TR	0.1
Massive quartz	TR	0.1	0.1	0.9	0.7	0.3	1.0	0.1	0.1	1.5	3.1	0.8	2.1	1.5	0.7	1.6	3.4
<b>Metamorphic rocks</b>																	
Gneissic	2.1	3.5	2.8	4.3	3.7	4.8	3.1	2.5	3.2	3.2	7.7	5.0	5.5	8.8	8.6	7.1	19.6
Weathered gneissic	0.2	0.2	0.2	0.4	0.3	0.2	0.4	0.2	0.5	0.3	1.3	0.2	0.3	0.5	0.5	0.7	2.1
Schistose	1.1	0.4	0.2	0.3	0.2	0.2	0.5	0.1	0.5	0.4	0.7	0.3	0.1	0.5	0.2	1.1	0.3
Weathered schistose	TR	TR	TR	TR	0	0	0.1	TR	TR	TR	TR	TR	TR	TR	TR	0.1	0.2
Metasedimentary	1.1	1.3	1.4	3.1	2.9	4.0	2.5	1.6	1.0	3.1	5.7	4.9	6.0	9.1	8.7	8.3	17.6
Weathered metasedimentary	TR	TR	0	TR	TR	0	0	TR	0	TR	0	TR	TR	TR	0.1	TR	1.1
Metagraywacke	0.8	1.0	1.1	1.2	1.3	2.2	0.8	1.3	0.8	1.4	2.3	1.3	1.2	1.1	0.8	1.3	4.5
Tillite	0	0	TR	TR	0.1	0.1	TR	0	TR	0.2	0.1	0.1	0.1	1.9	2.1	2.1	TR
Quartzite	TR	0.2	0.1	0.5	0.8	1.0	0.8	0.1	0.2	0.3	1.2	0.6	0.8	2.9	2.9	2.0	3.6

\*Sample obtained from product stockpile for rock type data only and not subjected to freeze-thaw or other quality tests.

and metamorphic rocks (group 4); and (e) a group of potentially deleterious rock types present in only very small amounts, including ironstone, silty dolomite, sandstone-siltstone, weathered rocks, and incompletely consolidated rocks (group 5).

Phase II rock-type data are summarized in table 3. Statistical analyses were made by comparing the weight percent of each of the five groups of rock types to the corresponding expansion data and then comparing the weight percent of each rock type within a group to the expansion data for its respective group.

### Rock-type variations

The major gravel deposits in Illinois from which the study samples were ob-

tained differ in their rock-type contents (table 3). In table 4, related rock types are combined; the samples are identified by stratigraphic names (Willman and Frye, 1970) and arranged by outwash-deposit area. Dolomite was most abundant (63% to 67%) in the McHenry County outwash plains and the closely related Fox River valley-train, and chert was least abundant (5% to 10%) in these deposits (table 4). All other samples were from valley-train deposits containing abundant chert (15% to 22%); the deposits containing higher amounts of chert tended to occur farther downstream at greater distances from the glacial-age ice fronts (figs. 3 and 4). Within each gravel production area the weight percent of several rock types varied widely; for instance, in the Illinois

River valley-train deposits, dolomite content varied from 24.0 to 39.6 percent and ironstone from 0.2 to 1.4 percent. The dolomite content was fairly distinctive in each valley-train system, ranging from 63 to 69 percent in the Fox, 47 to 57 percent in the Rock, 24 to 40 percent in the Illinois, 22 to 25 percent in the Wabash, to just 2 percent in the Mississippi River valley sample. The igneous and metamorphic rock contents (including weathered varieties) in the samples from the valley trains ranged from 9 percent in the Fox, 12 to 18 percent in the Rock, 19 to 30 percent in the Illinois, 30 to 33 percent in the Wabash, and 69 percent in the Mississippi River valley sample (table 4).

**Table 4** Summary of weight percent data for rock types identified in the study samples (table 3) and geologic names of deposits (fig. 4) and geographic sites from which samples were obtained (fig. 3).

DIVISIONS OF GEOLOGIC TIME Divisions of rock stratigraphy Geographic locations	Sample number	Rock types (wt %)							Igneous and metamorphic	Weathered igneous and metamorphic	
		Sedimentary					Sandstone-siltstone	Other sedimentary rocks			Ironstone
		Total dolomite	Limestone	Weathered carbonate rocks	Chert						
<b>QUATERNARY SYSTEM</b>											
<b>PLEISTOCENE SERIES</b>											
<b>WISCONSINAN STAGE</b>											
<b>Henry Formation</b> (outwash deposits)											
<b>Batavia Member</b> (outwash plains)											
McHenry County	1	66.1	5.1	5.1	5.2	2.9	4.6	0.5	9.7	0.6	
	2	67.1	3.9	2.9	5.9	2.1	5.0	0.6	12.6	0.7	
	3	64.8	4.4	3.2	7.5	2.7	7.7	0.3	8.8	0.5	
<b>Mackinaw Member</b> (valley trains)											
Rock River	4	47.3	3.6	4.4	20.3	2.8	7.3	0.1	13.3	0.6	
	5a	56.8	3.1	3.2	17.2	2.0	4.9	TR	12.3	0.4	
	5b	50.7	2.3	2.1	19.6	2.4	4.8	TR	17.8	0.3	
	6	50.9	3.2	7.0	18.3	3.2	5.3	TR	11.3	0.8	
Fox River	7	63.0	2.7	4.0	9.7	1.6	10.2	0.1	8.2	0.4	
	8	66.3	4.3	4.6	6.5	2.9	6.2	0.1	8.0	1.1	
Kickapoo Creek	9	39.9	7.7	8.5	15.0	5.7	7.3	1.5	13.6	0.7	
Illinois River	10	24.0	6.1	6.6	21.1	7.1	4.1	0.2	28.9	2.0	
	11	39.6	8.1	4.9	14.4	4.2	8.8	1.4	18.4	0.4	
	12	33.9	9.7	5.7	15.6	4.0	7.9	0.8	22.3	0.5	
Wabash River	13	19.8	9.4	2.0	19.0	5.9	10.3	0.4	32.0	0.8	
	14	24.6	10.9	2.3	15.0	5.2	10.2	0.4	30.4	1.0	
	15	22.2	9.4	2.1	21.4	8.2	6.9	0.2	28.1	1.4	
Mississippi River	16*	1.7	1.8	0.5	21.7	1.3	2.3	1.4	65.3	4.1	

\*Sample obtained from product stockpile for rock type data only and not subjected to freeze-thaw or other quality tests.

## STATISTICAL ANALYSES

The IDOT freeze-thaw expansion values for Phase I and Phase II test beams were compared (Statistical Analysis System (SAS), 1982 a,b) with the respective Phase I and II sets of rock-type data. Phase I expansion data were also compared with IDOT aggregate-quality test data on the same samples. The General Linear Models (GLM) (simple regression) and Stepwise Regression (SR) (multiple regression) procedures of SAS were used to determine which rock types were related to expansion.

Results of the statistical analyses were often difficult to evaluate or less than conclusive, and therefore must be used with caution. One problem was the relatively small number of samples analyzed

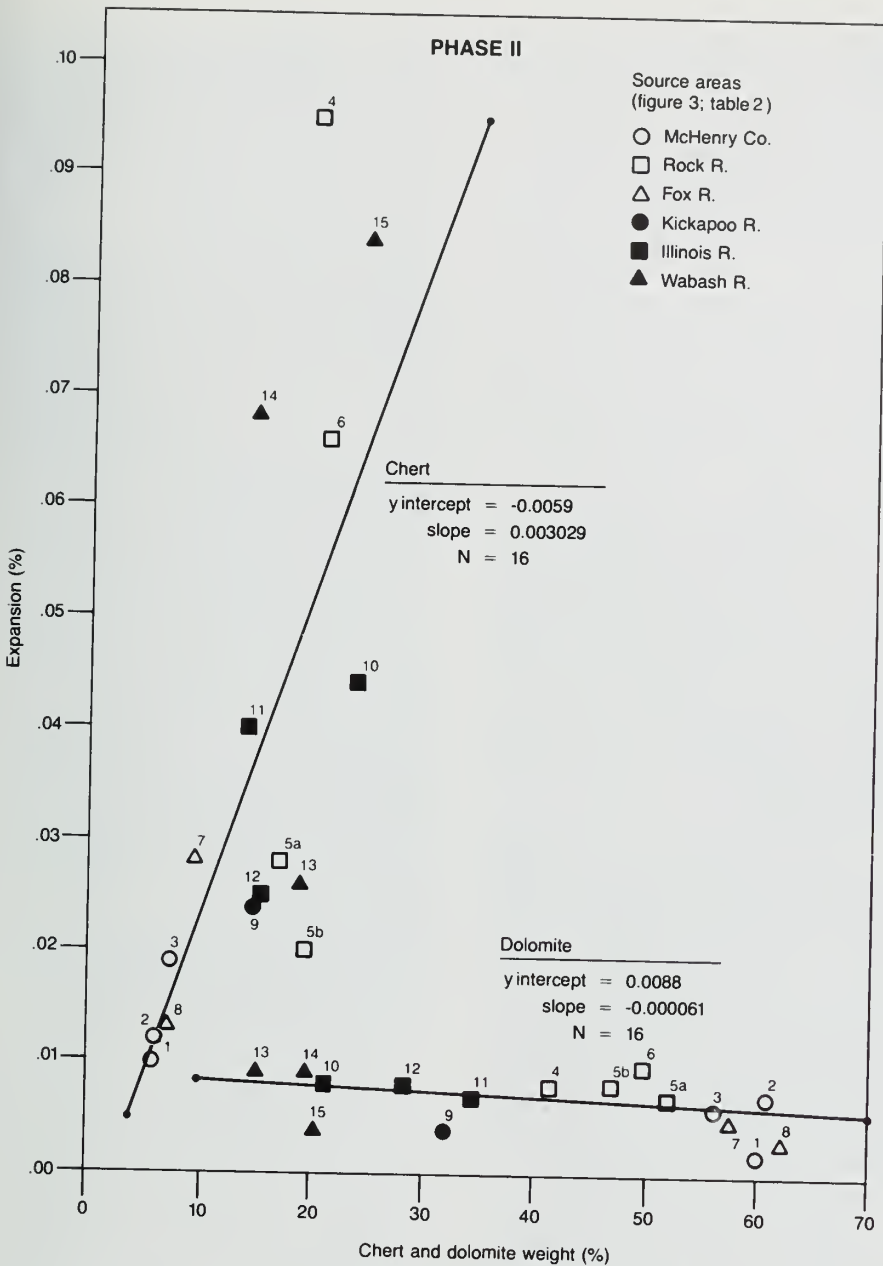
in comparison with the large number of unknowns (rock types) per sample. In addition, the relatively small number of expansive pebbles in the cores of the test beams probably increased the variability and reduced the reliability of the results. This paper includes only general statements about the statistical analyses; Masters and Evans (1987) contains detailed data and discussions.

GLM analysis of Phase I data indicated that the most abundant rock type, dolomite, is not directly related to expansion and that chert, sandstone-siltstone, and limestone may be related to expansion. Phase I SR data analysis generated a model consisting of the three rock types most likely responsible for expansion. Of these three, chert and ironstone were

strongly implicated and metagraywacke was weakly implicated in contributing to expansion.

Statistical analysis of Phase II data showed stronger correlations with expansion than did Phase I data; this result can be attributed to the fact that many more pebbles were identified, providing more accurate rock-type percentages, and many rock types were freeze-thaw tested in groups, effectively reducing the number of unknowns.

GLM analysis of Phase II data indicated that chert (group I) was the rock type most strongly related to expansion, even though the chert data were quite variable. Samples with total chert contents ranging from about 5 to 22 percent (25% is the maximum allowed by IDOT



**Figure 9** Relationship of chert and dolomite contents to the average percent expansion of their respective test beam sets. Each point is identified by its sample number and source area.

standards for gravel aggregate) had expansions ranging from <0.01 to as much as 0.095 percent (fig. 9). Although the positive slope of the regression line illustrates the general relationship between chert content and expansion, the line should not be used to predict the expansion for a particular chert content.

The wide range of expansion values for the fairly limited range of chert contents in the samples suggests that other factors besides lithology strongly affected expansion values. One factor was probably the wide range in specific

gravity of the chert pebbles in the samples. Most individual test beams probably did not contain a lithologically representative sample of the 60-pound gravel sample used to make them. IDOT specifications restrict the amount of low-specific-gravity (<2.35) chert to less than 3 percent of the total sample; however, there may be a direct relationship between chert with slightly higher specific gravity and expansion. Thus, chert pebbles with specific gravities near but greater than 2.35 may contribute sig-

nificantly to expansion if present in large amounts.

Another likely source of the variability in expansion values was the position of expansive pebbles within the test beams. As noted in the discussion of Phase I testing, expansive pebbles in the central core of the beam probably contributed the most to freeze-thaw expansion. In any one test beam, the presence of pebbles in critical positions may strongly affect the measured expansion value for that beam. It seems likely that increasing the size of the test beams to the maximum prism size (5x5x15 inches) recommended in ASTM C666-77 would help reduce variability in freeze-thaw data by increasing the overall lithologic representativeness of the pebbles in the beams and increasing the number of pebbles in the central cores of the beams.

Dolomite (group 2) content was essentially not related to expansion of the test beam sets (fig. 9). The neutral effect of dolomite in the gravel samples can be further demonstrated by comparing the average expansion of 0.0066 percent for 48 Phase II dolomite (group 2) gravel test beams with that of 0.0060 percent for 83 experimental control test beams containing nonexpansive crushed dolomite (quarry rock).

GLM analysis of freeze-thaw tests of group 3 rock types (table 2) indicated that this rock group contributed only slightly to expansion, and SR analysis identified cherty carbonate and pyritic dolomite as the group members responsible. Because these tests did not identify limestone as a contributor to expansion, it was dropped from further consideration in this study. However, this does not mean that other samples containing more limestone would not show increased expansion; it means only that the small amount of limestone in the study samples did not cause significant expansion.

Neither GLM nor SR analyses of test data on group 4 (table 2) indicated any significant relation of these rocks types to expansion. Therefore, although weakly implicated as a factor in expansion by the analysis of Phase I data, metagraywacke was dropped from further consideration in the study.

GLM analysis of data on group 5 rock types (table 2) indicated a surprisingly strong relationship of this group to expansion. However, this statement must be used cautiously, because many of the rock

types in group 5 were absent or occurred only in trace amounts in several samples. Thus, even though the data for chert are more variable, chert is still considered more closely related to expansion than are the group 5 rock types.

Combined GLM and SR analyses of data on the individual rock types in group 5 indicated that ironstone and silty dolomite were probably most responsible for the expansion observed for the group. Other observations concerning cracks and popouts indicated that ironstone was probably more deleterious than silty dolomite. Weathered carbonate, conglomerate, sandstone-siltstone, and shale may also have been related to expansion, but to a lesser degree. Any weathered rock type found in other samples in amounts greater than in the study samples should be considered suspect, because weathering typically weakens rocks, increases their porosity and permeability, and thus may make them more susceptible to expansion. In general, any gravel samples containing more group 5 rock types than the study samples could experience more expansion.

GLM comparison of IDOT aggregate-quality test data with average expansion values of the Phase I test beams revealed that the relationship of the individual test results with expansion ranged from very strong to unrelated. Even very small amounts (0.2% to 2.9%) of low-specific-gravity (<2.35) chert were strongly related to freeze-thaw expansion. The IDOT tests for total chert, specific gravity of the total sample, and high-specific-gravity (>2.35) chert showed a fairly strong relationship to expansion. These IDOT tests on chert further confirmed our findings that chert has a stronger relationship to expansion than any other rock type.

The percentage of soft and unsound materials in the samples (determined by IDOT) did not correlate well with Phase I expansion data, despite the fact that the soft and unsound category contains ironstone, an expansive rock type. This lack of correlation was probably due to the fact that the proportion of expansive to nonexpansive rock types in the category was unknown. Other IDOT test results indicated little or no relation to freeze-thaw expansion.

To summarize, although there are problems with the statistics, the results of GLM and SR analyses of Phase I, Phase

II, and the IDOT tests indicated that chert, especially low-specific-gravity chert, had a stronger relationship to unacceptable freeze-thaw test expansion than any other rock types. Ironstone was second to chert in its relationship to expansion; however, if ironstone had been as abundant as chert in the samples, it might have caused the test beams to crumble. Silty dolomite probably caused some expansion. Weathered carbonate, cherty dolomite, pyritic dolomite, conglomerate, sandstone-siltstone, shale, and weathered rocks in general were only suspected to contribute to expansion.

## CONCLUSIONS

The following conclusions apply directly only to the high-quality gravel sources used in Illinois portland cement pavement and to the percentages of the rock types found in the study samples. They should not be applied directly to gravel sources used in other states or gravels containing percentage ranges of rock types that differ from those of the study samples.

Statistical analyses of the Phase I, Phase II and IDOT test data indicated that (1) chert, especially low-specific-gravity (<2.35) chert, is related to expansion; (2) ironstone and to a lesser extent, silty dolomite, probably cause expansion; (3) weathered carbonate, cherty carbonate, pyritic dolomite, sandstone-siltstone, conglomerate, shale, and other weathered rocks are only suspected to contribute to freeze-thaw expansion; and (4) dolomite, igneous rocks, and metamorphic rocks are not associated with expansion.

Very small amounts of certain expansive rock types, most notably low-specific-gravity chert ranging from 0.2 to 2.9 weight percent, and ironstone ranging from a trace to 1.5 weight percent, can cause significant expansion.

Variability in expansion among test beams of the same sample and among different samples from the same source is probably related to inconsistent positions and nonrepresentative amounts of expansive pebbles within the cores of the test beams.

Cracks in expansive pebbles (especially chert pebbles) commonly extending into the surrounding matrix cement and through innocuous pebbles were the only abundant deterioration features found during petrographic study of slabs cut from the Phase I test beams.

Expansive pebbles near the surface of a test beam may cause popouts without affecting expansion, but pebbles in critical positions deep within the core of a test beam may contribute strongly to expansion without causing a popout.

## RECOMMENDATIONS

IDOT and the construction aggregate industry should continue to work together to eliminate the occurrence of D-cracking in new highway construction projects. Although freeze-thaw test results are helping to achieve this goal, problems still persist. Additional government- and industry-funded research should be conducted along several lines.

- Test alternate methods of constructing test beams that may eliminate the cause of the highly variable expansion values:
  - increasing the dimensions of the test beams from 3x4x15 inches to the maximum recommended size of 5x5x15 inches (ASTM, 1979, C666-77) or constructing cylinders 15 inches long with 5.6-inch diameters; this would increase the number of pebbles in the beams or cylinders, which should give a more representative population of rock types in the central core of each beam or cylinder.
  - experimenting with different ways of embedding the measuring pins in the beams to minimize the effects of expanding pebbles near their heads.
  - eliminating the +3/4-inch size-fraction from materials used to make test beams. This size-fraction contains far too few particles to provide a lithologically representative sample in each beam; in addition, the larger deleterious particles may have disproportionately large effects on expansion.
- Determine if the effects of internal cracking of the beams caused by the freeze-thaw test can be measured by another method, such as sonic resonance or electrical resistivity, rather than by measuring the length of the test teams.
- Determine the varieties or specific-gravity ranges of chert that are most susceptible to freeze-thaw expansion. A series of freeze-thaw tests could be performed on beams containing identical amounts of chert, separated according to ranges of specific gravity and studied petrographically before and after testing.

- Determine if the number of beams tested per gravel source can be increased to compensate for the variability of the expansion values, thus raising the statistical significance of the data.
- Determine if certain gravel processing techniques, such as the use of the vertical impact crusher, can eliminate significant amounts of expansive rock types.
- Determine if significant variations in rock types exist in different parts of a pit where samples show extremely variable expansion values.

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# The Chicago Stone Industry: A Historical Perspective

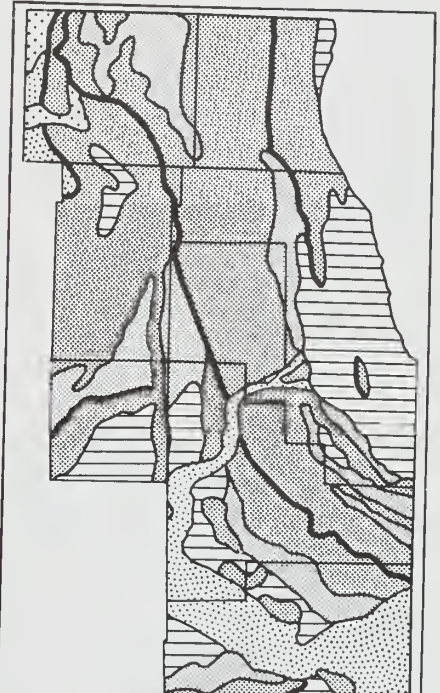
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## ABSTRACT

For more than 150 years, local deposits of Silurian dolomite have provided essential construction materials for the Chicago area. Lime and building stone were the major construction materials from the days of pioneer settlement until the turn of the century. In the early years, many local exposures were quarried, regardless of rock quality, to compensate for a poor and expensive transportation system. As canals, railroads, and roadways were built, however, higher quality rock became more accessible. The opening of the Illinois and Michigan Canal in 1848 allowed shipping of high-quality building stone from Lemont, Lockport, and Joliet to Chicago and other major markets. Quarries in high-purity dolomite closer to Chicago specialized in the production of lime during the latter half of the 19th century.

Around the beginning of the 20th century, economic conditions, changes in construction methods, importation of superior-quality stone, and production and marketing of concrete blocks caused the permanent collapse of the local building-stone industry. The use of cement also eliminated the major market for locally produced lime used as mortar. These events resulted in the abandonment of many quarry sites and the disappearance of several of the larger stone producers in northeastern Illinois.

Early in this century, crushed stone for aggregate became, and still remains, the primary product of the local stone industry. Consolidation of many small independent operators into the major producers that dominate the industry today coincided with this development. Geologic constraints and urban expansion continue to place severe and finite limits on crushed stone reserves in the Chicago Metropolitan area.



## HOLOCENE and WISCONSINAN

Alluvium, sand dunes, and gravel terraces

## WISCONSINAN

lake deposits

## WOODFORDIAN

moraine

front of moranic system

ground moraine

## ALTONIAN

till plain

Figure 1 Glacial geology of northeastern Illinois (after Willman, 1971).

## INTRODUCTION

Since early pioneer settlement, the local stone industry has figured prominently in the development of the Chicago metropolitan area. Here as elsewhere, the availability of stone affected the cost, style, amount, and quality of local construction. Whereas geological features and access to the resource constrain the availability of stone products, changes in economic trends, transportation, architecture, and technology affect the demand for and use of stone. The interaction of these factors is outlined in this brief history of the stone industry in the Chicago area, encompassing Cook, Will, Du Page, Kane, Lake, McHenry, and Kankakee Counties.

## GEOLOGY

The geological features of the Chicago area and their influence on the local stone

industry have been discussed in several papers (Mikulic and Goodwin, 1986; Mikulic and Kluessendorf, 1985; Mikulic et al., 1985; Willman, 1971, 1973). The availability of stone is limited primarily by the distribution, thickness, and composition of unconsolidated Quaternary sediments that cover most of the bedrock surface in northeastern Illinois (fig. 1). These sediments generally are too thick to remove economically in order to exploit the underlying bedrock. Rock is exposed or near the ground surface in only a few areas, primarily where stream or lake erosion has removed most of the sediment (fig. 2). Numerous exposures along the Des Plaines, Kankakee, and Fox River valleys are the result of stream erosion, whereas exposures in eastern Cook County are Silurian reef-controlled bedrock hills uncovered by lake erosion.

The character and distribution of the bedrock have played an important role in the availability of stone materials. Silurian dolomites underlie all of the Chicago region except the western edge (fig. 3). The composition and thickness of these dolomites vary from east to west across the region as the rock dips eastward at 10

to 15 feet (3.0 to 4.5 m) per mile (km) (fig. 4) (Willman, 1971). Silurian rocks are thickest (500 ft; 150 m) along the Lake Michigan shore and in eastern Will and Kankakee Counties. The base of these strata rises towards the west and the entire Silurian thins because of erosional truncation. Along the western edge of the study area, Ordovician rocks are exposed where the Silurian has been eroded completely. Ordovician strata also rise to the west and are thinned by erosion.

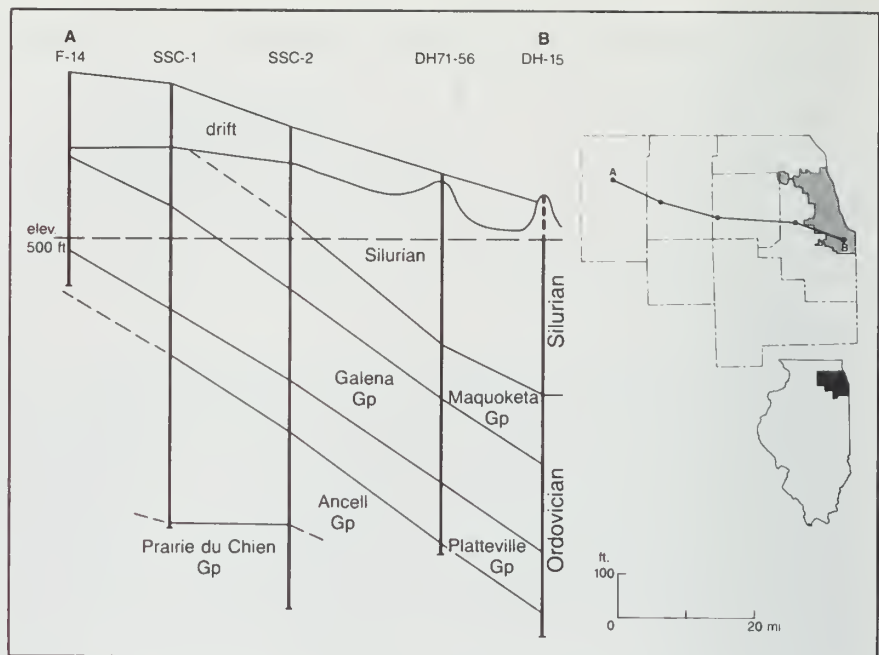
Silurian strata are classified into several stratigraphic units on the basis of general lithologic characteristics (fig. 5). These characteristics determine the use-



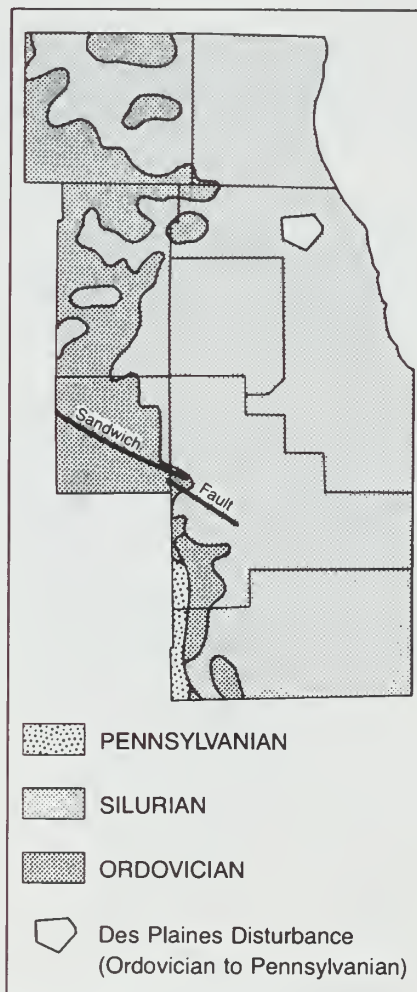
**Figure 2** Outcrop distribution in the Chicago region (modified from Willman, 1971).

fulness of each unit for various stone products. During the 19th century, limited amounts of flagstone, rough building stone, and lime were produced from all parts of the Silurian. The use of these products depended less on rock quality than on proximity of the exposure to the building site. Ultimately, however, improved transportation and higher rock-quality requirements resulted in greater selectivity.

The Racine Dolomite, the uppermost Silurian unit (fig. 5), is as thick as 300 feet



**Figure 4** Diagrammatic cross section of Paleozoic rocks from west to east in study area, showing westward truncation of Silurian strata and general eastward dip of rock units toward the Michigan Basin. Boreholes F-14, SSC-1, SSC-2 are based on core logs obtained by the IGS as part of the siting study for the proposed Superconducting Super Collider. Boreholes DH71-56 and DH-15 are based on core logs from the Metropolitan Sanitary District of Greater Chicago obtained during planning for the Chicago Tunnel and Reservoir Plan. The bedrock surface at DH-15 has been extended up to an elevation of 600 feet to show the nature of nearby Stoney Island, a reef-controlled bedrock hill.



**Figure 3** Bedrock geology of northeastern Illinois (after Willman, 1971).

(100 m) in places; it forms the bedrock surface throughout much of the eastern half of the study area. In general, the Racine varies from argillaceous to pure dolomite. The unit is thickest in the eastern part of Cook County where large, high-purity dolomite reef structures occur, accounting for most of the Silurian exposure in this area (fig. 2). Their proximity to the center of the city and their high purity always have made the Racine reefs the most desirable northeastern Illinois source of many stone products, including lime, riprap, aggregate, foundation stone, and flux.

Underlying the Racine is the Sugar Run Dolomite (fig. 5), which is as thick as 30 feet (9 m). The Sugar Run is exposed primarily along the Des Plaines River valley in the vicinity of Lemont and Joliet (fig. 2), where it forms the bedrock surface in places. This well-bedded,

even-textured rock was the major source of building stone in the late 19th century.

The Joliet Dolomite (fig. 5) underlies the Sugar Run Dolomite. The Romeo Member at the top of this unit consists of up to 35 feet (10.5 m) of high-purity dolomite similar in appearance and character to Racine reef rock. The Romeo Member has been used for lime and flux around the towns of Joliet and Romeoville. The underlying argillaceous and thick-bedded Markgraf Member, as much as 28 feet (8.5 m) thick, also has been used for building stone.

The remaining Joliet Dolomite, the Kankakee, Elwood, and Wilhelmi Formations (fig. 5) that constitute the lower 100 to 200 feet (30 to 60 m) of the Silurian, generally consist of thin-bedded, argillaceous dolomite. This part of the Silurian is well exposed along the Fox River valley in Kane County, in western and southern Will County, and along the Kankakee River in northwestern Kankakee County. Active and abandoned quarries are numerous in these areas, but variable stone quality, decreased thickness of this part of the Silurian, and distance to the Chicago market make these areas less desirable for quarrying than sites farther east.

Shale belonging to the Ordovician Maquoketa Group (fig. 5) ranges from 100 to 250 feet (30 to 85 m) in thickness. It underlies the Silurian strata throughout the study area except in the western part of the Chicago metropolitan area, where it occurs at the bedrock surface (fig. 3). The thin (0 to 30 ft; 0 to 9 m) Fort Atkinson Limestone is the only part of the Maquoketa that has been quarried in this region. Maquoketa strata are generally too thick to remove economically in order to exploit dolomite and limestone of the underlying Ordovician Galena Group (fig. 5), which is exposed only in Kendall County and farther west. As thick as 200 feet (60 m), the Galena is a rarely utilized source of aggregate that lies beneath the entire Chicago region. However, underground mining of the Galena has become economically feasible at one location in Du Page County, and the Galena is potentially a very large, high-quality source of aggregate and subsequent underground storage space.

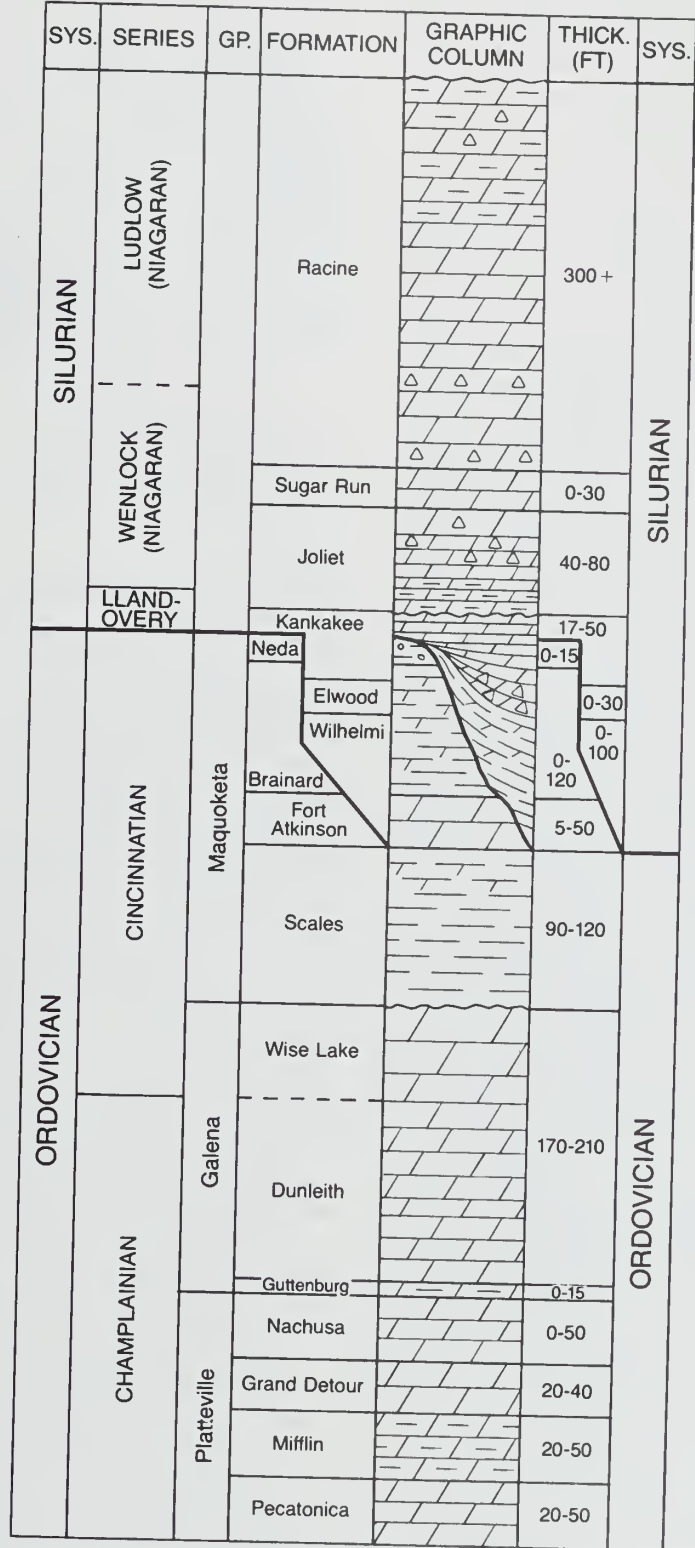


Figure 5 Stratigraphic column showing Silurian and Ordovician rocks in northeastern Illinois (after Mikulic et al., 1985).

## GROWTH OF THE BUILDING STONE INDUSTRY

When pioneer settlement of the region began in the early 1800s, construction materials such as lumber, brick, building and foundation stone, and lime for mortar had to be obtained locally because of the severely limited and expensive transportation system.

Although the nearest exposures were located several miles from the center of the original Chicago settlement during the 1830s to the 1850s, quarries had been opened in many of the low exposures of Racine reef rock on the Lake Chicago Plain (figs. 1, 2). The earliest documented stone production occurred in 1833 when a quarry was opened in what is now the Bridgeport neighborhood of Chicago. Stone from this quarry was towed in barges 4 miles along the South Branch of the Chicago River to Lake Michigan and was used in the original construction of the Chicago Harbor pier (Andreas, 1884).

Stone was readily accessible near Joliet and other settlements along rivers and streams and was used in many early local building projects. Shepard (1838) mentioned that Chicago quarries were producing "a valuable flagging-stone," whereas quarries at Joliet were extracting "very valuable building material." These quarries were small, labor-intensive, intermittent operations that generally were worked only to supply construction materials for a specific project.

The rapid development of Chicago in the middle and late 1800s created a major market for stone products, and a continually improving transportation system permitted the economical and reliable shipment of stone materials to this market. In 1847, stone from Joliet was hauled by wagon to Chicago for construction of Scammon School (Goodspeed Publishing Company, 1891). The Illinois and Michigan Canal, which opened in 1848, was the most important advancement in transportation for the stone industry because it provided a direct and inexpensive means of shipping building stone from Joliet, Lockport, and Lemont to the Chicago market.

The closest source to Chicago of well-bedded, high-quality Sugar Run Dolomite (then known as Athens, Lemont, or Joliet limestone) was discovered during construction of the canal itself at Lemont (then called Athens). During the late 1800s this stone became a popular build-

## STONE DRESSERS.

## TO STONE DRESSERS AND BUILDERS.

# The Illinois Stone Company, OF LEMONT, ILL., ORGANIZED FEBRUARY 24, 1855. Chicago Office, - Corner Wells and Taylor Streets.

President.....S. F. GALE.  
Treasurer.....H. G. LOOMIS.  
Secretary and Superintendent...J.W. MCGENNISS.  
General Agent.....A. S. SHERMAN.

**TRUSTEES.**  
S. F. GALE, W. S. GURNEE,  
H. G. LOOMIS, A. S. SHERMAN,  
O. SHERMAN.

THE ILLINOIS STONE COMPANY is now prepared to contract for the delivery of Stone from its celebrated quarries at "Athens."

For beauty and uniformity of color, this stone stands unsurpassed by any in the United States. The Company's facilities are ample for furnishing any desired quantity of Dimension, Block, or Rubble Stone.

Orders through the Post office or left at the office of H. G. Loomis, Treasurer, 8 Clark street, up stairs, addressed to the undersigned, will receive prompt attention.

J. W. MCGENNISS, Superintendent.

Figure 6 Advertisement for the Illinois Stone Company from the *Northern Counties Gazeteer and Directory* for 1855-56, Robert Fergus, Book and Job Printer, 1855.

ing material used in many major residences, businesses, government buildings, and churches, and extensive quarries were opened at Lemont in the early 1850s by the Singer and Talcott and the Illinois Stone companies (fig. 6).

During the late 1800s, Lemont-Joliet building stone was the major product of the stone industry in northeastern Illinois. Singer and Talcott, the largest building stone producer throughout much of this period, employed 300 workers at its Lemont quarries and 150 at its Chicago yard (Land, 1883).

Increasing amounts of Lemont-Joliet stone were shipped outside northeastern Illinois as an extensive railroad network developed throughout the Midwest. In the 1860s, W. A. Steel's quarry, located between Lockport and Joliet, supplied stone for government construction projects in Illinois, Wisconsin, Missouri, Iowa, and Indiana (Steel, 1871). In 1870, Joliet quarries produced 1,125,000 cubic feet (12,000 railroad carloads) of stone and employed 700 men (Steel, 1871). By 1886, building-stone quarries were located at many communities; those listed by Polk (1886) include: Aurora (2), Batavia (6), Bonfield (1), Joliet (15), Kankakee (1), Lemont (5), Naperville (2), and Sag Bridge (1). Undoubtedly, more quarries existed.

Major consolidation of the industry took place in 1889 when Singer and Talcott combined with five other building-stone companies, forming the Western Stone Company (fig. 7) (Goodspeed Publishing Company, 1891). This new company employed about 1500 men and operated 35 boats, tugs, and barges at its Joliet, Lemont, and Chicago yards (Goodspeed Publishing Company, 1891).

## DECLINE OF THE BUILDING STONE INDUSTRY

Despite continued growth of the building-stone industry through much of the late 1800s, several problems that arose led to its total collapse by World War I. The first problem concerned a change in the way large buildings were constructed. Massive stone blocks had been used as a foundation to support most of the weight of a building, but during the 1870s steel internal supports came into use, eliminating the need for stone foundations. As this type of construction became more widespread, stone was relegated to an ornamental function, primarily as veneer; such use placed more emphasis on the appearance of the stone facing and less on the quality of the rock.

By the 1890s building stone was being imported from outside the area, constituting another problem for the Lemont-Joliet stone industry. Improved



**Figure 7** Western Stone Company operations at Lemont, 1907; photo by E. F. Burchard, U.S. Geological Survey Library.

transportation methods permitted a variety of stone types such as Lake Superior sandstone from Michigan and Wisconsin, granite from Wisconsin, and limestone from Indiana to be sold competitively with local building stone in the Chicago market. Labor and economic problems during this period also had a negative impact on the industry.

Another critical factor in the decline of the Lemont-Joliet stone industry was the increasing use of concrete in construction. Concrete now could be used to produce a more uniform, durable, and ultimately cheaper building material than natural stone. Sidewalks, previously constructed of large slabs of cut or rough stones, had provided a major market for stone in Chicago; now, stone slabs were rapidly being replaced by uniform slabs of concrete. Rough and cut foundation stone was replaced by cheaper, more durable, and uniform concrete blocks. Old habits died hard, however, and many early concrete blocks were molded to look like dressed stone. The only building-stone market remaining was ornamental stone, for which most of the local Silurian rock was not well suited.

By World War I, the once prosperous Lemont-Joliet building-stone industry had almost disappeared. Since that time some cut stone has been produced from Silurian rocks in northeastern Illinois, but only a few flagstone quarries remain in the once extensive Lemont-Joliet quarry district. None of the major building-stone concerns, even those that were the largest companies in the Chicago stone industry for several decades, survived this collapse.

The Chicago area quarries did produce some building stone. However, local rock seldom could be excavated in large slabs, did not have an acceptable appearance, and was more difficult to fashion into blocks than the stone at the Lemont-Joliet quarries; therefore, it could never compete with the latter for the building-stone market. Some of the Racine Dolomite from the Rice and Artesian quarries near Grand and Campbell streets in Chicago contained asphalt-filled pores and was marketed as "blackstone." This stone was used in such buildings as the Second Presbyterian Church and the castellate walls and towers of the Libby Prison-Coliseum in Chicago.

### LIME PRODUCTION

During the rise and fall of the Lemont-Joliet building-stone industry, lime-burning continued to be a major part of the Chicago area stone industry. Although Cook County operations, which quarried high-purity Racine Dolomite, were the main source of lime, kilns were also operated at Kankakee, Batavia, Joliet, and elsewhere. Several prominent companies, including Stearns Lime and Stone and Chicago Union Limeworks, were formed in and around Chicago to take advantage of this high-quality rock close to the city market (Mikulic and Kluessendorf, 1985). The demand for lime apparently far exceeded the capacity of local kilns, and from the 1860s through the 1890s, a number of Wisconsin firms maintained offices in Chicago where they sold lime that was similar if not identical to that produced in the local quarries.

However, the lime industry eventually suffered a fate similar to that of the building-stone industry, for some of the same reasons. Like the building-stone industry, the manufacture of lime was affected adversely by an increase in the use of cement.

Although lime is still produced at the Thornton and McCook quarries, most quarries abandoned its production early in this century.

### CRUSHED STONE PRODUCTION

With the development of a more extensive and enduring transportation system, crushed stone, utilized mostly for macadamizing roads and for railroad ballast, became an important product of the mid- to late 1800s. Large-scale macadamizing, beginning in the 1860s, led to the formation of the Dolese and Shepard Company in 1868 by John Dolese and Jason Shepard (Goodspeed Publishing Company, 1891). Dolese and Shepard operated a number of quarries in Cook County; in 1870, the company employed 600 men at its Hawthorne and Cheltenham quarries and an additional 200 men for street construction.

By the late 1800s, many of the older quarries that had initially been located in rural areas were surrounded by residential neighborhoods, and there was no room for lateral expansion. The only recourse was to quarry to greater depth; however, strata beneath the Racine Dolomite in Cook County generally are unsuitable for high-quality lime. Crushed stone for street paving and concrete aggregate therefore became the main product of many older quarries. The advent of crushed-stone production marked the beginning of the modern Chicago stone industry.

As city quarry sites were exhausted, the extensive Chicago railroad network provided cheap transportation of crushed stone into the city center from outlying regions, primarily the Thornton and McCook areas. With the development of these two outlying districts and exhaustion of high-purity reserves, most of the Chicago quarries closed down; by 1930, only the Stearns quarry at Bridgeport remained (Mikulic and Kluessendorf, 1985). Formation of the Material Service Corporation by the Crown family in 1917 and expansion of the Consumer Company in 1921 led to major consolidation of the crushed-stone industry. The Dolese

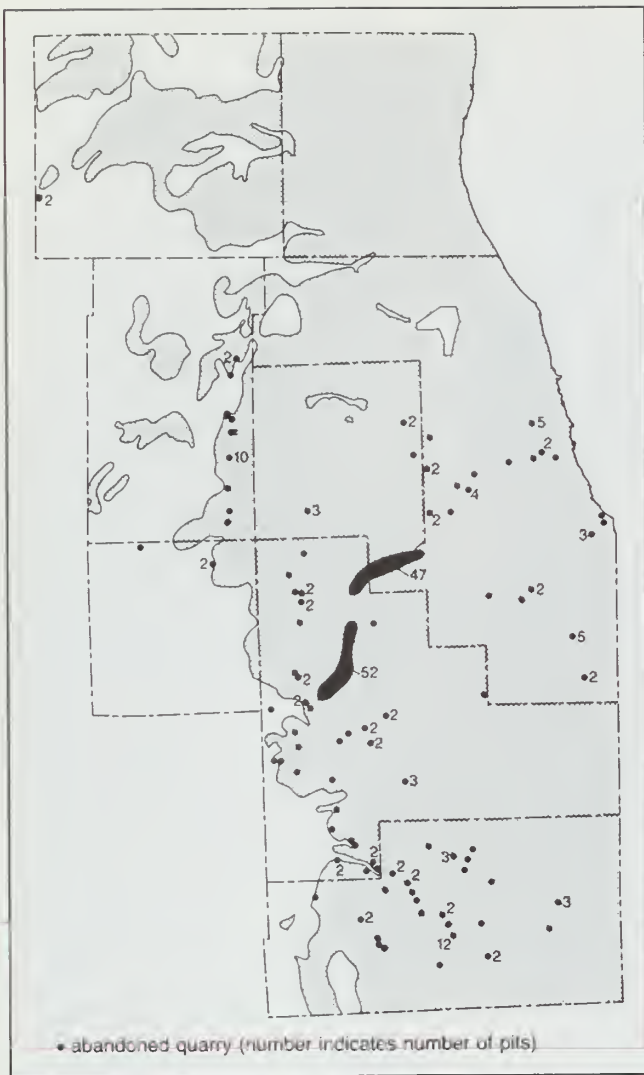


Figure 8 Abandoned stone quarries in northeastern Illinois.

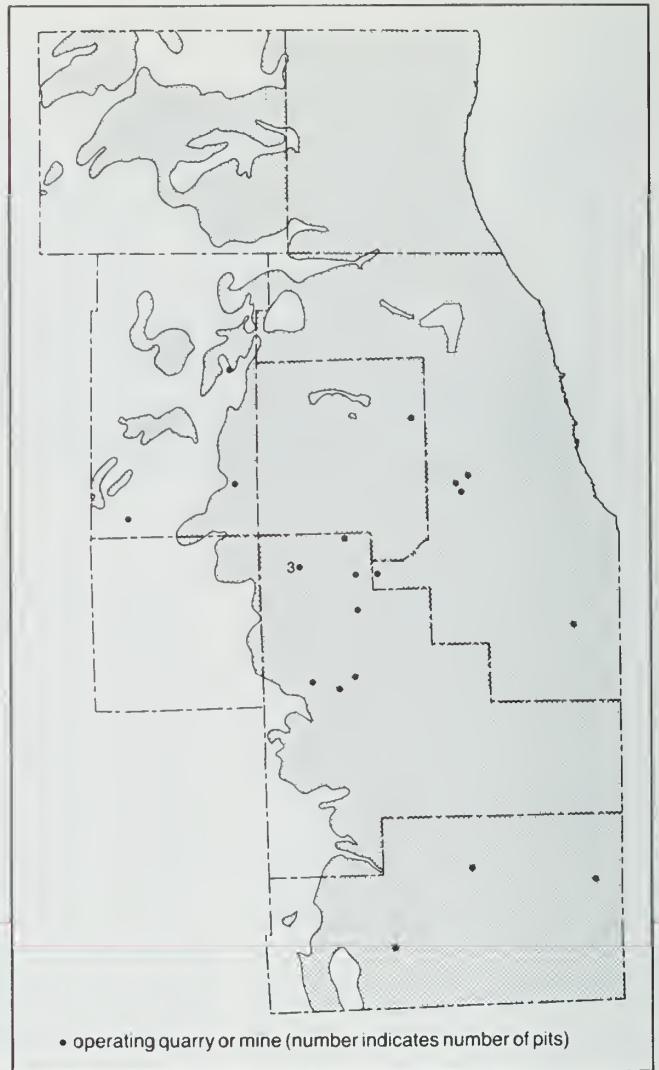


Figure 9 Active stone quarries in northeastern Illinois.

and Shepard Company, long the largest Chicago area stone concern, ended nearly 100 years of operation when it was acquired by the Consumer Company (now Vulcan Materials) in 1967.

With industry consolidation, urban expansion, and increasing demand for higher-quality materials, the number of quarries in northeastern Illinois has decreased dramatically since the turn of the century. More than 250 abandoned stone pits are known in northeastern Illinois (fig. 8), but only about 20 sites are quarried today (fig. 9).

#### STONE INDUSTRY PROSPECTS

Most of the once-rural quarries are now surrounded by urban development; little room exists for expansion, and near-surface reserves are limited (Mikulic and

Goodwin, 1986). The northeastern Illinois stone industry is faced with the problem of securing new long-term sources of local stone to meet the increasing demand of the growing metropolitan area. This growth has created and maintained an enormous market for stone products, but at the same time has eliminated many local sources of such materials from exploitation. Urban expansion has limited the number of potential quarry sites in areas where the Silurian is thick and consists of high-quality Racine Dolomite. Localities in the western part of the metropolitan area are available for quarrying, but the thin section of Silurian rock in this region and the poorer quality of stone produced from some lower Silurian units suggest that

these localities would not be a promising source for the future.

When the few remaining quarries eventually close down, stone for Chicago area needs will have to be imported from more distant areas at increasingly higher transportation costs. An alternative solution may be underground mining of the Ordovician Galena and Platteville Groups at existing quarry sites, as is now done at Elmhurst-Chicago Company's Elmhurst plant (fig. 9). Quarries such as those at McCook and Thornton would be particularly suitable for this approach because of their large size.

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# An Overview of the Geology of Clays, Shales, and Slates Utilized in Ceramic and Structural Clay Products in Georgia

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## ABSTRACT

Since the turn of the century, a wide variety of argillaceous materials have been used in the manufacture of ceramic and structural clay products in Georgia. In the Valley and Ridge and the Cumberland Plateau Provinces in northwest Georgia, widespread deposits of clay and shale consist mostly of weathered Paleozoic shales but also include Tertiary to Recent alluvial, colluvial, and residual clays, and Paleozoic slates and phyllites. Present production comes principally from Cambrian, Silurian, and Mississippian illitic shales that are used for common brick and quarry tile. Ordovician slates are used for lightweight aggregate. In the past, other products such as sewer pipe, flue tile, fire brick, roofing granules, and portland cement were produced locally in northwest Georgia. Much of the shale mined in this area is shipped to plants in other parts of the state and in adjacent states.

In the Piedmont and Blue Ridge Provinces, common brick, flue tile, and portland cement are manufactured from Quaternary alluvial clays and/or saprolitic residuum developed on the Precambrian to Paleozoic bedrock (weathered slate, phyllite, phyllonite, and fine-grained schist). In the Coastal Plain Province, common brick is produced from Quaternary alluvial and colluvial clays along the major river flood plain systems adjacent to metropolitan areas along the Fall Line. These alluvial deposits are similar to but more widespread than those in the other provinces. They all contain mixtures of kaolinite, illite, smectite, chlorite, and vermiculite and are commonly blended with clay and/or shale from the other provinces. Tertiary kaolinitic clays and smectitic clays are locally used in making portland cement. Lightweight aggregate has been produced from Paleocene smectite- and chlorite-bearing clay in southwest Georgia, but this operation is not currently active. In the past, other products such as sewer pipe, structural tile, and roofing tile were also produced in the Coastal Plain Province.

## INTRODUCTION

Many geologists are aware of the fact that Georgia is a major producer of kaolin and fuller's earth; however, most do not realize that a substantial amount of common clay and shale is also produced in the state. This article summarizes information on the geology, commercial utilization, and historical development of these common clay and shale deposits.

Table 1, which shows the total clay production in Georgia for 1985, is based on the most recent data available from the U.S. Bureau of Mines (Ampian, 1987; White and O'Connor, 1987). This table illustrates the relative importance of the

three major clay commodities (kaolin, fuller's earth, and common clay and

shale) in Georgia and their ranking in value and tonnage with respect to the total national production. Kaolin is clearly the predominant clay commodity mined in the state. However, common clay and shale, as well as fuller's earth, also make a substantial contribution to the total clay output. Nationwide, Georgia ranked fifth in production (tonnage) but eighth in value of common clay and shale among the 43 clay and shale producing states. (Texas and North Carolina ranked first and second in common clay production in 1985.)

## GEOLOGY

Georgia is divided into five physiographic provinces (fig. 1) that can be grouped into three major geologic areas (Georgia Geological Survey, 1976). From north to south these are: the Cumberland Plateau and Valley and Ridge Provinces in northwest Georgia; the Blue Ridge and Piedmont Provinces in north central Georgia; and the Coastal Plain Province in south Georgia. The predominant features of each of these areas are summarized in table 2.

### Northwest Georgia

The Cumberland Plateau and the Valley and Ridge Provinces of northwest Georgia consist predominantly of folded Paleozoic sedimentary rocks (mostly car-

Table 1 Clay production in Georgia for 1985\*.

Type	Tons (x 10 <sup>6</sup> )	Value (\$) (x 10 <sup>6</sup> )	National rank	
			Tonnage	Value
Kaolin	6.3	535.0	1	1
Fuller's earth	0.6	34.6	1	1
Common	1.7	5.5	5	8
Total	8.6	575.1	1	1

\*Data from Ampian, 1987.



Figure 1 Physiographic provinces of Georgia.

bonates, sandstones, and shales) ranging from Cambrian to Pennsylvanian in age. They are essentially unmetamorphosed except along the eastern and southern border of the area where slaty cleavage is well developed. In this region the Paleozoic strata have been overthrust by the igneous and metamorphic rocks of the Blue Ridge Province along the Great Smoky-Cartersville Fault.

Clay and shale deposits have been mined from a number of stratigraphic units in this area. For the most part these are illitic lithologies that are best utilized in the manufacture of structural clay products such as brick, tile, and sewer pipe. The most important of these lithologies are the shales (and residual clays weathered from them) of the Conasauga Group (Cambrian), the Red Mountain Formation (Silurian), and the Floyd Shale (Mississippian)—all used in making brick and/or quarry tile. In addition, the Rockmart Slate (Ordovician) is expanded for lightweight aggregate.

In the past, various products were produced from other local deposits. Quaternary to Recent alluvial clays mined from many of the local flood plains

were used in making brick, tile, sewer pipe, and flue liner. In several areas, Tertiary siliceous and kaolinitic residual clays derived from Cambrian-Ordovician Knox Group carbonates were used in making fire brick. Residual clay of the weathered Rockmart Slate (Ordovician) has been used in the production of portland cement, and slates of the Conasauga Group (Cambrian) at one time were crushed for roofing granules.

### Blue Ridge and Piedmont

The Blue Ridge and Piedmont Provinces are composed of igneous and metamorphic rocks (mostly gneisses, schists, and granites) that range in age from Precambrian to Paleozoic. Much of the bedrock in this area is blanketed with a thick residual clay mantle (saprolite), and Quaternary to Recent alluvium is commonly thick along the major streams and rivers in this area. Common brick, flue tile, and portland cement are locally manufactured from these alluvial and saprolitic clays, which can be improved by blending with clays from northwest Georgia. In most cases, the preferred saprolitic clays are those derived from relatively fine-grained argillaceous bed-

Table 2 Structural clay deposits of Georgia.

Province	Age	Type	Clay Minerals	Use
Cumberland Plateau and Valley and Ridge	Paleozoic	Shale	Illite	Brick, tile, quarry tile and pipe (cement)
	Paleozoic	Slate	Illite, chlorite	Lightweight, aggregate (roofing granules)
	(Tertiary)*	(Clay)	(Kaolinite)	(Fire brick)
	(Quaternary to Recent)	(Clay)	(Illite, chlorite, smectite, kaolinite and vermiculite)	(Brick, tile, pipe, and flue liner)
Blue Ridge and Piedmont	Paleozoic and Precambrian?	Weathered phyllite and phyllonite residuum	Illite, chlorite, and kaolinite	Brick, cement, and flue liner
	Quaternary to Recent	Clay	Illite, chlorite, smectite, kaolinite and vermiculite	Brick, flue tile, and cement
Coastal Plain	Quaternary and Recent	Alluvial clay	Illite, chlorite, smectite, kaolinite and vermiculite	Brick (pipe)
	Tertiary	Clay	Smectite and chlorite	Cement and lightweight aggregate
	Tertiary	Clay	Kaolinite	Cement

\*Items in parentheses indicate deposits no longer being worked and types of clay no longer used.

rock such as slate, phyllite, phyllonite, and fine-grained mica schist. However, in some places brick was made from residual and colluvial clays derived from gneissic bedrock, and saprolite clay developed on a small gabbro body was blended with other clays to make sewer pipe.

### Coastal Plain

The Coastal Plain occupies the southern half of the state and is composed of poorly consolidated sediments—mostly clays, sands, and marls ranging in age from Cretaceous to Recent. The northern boundary of the Coastal Plain is called the Fall Line; this is the surface expression of the unconformity between the igneous and metamorphic rocks of the Piedmont and Blue Ridge and the overlying Coastal Plain sediments.

Quaternary alluvial deposits are well developed along the major stream and river flood plains across the Coastal Plain. This alluvium is extensively mined for commercial brick clay adjacent to major metropolitan centers near the Fall Line. These clays are composed of mixtures of kaolinite, illite, smectite, chlorite, and vermiculite with varying amounts of

detrital mica, quartz silt, and organic debris. They are commonly blended with clay and/or shale from other areas.

Relatively little common clay has been mined further to the south. Lightweight aggregate was produced from carbonaceous clays of the Tusahoma Formation (Paleocene) for a short period of time in Clay County. Kaolinitic clays (Eocene undifferentiated) and fuller's earth of the Twiggs Clay (Eocene) in Houston County are used in making portland cement; however, most of the clay mined south of the Fall Line is high-grade kaolin and fuller's earth, which are used in more spe-

cialized applications such as coatings, fillers, absorbents, and catalysts.

### HISTORICAL DEVELOPMENT

Common clay has been mined in Georgia for the manufacture of building brick since at least the late 1700s (Smith, 1931). However, very little comprehensive information is available on clay mining and production of structural clay products that occurred before early in the 20th century.

Shortly after the turn of the century, common clay and shale were mined at numerous locations that were, in general, evenly distributed among the three areas

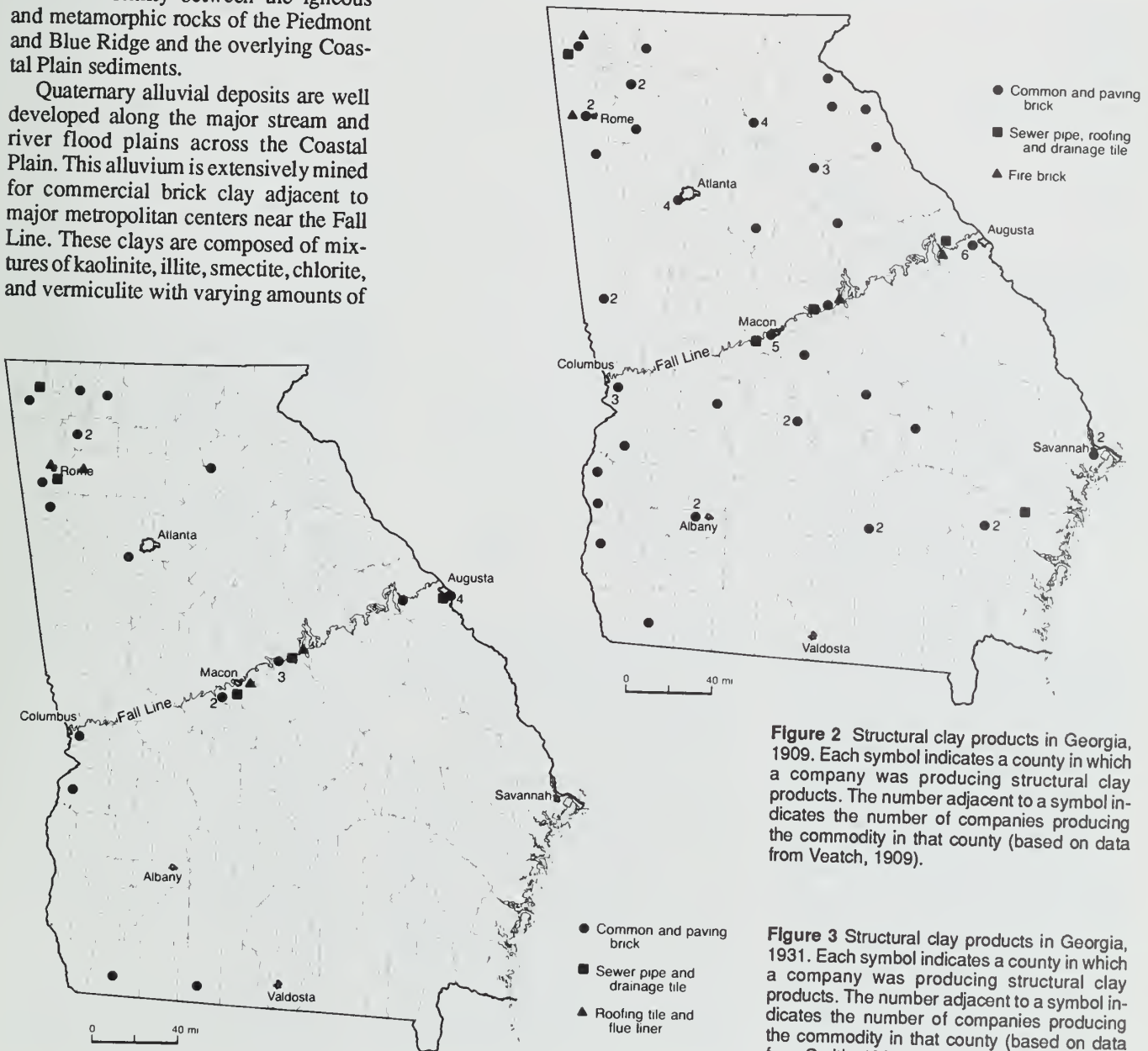
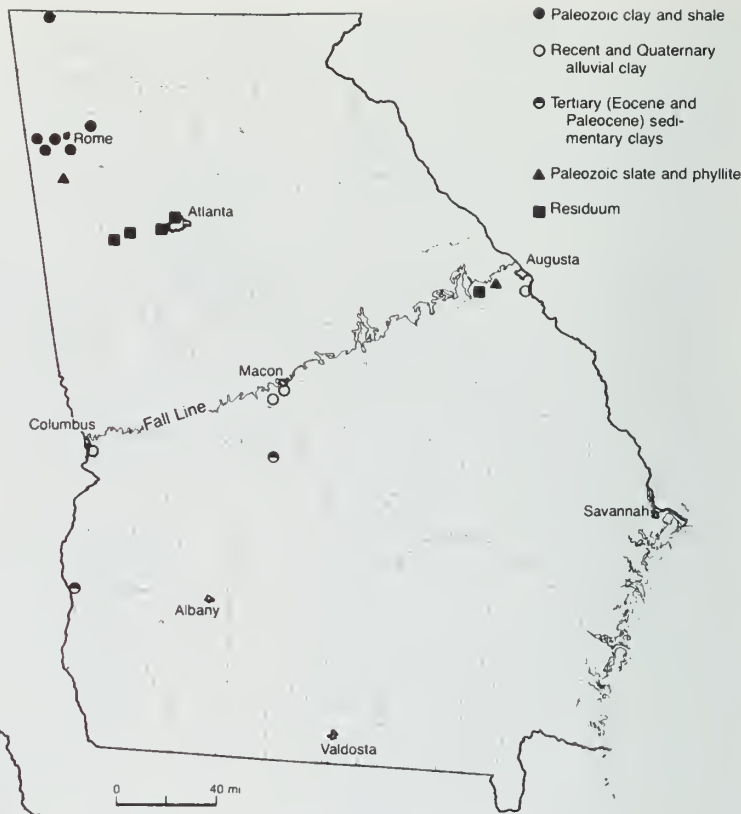
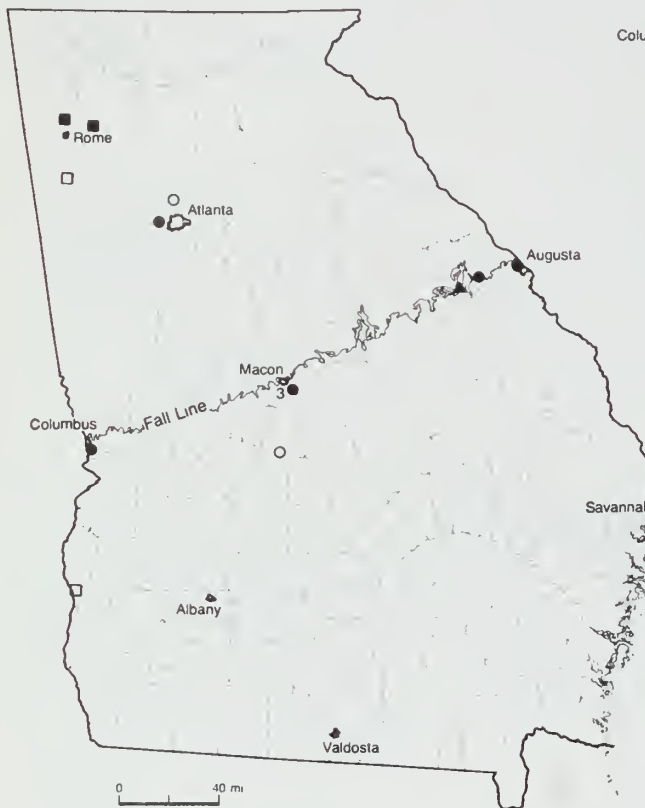


Figure 2 Structural clay products in Georgia, 1909. Each symbol indicates a county in which a company was producing structural clay products. The number adjacent to a symbol indicates the number of companies producing the commodity in that county (based on data from Veatch, 1909).

Figure 3 Structural clay products in Georgia, 1931. Each symbol indicates a county in which a company was producing structural clay products. The number adjacent to a symbol indicates the number of companies producing the commodity in that county (based on data from Smith, 1931).

**Figure 4** Current mining of structural clay and related materials in Georgia. Each symbol indicates a county in which a company (or companies) is mining the commodity in that county (based on data in Steele and O'Connor, 1987)

**Figure 5** Structural clay products currently produced in Georgia. Each symbol indicates a county in which a company (or companies) is producing structural clay products; the number adjacent to a symbol indicates how many companies are producing the commodity in that county.



somewhat more numerous and widespread than the manufacturing plants; the explanation for this is that the most desirable raw materials are not always available close to established plants and that most plants use a blend of several grades of clay in manufacturing their products.

The data summarized in figures 2 to 5 suggest the combined effects of several factors over time. Near the turn of the century, clay and shale were mined from numerous small pits used mostly to supply relatively small plants that generally served small, local markets. Because demand was relatively small, local deposits could easily be found near these plants. Over time, many of these deposits were exhausted. In addition, urban growth has encroached on many potential reserves, and the increasing popularity of wood, concrete, and plastic has reduced the overall demand for structural clay products. Increased energy costs have forced many older, inefficient plants to close or consolidate with other operations.

The net result of these factors is that a relatively small number of companies is now producing a limited range of structural clay products from large plants near

of the state. Figure 2 shows the location (by county) of clay and shale plants producing a variety of structural clay products in the early part of the century (from data in Veatch, 1909). Nearly all of these manufacturing facilities used clay and shale mined from local deposits, and most of the plants were producing brick. However, at a few locations fire brick was made from residual and sedimentary kaolinitic clays in northwest Georgia and along the Fall Line. Other specialized products, mainly sewer pipe, drain tile, and roofing tile, were produced at widely scattered locations across the state.

By the 1930s, common clay and shale production was largely concentrated along the Fall Line and in northwest

Georgia (fig. 3). According to data in Smith (1931), most of these deposits were used for brick manufacture, although sewer pipe, drain tile, roofing tile, and flue liner also were made locally.

Data on current mining of structural clay and related materials and the manufacture of structural clay products are summarized in figures 4 and 5 (from data in Steele and O'Connor, 1987). These figures illustrate the continued trend of consolidation near the larger metropolitan centers of Rome (in northwest Georgia), Atlanta (in the Piedmont), and near Columbus, Macon, and Augusta (along the Fall Line). A comparison of the two figures, however, indicates that the clay and shale mines are

their market areas. Because of the limited availability of large volumes of clay and shale raw materials, some of the clay is transported long distances from the mine to the plant.

#### **FUTURE DEVELOPMENTS**

Future developments are difficult to predict; however, we anticipate continued stability accompanied by a modest growth in the ceramic and structural clay industry in Georgia. Raw materials are adequate and the economy of the state is strong. The construction industry is healthy. Although building-brick faces strong competition from alternative construction materials, Georgia's brick industry

has a broad market base throughout the southeastern United States. In addition, the ceramic tile market is growing and will probably offset any losses the brick industry may experience in the future. The ceramic tile industry continues to express a strong interest in Georgia's structural clays, and numerous foreign and domestic companies have studied the area for the possible establishment of new ceramic tile plants in the state.

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# Industrial Minerals and the CUSMAP Program

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## ABSTRACT

*Explorationists, geologists, and geological surveys have for many years been studying mineral deposits and attempting to extrapolate their findings into mineral resource appraisals of large regions. One such attempt has been the Conterminous United States Mineral Assessment Program (CUSMAP) of the U.S. Geological Survey. The Missouri Department of Natural Resources, Missouri Geological Survey Program, has been involved in four CUSMAP appraisals and is currently starting a fifth. The work began with the Rolla 1° x 2° Quadrangle in 1975, continued through the Springfield, Harrison, and Joplin 1° Quadrangles, and now includes the Paducah 1° x 2° Quadrangle. The original intent of these studies, at least that of the U.S. Geological Survey, was to evaluate metallic mineral resource potentials. Over time, because of changing interests of the mineral industry and different geologic conditions in various quadrangles, these studies have evolved into more broad-based mineral resource appraisals. In the Rolla Quadrangle, assessment of a wide range of minerals was an essential part of the study from the beginning. Industrial minerals are the only mineral resources that are currently exploited and that have any realistic economic potential for further development in the Springfield Quadrangle. In the Joplin Quadrangle the appraisal will include mineral fuel potential. An opportunity exists for further development and refinement of these appraisals in the upcoming study of the Paducah Quadrangle for the states of Kentucky, Illinois, Indiana, and Missouri.*

## INTRODUCTION

The Conterminous United States Mineral Assessment Program, commonly known as CUSMAP, is the result of several long-term U.S. Geological Survey (USGS) efforts to prepare regional mineral resource appraisals. The general objective of this program is to assess the mineral resource potential of the lower 48 states, using geology, geochemistry, geophysics, remote sensing, statistics, and computer science. The USGS has organized the program and directs the work, but seeks the cooperation and contributions of state geological surveys and individuals knowledgeable in the various disciplines and areas of study.

Criteria considered important in the selection of CUSMAP quadrangles are known or suspected mineral resource potential, percentage of federal land within the quadrangle, land changes that may affect future mineral resource development, and need for improved

geologic information about the area (Ovenshine, 1982, personal communication to W. B. Howe). A characteristic of CUSMAP appraisals, however, is their temporary significance because of changing economics and technological advancements in the mineral industry; hence, they are not meant to be final judgments on areal or regional mineral potentials.

## APPRAISAL METHODOLOGY

The methodology used in these appraisals was developed at a resource appraisal workshop convened by the USGS in 1979 (Shawe, 1981). As part of that workshop, scientists from government, industry, and academia designed a methodology for appraising the resource potential of several 2° quadrangles, one of which was representative of the mid-continent geologic province. This evaluation procedure comprises six basic steps (Pratt, 1981):

1. Compilation of geologic, stratigraphic, geochemical, and geophysical maps of a quadrangle to identify known and inferred geologic environments and structures present.
2. Determination of types of mineral deposits that could be expected to occur in the quadrangle on the basis of (a) known mineral deposits and occurrences that exist in the quadrangle and (b) known worldwide associations of certain mineral deposit types with geologic environments and structures that exist in the quadrangle.
3. Development of descriptive models for each mineral deposit type.
4. Development of "recognition criteria" (based on the descriptive model) for the occurrence or nonoccurrence of each type of mineral deposit.
5. Careful study of the available data for the existence of recognition criteria.
6. Evaluation of the distribution and importance of various recognition criteria to appraise the probability of occurrence and generalized location for each mineral deposit type throughout the quadrangle.

Recognition criteria, essential to this methodology, are the geologic characteristics that affect favorability for occurrence of mineral deposits in an area or region. They may be diagnostic, permissive, or negative.

*Diagnostic criteria* are characteristics present in all, or most, known deposits that are commonly considered to be necessary for the occurrence of mineral deposits. The known absence of such criteria may either severely limit or definitively rule out the possibility of deposits. Although diagnostic criteria indicate that deposits may be present, they do not guarantee it.

*Permissive criteria* are so commonly associated with deposits that they are considered to favor their presence. Their existence greatly enhances the possibility of mineral deposits, especially if diagnos-

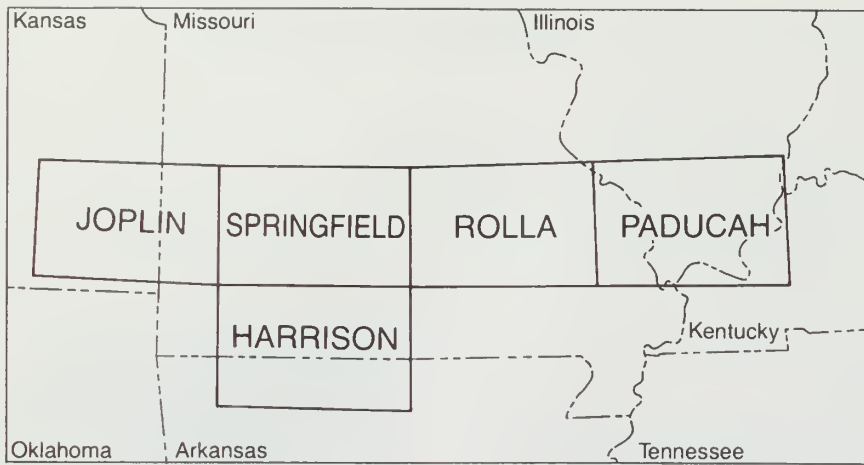


Figure 1 Index map.

tic criteria are present. Absence of permissive criteria, however, does not necessarily reduce the possibility of mineral deposits so long as diagnostic criteria occur.

*Negative criteria* generally can be equated with the known absence of diagnostic criteria.

### RECENT CUSMAP STUDIES

CUSMAP studies have traditionally concentrated on the identification of areas or regions favorable for metallic mineral deposits, but during several recent studies in Missouri this approach began to change.

The study of the Rolla Quadrangle by the Missouri Geological Survey and the USGS that began in 1977 was completed in May, 1981; this study was followed by a study of the Springfield Quadrangle, completed in 1985, and by studies of the Harrison and Joplin Quadrangles, still in progress. In October, 1986, preliminary planning began on assessment of the Paducah Quadrangle. Locations of these quadrangles are shown in figure 1.

The Rolla Quadrangle, in southeast Missouri, includes all the southeast Missouri lead district, much of the Washington County barite district, and several important iron mining areas. The quadrangle contains the largest known lead reserves in the world; in 1986 the region accounted for approximately 96 percent of the country's domestic lead production (Wharton, 1987).

Industrial minerals are also an important economic asset in this quadrangle. The mineral operation producing the largest tonnage of ore annually is a

crushed stone quarry, and the mineral operation with the largest annual value of output is a lime plant.

### RESOURCE EVALUATION

#### Development of descriptive model

The first step in evaluating a resource commodity is to develop a descriptive model. An example is the following model, slightly modified from Pratt and Wharton (1981); this example, developed by Pratt and Wharton from available descriptive data, represents the known facts or features of barite deposits in the Rolla Quadrangle.

#### *The typical ore body: barite*

- *Size: a few acres to more than 1500 acres; thickness up to 50 feet, averaging about 10 feet.*
- *Mode of occurrence: barite fragments, mostly 1/2 inch to 6 inches long contained in clayey, cherty residuum.*
- *Tonnage:  $n \times 10^6$  tons of ore.*
- *Minimum grade: 75 to 100 lbs barite per  $yd^3$  residuum represents the economic cutoff as of September 1980.*
- *Mineral content: barite, commonly accompanied by galena, marcasite, and sphalerite. Marcasite may be completely altered to limonite.*
- *Depth: ground surface to top of bedrock.*

#### *Lithology of host rocks*

- *Principal ore host rock is residuum from the upper half of the Potosi Dolomite or the lower two-thirds of the Eminence Dolomite. Minor production has also been reported in residuum from the Gasconade, Roubidoux, and Jefferson City formations, and occasionally from*

*the Bonnetterre, Davis, and Derby-Doerun formations.*

- *Some decomposed bedrock zones at the base of residual ore occurrences are barite rich and soft enough to mine with conventional equipment.*

#### *Structural controls*

- *Principal controls are highly permeable zones related to fractures and voids in bedrock and algal reef structures.*
- *The main producing areas are bounded by a major fault-block system. Faulting in the block has probably increased host-rock permeability for penetration of ore-forming fluids.*

#### *Geochemical indicators*

- *Barite occurrences in insoluble residues of carbonate rocks from drill holes; barite in panned concentrates of stream sediments.*

#### *Geophysical indicators*

- *Positive gravity anomalies in very detailed surveys.*

#### *Preparation of the resource appraisal*

The descriptive model was used with the following criteria to prepare the resource appraisal:

#### *Diagnostic criteria*

1. Outcrop areas of Potosi Dolomite and lower part of Eminence Dolomite; outcrop areas of Bonnetterre, Davis, and Lower Ordovician formations are less important.
2. Proximity to major and minor faults in a major fault block.
3. Anomalous barium (100 ppm) in insoluble residues of uppermost 300 ft of drill holes.

#### *Permissive criteria*

1. Barite deposit or occurrence at surface.
2. Reported barite occurrence in drill-hole samples.

### ROLLA QUADRANGLE: BARITE POTENTIAL

The distribution of recognition criteria is shown in figure 2, a compilation of all diagnostic and permissive criteria. This figure also delineates the four areas considered to have potential for barite resources.

*Potential Resource Area 1* contains an estimated 2 million tons of residual barite; in addition, 750,000 tons are estimated to be present in tailings ponds. Much of the area has been thoroughly



prospected, so large undiscovered deposits are unlikely; however, there is high potential for the discovery of small additional deposits in the area.

*Potential Resource Area 2* contains reported but unverified occurrences of barite in the Potosi-Eminence outcrop area and high barium in insoluble residues of several drill holes (diagnostic criteria 1 and 3). The barite occurrences lie between the Black fault to the north-east and the Ellington fault to the south-west; these faults may define a structural block in the sense of diagnostic criteria 2. The extent of minor faulting in this area is unknown. Any existing major residual barite deposits would probably have been penetrated by some of the many holes drilled for water supply or base metal exploration or simply observed in outcrops, so there is little potential for the discovery of additional large deposits.

*Potential Resource Areas 3 and 4* contain insoluble residues with high barium content in several drill holes in the Potosi-Eminence outcrop area. These areas possess potentially favorable structure but have been intensely explored by drilling. Only small deposits are likely to be found here.

The remainder of the Potosi-Eminence outcrop area and the remainder of the outcrop areas of the Bonneterre, Davis, and Lower Ordovician formations theoretically have some potential for residual barite deposits.

### SPRINGFIELD QUADRANGLE: CRUSHED STONE POTENTIAL

Following completion of the Rolla Quadrangle study, work began on the Springfield Quadrangle, immediately west of the Rolla Quadrangle. The mineral resources of the Springfield Quadrangle differ greatly from those in the Rolla Quadrangle. All active mineral production is from industrial minerals; there has been no metal production since 1950. For this reason industrial minerals were included in the evaluation from the beginning of the study. The following industrial minerals were considered worthy of evaluation: barite, clay and shale, crushed stone, dimension stone, industrial sand, and construction sand and gravel. Of these, only crushed stone and construction sand and gravel are of current economic importance. Lime, a product manufactured from crushed limestone, was not considered separately.

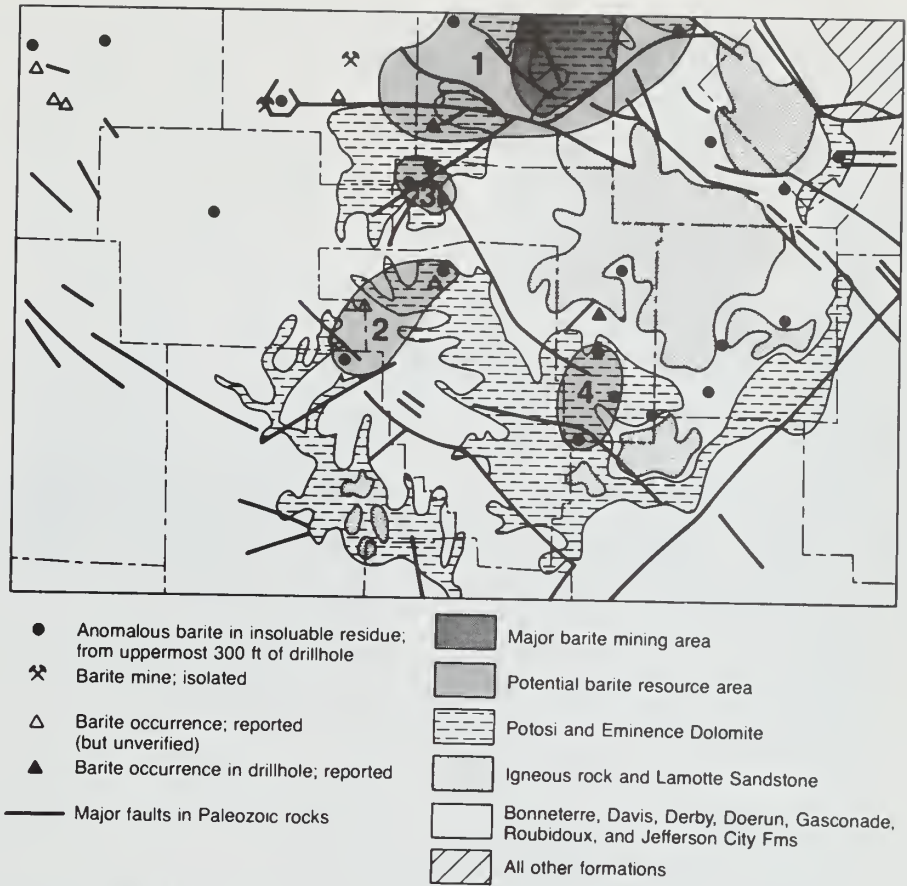


Figure 2 Barite resources (adapted from Pratt and Wharton, 1981).

The following brief description of the appraisal of one mineral commodity, crushed stone, illustrates the approach used in the Springfield Quadrangle.

The crushed stone resources of the Springfield 2° Quadrangle are limestone and dolomite; sandstone is considered an industrial sand resource. Some rock units are suitable for all types of construction aggregate, others for aglime, and many for chemical and industrial use; some resources have little or no economic value.

For the appraisal, the nine mapped geologic units were grouped into use categories based on the known or potential suitability of the unit for various commercial uses. The use category for each unit was determined by present or past performance records of material from that unit, laboratory tests, and similarity to other rock units with a history of use either within the quadrangle or elsewhere in southwest Missouri. Six use categories were developed for stone resources in the quadrangle (modified from Rueff, 1985):

1. Known resources of high-purity limestone. (High-purity limestones are formations that contain a significant section of limestone with a minimum CaCO<sub>3</sub> content of 95 percent.) This criterion is met in two limestones.
2. Known resources of high-specification aggregate. (High-specification aggregate is an arbitrary category to designate stone that meets standard specifications established by the Missouri Highway and Transportation Department and other agencies for aggregate used in portland cement concrete.) Stone resources meeting this criterion occur in two limestones.
3. Known resources of commercial limestone and dolomite. (Commercial limestone and dolomite is a category that includes rock units in the high-purity limestone and high-specification aggregate categories and those suitable for less stringent aggregate and aglime use.) Stone resources in this category are present in seven formations.
4. Known resources of high-purity dolomite. (Resources in this category

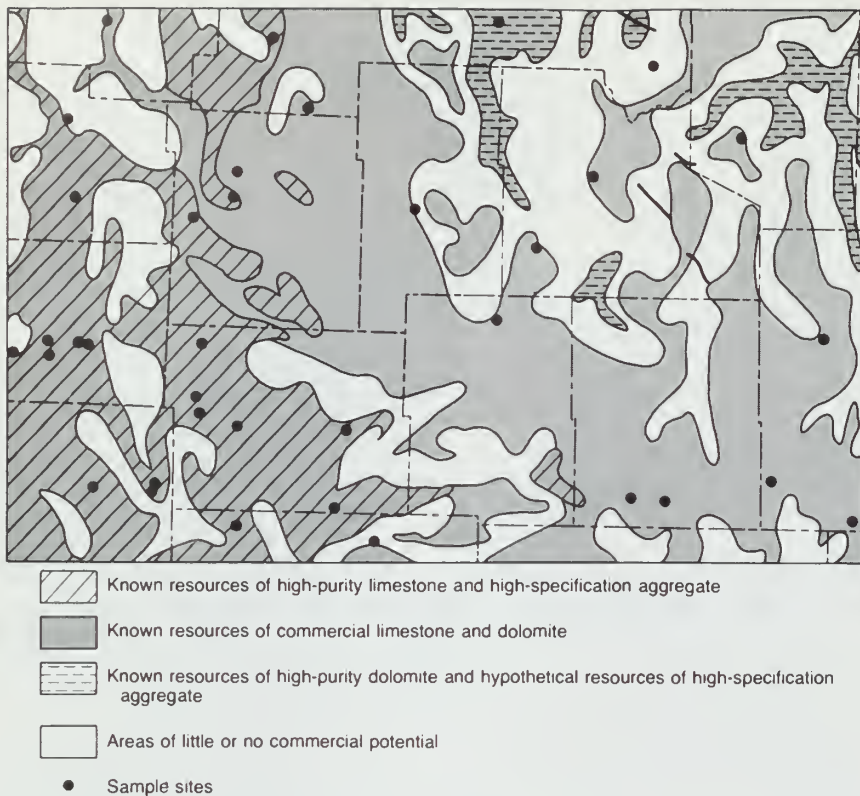


Figure 3 Crushed stone resources (adapted from Rueff, 1985).

are dolomite formations that contain a section of stone with a combined  $\text{CaCO}_3$  and  $\text{MgCO}_3$  content of at least 95 percent and at least 40 percent  $\text{MgCO}_3$ .) This criterion is met in two formations.

5. Speculative resources of high-specification aggregate. (This category consists of stone resources that may contain a section of stone meeting the specifications of high-specification aggregate, as defined earlier.) Only one formation is in this category.
6. Areas and/or units with little or no commercial potential. Large areas of the Springfield Quadrangle are underlain by formations having little or no commercial value as crushed stone aggregate. Five mapped rock units and areas near several major structural features are included in this category.

The crushed stone resource map (fig. 3) is derived from the geologic map and from the earlier defined use categories.

This map and the other industrial mineral maps are at a scale of 1:500,000 rather than the 1:250,000 scale used in the Rolla Quadrangle; they are intended only as guides because detailed geologic open-file maps at a scale of 1:62,500 are available. Tables providing average chemical and physical data for each unit having commercial potential also were prepared. Detailed chemical and physical test data also are available as open-file reports. Graphs were prepared to show past stone production and current stone use.

#### FUTURE APPRAISALS

The purpose of this report is not to discuss what has been done on the preceding CUSMAPs but to stimulate discussion about what can and should be done for future industrial mineral appraisals. Although the methodology used in the previous studies has worked well for metallic minerals, it does not, for example, address cultural factors and other factors that so greatly affect many in-

dustrial minerals. Kentucky, Illinois, Indiana, and Missouri have recently begun a CUSMAP appraisal of the Paducah  $1^\circ \times 2^\circ$  Quadrangle, and an excellent opportunity exists to develop an appraisal system to meet the needs of the mineral industry, the government planner and regulator, and the general public.

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# Silica Sands: Grassroots Exploration or Acquisitions

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## ABSTRACT

*In the United States, companies interested in silica sands have done little grassroots exploration (that is, discovering a silica sand deposit by geologic methods and then developing it). Simple reconnaissance of known sites is common, but actual development of a deposit is rare. Most of the silica sand sites, whether consolidated or unconsolidated, are known. When these sites are rediscovered, as they often are, individuals wishing to capitalize on their discoveries solicit financial backing from large companies. However, although these rediscovered deposits may at first attract considerable interest, they seldom get developed.*

*In recent years the silica sand industry in the United States has consolidated to a few major companies, and further consolidation is expected. Only three new plants have been built in the past 6 years. Many factors associated with this industry prompt companies to acquire ongoing operations rather than explore for new deposits.*

*This paper includes an examination of markets, industry trends, processing, and product specifications, and a brief discussion of some of the factors that favor acquisitions over grassroots exploration.*

## INTRODUCTION

In recent years the silica sand (industrial sand) industry has experienced some major changes, especially consolidation of operations by acquisitions. In 1978 approximately 31 companies in the United States were operating about 59 plants that provided most of the silica sand needed by industry consumers. Today approximately 10 major companies operate about 55 plants. The most recent U.S. Bureau of Mines survey (1987) lists 94 companies that control 166 active operations. However, approximately six companies are closed, the industrial sand-producing status of 36 companies is questionable, one company no longer exists, one is sand and gravel only, and one is listed twice. The remaining companies have consolidated with the majors or produce only one or two industrial sand products such as blasting, traction, or molding sands. Although the acquisition of companies is not new for this industry, in recent years a few com-

panies have taken significant steps to acquire silica sand plants. In the late 1970s, some established silica sand producers began acquiring other such companies. The Jesse S. Morie Company of New Jersey (now the Morie Company) bought the Georgia Silica operation from the Georgia Marble Company and subsequently acquired the Hardy Sand Company's plants in West Tennessee and Alabama. Unisil Corporation had already begun to buy small operations and eventually became Unimin Corporation (financed by Shibelco, Belgium).

In the early 1980s, Martin Marietta's purchase of the Wedron Silica Division of Del Monte properties increased its holdings from 6 to 11 plants. British Industrial Sand bought two plants from Manley Brothers and reportedly planned to acquire additional plants. Unimin Corporation, probably the most aggressive silica sand company in the United States, bought Carolina Silica near Marston, North Carolina, and plants that Martin

Marietta had purchased previously from Wedron. Unimin built a plant near Kasota, Minnesota, and reconstructed a Whitehead Brothers (now WHIBCO) plant in southern New Jersey. Unimin recently purchased Minnesota Frac Sand Company from J.L. Shiely Company near Jordan, Minnesota, Mid Continent Glass Sand Company at Roff, Oklahoma, and the International Minerals and Chemicals (IMC) operation at Spruce Pine North Carolina; the latter operation is silica-related but also produces other industrial minerals from the deposit. The one Wedron plant not sold to Unimin by Martin Marietta was at Wedron, Illinois; this plant was purchased by Best Silica Company (owned by Fairmount Minerals, Limited) and some of the remaining Wedron personnel.

The most recent and largest acquisition was U.S. Borax's purchase of Pennsylvania Glass Sand Company, an ITT subsidiary, and subsequent acquisition of Ottawa Silica Company's six plants. Olgabay-Norton purchased a plant near Riverside, California, from Owens-Illinois and lands near Marston, North Carolina, to continue its activity in this business. During this time only three new plants were built. Vulcan built a plant near Brady, Texas, for the production of frac sand, Unimin built near Kasota, Minnesota, to produce frac or proppant sand, and J.L. Shiely Company built near Jordan, Minnesota, to produce frac or proppant sand. Another new plant is currently producing glass sand and other sands for specialty uses near Corona, California.

## PRODUCT SPECIFICATIONS

The rigid specifications placed on silica sand products such as glass sand, foundry sand, blast sand, and filter sand dictate the products a company can produce from a deposit. Generally, the most stringent requirements regarding grain

size, chemistry, and heavy minerals content apply to glass sand. Grain sizes for melting sands require that less than 1 weight percent be retained on a 30-mesh sieve. The amount retained on the 40-mesh sieve is usually held to 8 to 10 weight percent and that passing a 140-mesh sieve to less than 5 weight percent. The SiO<sub>2</sub> content must exceed 99.5 percent. The Al<sub>2</sub>O<sub>3</sub> content should be less than 0.30 percent and the iron content, expressed as Fe<sub>2</sub>O<sub>3</sub>, should not exceed 0.30 weight percent. Limits are also placed on the amount of heavy refractory minerals (commonly found in young, unconsolidated quartzose sand deposits) melting sands can contain. Furthermore, heavy refractory minerals are likely to come under even more stringent specifications by the container industry (Kephart and DeNapoli, 1981).

Foundry sand users may require a four-screen distribution, free from calcium carbonate and other impurities. Filter sand users demand a strict uniformity coefficient and effective size specifications, and even blast and traction sand users impose certain specifications. Grain shape and screen size requirements generally limit the products and determine the processing requirements and the markets for deposits exploited for proppant sands. Favorable geology, selective mining, quality processing, and creative marketing are required for a company to successfully meet the many specifications for these markets.

## MARKETS AND TRENDS

To a large extent, markets dictate a company's decision to acquire or to explore and build. For example, the supply of silica sand for midwestern consumers has always been plentiful. Long-established companies produce from the St. Peter and Jordan Sandstones, Michigan dune sand, the Sylvania Formation, and the Sharon Conglomerate. Price wars are common, especially in the Illinois-Indiana area. The odds are against a newcomer's building a successful plant in this area. For example, the J.L. Shiely Company built Minnesota Frac Sand Company to produce frac sand, but failed; Unimin subsequently bought the plant and is reported to be working from the stockpiles only. Unimin also closed its plant at Kasota, Minnesota. Although the oil industry slump did not help these two plants (which produced mostly proppant

sand), the market was already saturated. In addition, major silica sand markets (for container glass) are being adversely affected by strong competition from aluminum cans and plastic bottles, as well as the cardboard drink boxes recently introduced to the market. The use of these nonglass containers is expected to grow in the next several years, possibly further reducing the demand for glass containers.

Cullet (recycled glass) is also utilized in the container glass manufacturing process, generally replacing 15 percent of the raw material mix. The cullet used in a container plant is obtained both from in-house sources and from outside recycling. The bottle bills of the Northeast requiring use of returnable bottles have created a large supply of cullet. Major beer and soda distributors have set up recycling plants to separate and clean cullet for shipment, usually by rail, to glass-container plants. Cullet is an important ingredient of the manufacturing process because it takes less energy to melt cullet than the same weight of raw sand. This recycling trend will continue to expand and reduce consumption of silica sand.

The volume of float glass (window and plate glass) produced generally varies with the economy, since this glass is used in the automotive and construction industries. Because new cars are smaller and require less glass and because the thickness of windshields and lights is decreasing, not much increase in glass-sand demand is expected in the automotive industry. Recent increases in residential and commercial construction will boost float glass consumption.

Some increase in silica sand (industrial sand) sales may come from products such as roofing sand, blasting sand, filter sand, and other specialty sand products. These areas demand new processing and marketing skills. One major new market for silica sand consists of high-purity products requiring unique processing for relatively small tonnages for piezoelectric and glass crystal products.

Any company has difficulty breaking into the market as a new supplier of silica sand. The customer may at first insist on using a new supplier's product on a trial basis and later, if the product meets approval, as a second source. No longer will consumers buy all their silica sand from one plant unless special contracts, providing for a continual supply of qual-

ity material at a fixed price for a number of years, are considered.

## PROCESSING AND PLANT COSTS

Beneficiation is usually the key to making a high-quality glass or foundry sand. To remove clay coatings from the sand grains, the sand is pumped to a primary desliming processor in which hydrocyclones remove the -0.01-mm particles. The slimes are pumped to settling ponds. After primary desliming, the clean sand is sized, either by hydrosizing in settling tanks or on screens or possibly a combination of both to remove the coarser fraction, usually the +0.5-mm particles (about 30-mesh size). Some plants also use air classifiers to size dry sands. The sand is dewatered by hydrocyclones to 70 percent solids for attrition scrubbing. Scrubbing is done in a compartment (sump) with propeller-type scrubbers. This process loosens and disperses any remaining coatings of iron oxides, clay, and other materials affecting the sand grain surfaces; it also prepares fresh surfaces on the nonquartz minerals (heavy minerals) for reagent attachment in the next step, which is flotation. Sometimes sodium hydroxide is used in the attrition scrubbing process to help clean the grain surfaces.

Flotation, if necessary, is the next step in processing the sand. Two types of flotation, cationic and anionic, are used. Anionic flotation is probably the more common; this process uses a petroleum sulphonate collector in an acid circuit to remove the heavy minerals.

The sand must be dried for most uses, and this is normally done in rotary or fluidized-bed dryers with wet scrubbers for dust removal on the exhaust system. Many glass sand producers use the fluidized-bed-type dryers because of their high reliability and thermal efficiency. After drying, the sand may be conveyed to another set of screens for final size control. Foundry sands generally have to be cooled before shipment; therefore, a cooling cycle must be introduced to the sand processing operation.

The type of processing just described is generally more elaborate, and therefore more costly, than that required to produce construction-grade sand and gravel. A plant built to produce 500,000 tons of high-quality glass or foundry sand per year may require more than 10 million dollars in engineering and construction

costs. A new plant being built near Corona, California, with a capacity to produce 600,000 tons per year reportedly will cost \$16 million, whereas a plant designed to produce comparable tonnages of sand and gravel for construction aggregate may cost \$3 to \$5 million dollars to build.

The following list (adapted from Shuffelbarger, 1983) includes processing steps that may be used in producing high-quality glass sand. The exact mix of processes is a function of the deposit, products, and markets.

1. drying and screening only (rare)
2. washing (generally the removal of clay), drying, and screening
3. washing, scrubbing, flotation, drying, and screening
4. washing, scrubbing (high pulp density attrition), drying, screening, and sizing by air separation
5. iron removal (magnetic, wet, or dry)
6. grinding (rod mill, ball mill), regardless of process used, if indicated by natural grain-size distribution
7. acid leach for high-purity products
8. bagging facilities for some or all products
9. cooling facilities for foundry sand

## TRANSPORTATION

The availability of good truck and rail routes near a potential silica sand site is essential for profitable operation. Although silica sand, unlike sand and gravel, may be shipped great distances to a customer, it is not always possible to have a distribution center close to the market and still be competitive: high labor and equipment costs are required to place a large number of trucks or railcars at the disposal of the glass customers or users to ensure that the carriers can be loaded as required to meet the delivery schedule (Peddicord and Regis, 1981). Truck delivery is usually more dependable and more costly than rail delivery.

## ENVIRONMENTAL FACTORS

The silica sand industry, like most other mining industries, must obtain various types of permits to comply with

local, state, and federal environmental regulations concerning water and air quality control and mined land restoration. According to Peddicord and Regis (1981), the most important and costly of the permits required, the Conditional Use Permit Requirements, typically include:

- zoning and integration with a local master-planning and development scheme
- restrictions on operating times
- implementation of detailed water-well monitoring programs
- noise and seismic survey of blasting on and off the property
- monitoring and mitigation of fugitive dust
- complete reclamation plans
- detailed site plan

It is not uncommon today for a silica sand mining company to be permitted to mine only 50 percent of its property (Peddicord and Regis, 1981).

Health and safety constraints have been placed on silica plants, especially those manufacturing silica flour, by the Mine Safety and Health Administration (MSHA) and the Occupational Safety and Health Administration (OSHA). A bagging facility for silica sand products installed at one operation cost \$3 million to meet the health and safety requirements of the various regulatory groups.

## SILICA INDUSTRY PROSPECTS

Although most of the potential silica sand sites have been located and investigated at one time or another, "new" sites are still available as they are rediscovered. Operators of some small companies near the markets who believe that their deposits contain quality materials often fail to understand why they cannot obtain financial backing; the following factors may help explain their difficulty:

- The size of the market for industrial sand is decreasing, not growing.
- A new company must take customers away from existing companies in a saturated market.
- The high initial capital cost of opening a new mine and processing plant probably means that a newcomer will

not be able to be as cost-competitive as long-established competitors.

- In general, the deposits least costly to mine, process, and distribute, are already being exploited. If an apparently excellent deposit isn't being used, there are probably good reasons why: it may be too expensive to mine or process, it may be too distant from markets, or transportation facilities may be poor.

Extensive testing to assure product quality is often required before new raw materials can enter a market.

## CONCLUSION

I have discussed briefly some factors that persuade company officials to acquire existing operations rather than explore, purchase land, and build new silica plants. Officials of most major companies prefer to purchase a producing facility, possibly change some processing equipment, obtain new permits, and have established customers waiting for their products rather than to struggle to overcome the many obstacles (including high costs) associated with grassroots exploration and development.

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# Enhancement of Sulfur Dioxide Sorption Reactivity of Limestone

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## ABSTRACT

Direct sorbent injection for control of sulfur dioxide emissions from the combustion of high-sulfur coal is an emerging technology. In this process a pulverized, dry, calcium-based sorbent is injected directly into the furnace cavity of an existing coal-fired boiler. The sorbent (limestone or hydrated lime, for instance) decomposes to lime, which reacts with the sulfur dioxide to produce calcium sulfate. Previous studies indicate that hydrated limes are more reactive than carbonates because they produce calcines having higher surface areas.

The objective of this work was to compare the surface areas and reactivities of products prepared by three hydration methods. In the first method, lime was hydrated with alcohol-water solutions. In the second method, hydrated limes were prepared at high-pressure, 120 to 475 atmospheres and subsequently ejected to atmospheric conditions. In the third method, lime was hydrated with steam, surface areas were determined by the BET (N<sub>2</sub>) technique, and sulfur dioxide sorption reactivities were obtained by thermogravimetry.

Surface areas and sulfur dioxide sorption capacities of the hydrated limes depended on sorbent type, calcination conditions, and the method of hydration. For a particular limestone, steam hydration improved calcium utilization (conversion of calcium oxide to calcium sulfate) by 84 percent over the lime from which it was prepared. Surface areas between 34 and 49 m<sup>2</sup>/g and calcium utilizations between 50 and 93 percent were observed for limes hydrated with water at atmospheric pressure. Pressure-hydrated limes had lower surface areas (10 to 20 m<sup>2</sup>/g) and utilizations (70% to 100%). A linear relationship was observed between reactivity and surface area for these hydrates. Results from this study showed the following order of reactivity:

Ethanol-water hydration = methanol-water hydration pressure hydration \* steam hydration = with water lime.

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## Reserve Evaluation and Planning on an IBM Personal Computer

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## ABSTRACT

An IBM desktop personal computer and a proprietary software package (PC/Cores) were used for evaluating the quantities and qualities of a diabase deposit on the East Coast. The entire project is described, from borehole data capture through database development, model building, contour mapping, and interactive, multiple-seam planning. A unique method of applying quality constraints to the model and to the display of histogram and quality-array tables is also described.

The main advantage of the software is that geologists and mining engineers can use it themselves without having to consult a computer programmer.







