Relationship of Pore-Size Distribution and Other Rock Properties to Serviceability of Some Carbonate Aggregates

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The pore-size distribution present in some carbonate rocks was determined as part of an investigation concerned with the properties of aggregate which affect the durability of portland cement concrete. Pore-size distribution is considered critical in controlling flow and retention of water in aggregate. Those properties, in turn, might be related to an aggregate's effect on the resistance of concrete to freezing and thawing and to potential chemical reactivity of the aggregate in concrete.

Pore-size distributions were determined by use of a mercury capillary pressure apparatus manufactured by Ruska Instrument Corporation, Houston, Texas. Several other properties of the rocks were also determined. Those properties are: (a) effective porosity; (b) total porosity; (c) calcium-to-magnesium ratio; (d) insoluble residue; (e) amount of clay present; (f) loss in weight in freezing-and-thawing; (g) service record; and (h) texture.

Questions were then asked about the relationships between the various properties and answered tentatively on the basis of the frequency of occurrence of the properties in these data.

This investigation shows:

1. A large number of the rocks sampled possess pore entry distributions with maxima in the region of radius less than, or equal to, 0.5 micron.
2. Knowledge of a rock's pore-size distribution might be of value in attempting to predict its service record as aggregate.
3. The "frequency-of-occurrence" method lends itself well to the characterization of a rock. One may ultimately be able to thus describe a typical "poor" aggregate.

● THIS STUDY was carried on as part of a research project sponsored by the Iowa State Highway Commission. The purpose of this program is to discover those properties of a carbonate rock which determine whether or not it will be a satisfactory aggregate for concrete highway construction.

Literature concerned with the behavior and suitability of carbonate rocks as concrete aggregate is not extensive. One of the earliest investigations of this subject was that by Laughlin (9). Others of significance are those by Sweet (21) and Mather (12, 13).

Investigations concerning the suitability of Iowa carbonates as concrete aggregate were inaugurated at Iowa State College by Dorheim (5). It had been recognized that certain carbonate rocks, when used as aggregate, did not always behave as indicated by standard acceptance tests. The purpose of that investigation was to find, through consideration of some of the geologic aspects, new criteria for the selection of limestone aggregates to be used in concrete. From the parallelism between the service record and the clay content he concluded that the presence or absence of clay could be used as an additional guide upon which to select more discriminatingly limestone aggregates.

The work was continued by Roy, Thomas, Weissman, and Schneider (19). Of special interest in their investigations were the Mississippian rocks quarried in the vicinity of LeGrand, Iowa, which have a poor service record when used in concrete highways. The study of concrete cores taken from satisfactory and from distressed pavements indicated that fresh stone gave satisfactory service, whereas weathered stone produced distress. The absorption capacity, in itself, did not indicate unsatisfactory stone. It was concluded that increased absorption and presence of swelling clay favored reactions between cement and aggregate which produce distress.

Bisque and Lemish (2) investigated some of the chemical properties of carbonate rocks as related to durability of concrete. Carbonate rocks of Devonian age were sampled and rocks from the Glory quarry, which have an unsatisfactory service record as aggregate, were used as a basis for comparison. They reported the presence of silicified reaction shells on the periphery of some carbonate rocks present in affected concrete.

Lemish, Rush, and Hiltrop (10) reported on some of the physical properties of carbonate aggregate as related to the durability of concrete. Devonian rocks from the Cedar Valley formation were sampled on a bed-by-bed basis in four quarries, one of which was the condemned Glory quarry. It was concluded that impure carbonate rocks characterized by high insoluble residue and high clay content were poor aggregate. Also, preliminary investigation of pore-size distribution indicated that property would be important in determining a rock's suitability for concrete aggregate. This led to the advisability of a more detailed study of pore-size distribution (7).

In the present work, the pore-size distributions of some Devonian and some Mississippian rocks have been determined and classified. It can be seen (Fig. 1), that shape of the pore-size distribution curve alone is not indicative of whether or not a specimen will pass freezing-and-thawing tests. For example, 10 out of 13 Devonian rocks possessing curves of type 7 fail freezing-and-thawing tests[1]. On the other hand, all rocks of Mississippian age possessing distributions of type 7 pass. Considerations of this nature lead one to suspect that a rock's freezing-and-thawing performance is very probably a function of several parameters. Also, serviceability of concrete aggregate is possibly a function of several variables, one of which might be pore-size distribution. It is the purpose of this paper to state how pore-size distribution is determined, to show the shape of the distribution curve possessed by the several specimens from the eight quarries investigated, and to show the relationships (where relationships are seen to exist) between serviceability, pore-size distribution, freezing-and-thawing data, insoluble residue, calcium-to-magnesium ratio, amount of clay mineral present, texture, effective porosity and total porosity.

It is not possible at this time to write the mathematical expression which describes the relationship between serviceability and the variables listed above. In order to gain some idea of the answer to the question of how serviceability varies with a change in

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[1] The present acceptance tests and standards for coarse concrete aggregates are described in the "Standard Specifications" Series 1956, Iowa State Highway Commission, Section 4107. Abrasion, soundness, former service record, and amount of objectional materials present are the main considerations in the specifications. The soundness test consists of a water-alcohol freezing-and-thawing test as described in Section 4101-2C, of the "Standard Specifications". The freezing-and-thawing values, reported in the tables of this paper and used for the purpose of correlation with the various properties of the carbonate rocks, represent the percentages of material passing a No. 8 sieve after 16 cycles of freezing and thawing in a water-alcohol solution containing 0.5 percent alcohol (by weight). A maximum of 6 percent material loss is allowed for concrete aggregate.

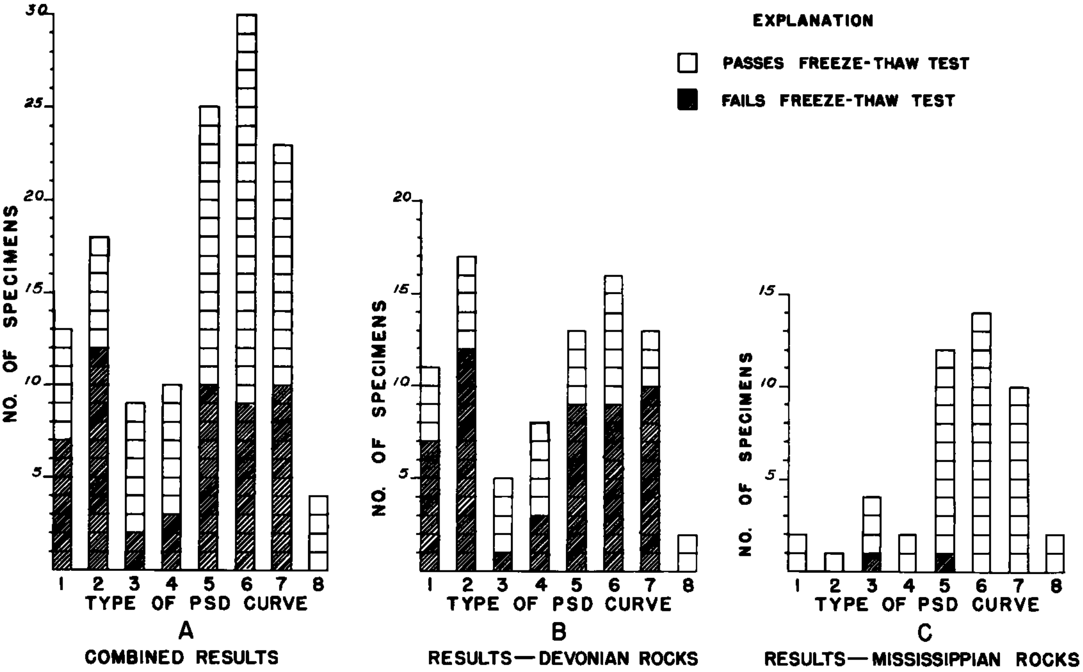


Figure 1. Relationship of freeze-thaw results to the type of pore-size distribution (psd) curve.

these independent variables, frequencies of occurrence were observed. This method allows one to compare frequencies of occurrence of the parameters involved and to see which of these parameters appear critical.

QUARRIES

In order to have representative specimens and at the same time have rocks on which as many other pertinent data as possible were available, specimens were collected from eight quarries in northeastern Iowa. Specimens of approximately 100 lb each were taken on a bed-by-bed basis to represent all lithologic variations present. The location of each quarry is shown in Figure 2. Five are in the Devonian Cedar Valley formation; three, in Mississippian rocks.

The Cedar Valley formation is composed almost entirely of carbonate rocks, the lithology of which varies both horizontally and vertically. The composition ranges from pure carbonates (limestones and dolomites) to those which are argillaceous or cherty. The formation is divided into three members, from bottom to top, the Solon, Rapid, and Coralville. The Solon member is a massive, fine-grained, light gray to buff limestone with characteristic zones of black fossil fragments. The Rapid member consists mainly of a gray, argillaceous, calcitic dolomite with abundant chert moduls. The Coralville is generally a buff-colored rock and varies in composition both horizontally and vertically from calcitic dolomite to beds of very dense ultra- fine-grained (lithographic) limestone.

Figure 3 shows the stratigraphic range of rocks exposed in the five quarries which produce from the Cedar Valley formation. It also shows which portion of the stratigraphic interval is, or was, used for concrete aggregate.

The Solon member yields acceptable concrete aggregate at the Burton Ave. quarry. The Coralville is acceptable at Burton Ave., but not enough is present to be economically quarried. Concrete aggregate is quarried from the Coralville at the Pint, Newton, and River Products quarries and a small portion of the Coralville present at the Glory

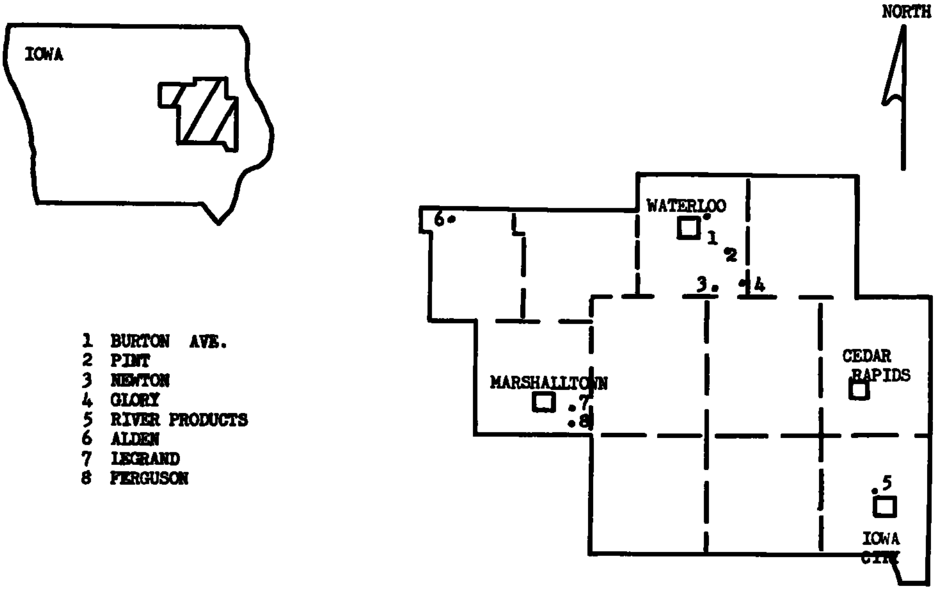


Figure 2. Index map showing the approximate location of the eight quarries sampled.

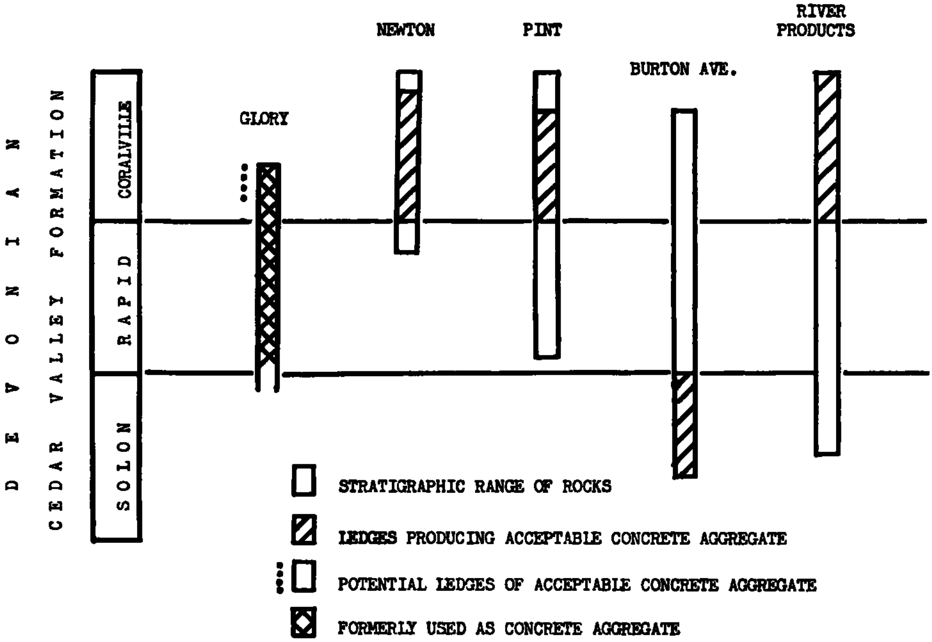


Figure 3. Startigraphic range of the rocks exposed in five quarries and the position of the concrete aggregate ledges.

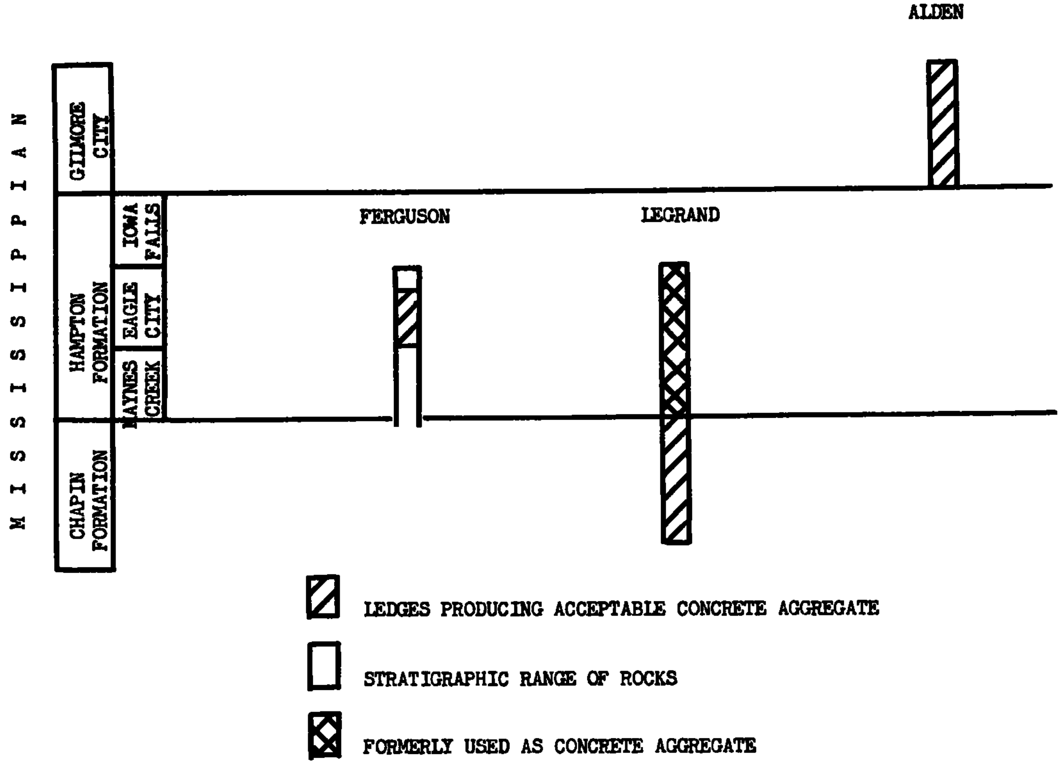


Figure 4. Stratigraphic range of the rocks exposed in three quarries and the position of the concrete aggregate ledges.

quarry will also pass acceptance tests. According to present acceptance tests, the Coralville is probably the best source of concrete aggregate in the Cedar Valley formation.

The Rapid member is found in all five quarries and generally does not pass the present acceptance tests for concrete aggregate. Most of the face exposed in the Glory quarry belongs to the Rapid member and was used extensively as concrete aggregate throughout eastern Iowa but is unacceptable by present standards It has also proven itself to be unsatisfactory in service.

In the remaining three quarries, Ferguson, LeGrand, and Alden, aggregate is quarried from Mississippian rocks (Fig. 4).

The LeGrand quarry is in the Chapin and Hampton formations. Aggregate from the "basal" oolite, or Chapin formation, has been used in a few short sections of pavement with satisfactory results. Aggregate from the overlying Hampton, however, passes acceptance tests but fails in service. This fact presents one of the interesting enigmas of the carbonate aggregate problem.

The Ferguson quarry, though underlain by the Chapin oolite, provides aggregate from the overlying Hampton formation. A part of the Hampton formation from this quarry is used as concrete aggregate but, to date, insufficient evidence has been compiled to be able to designate it as either satisfactory or unsatisfactory in service

The Alden quarry provides aggregate from the Gilmore City formation That unit is known to unconformably overlie the Iowa Falls member of the Hampton Rocks from the Alden quarry are generally coarsely crystalline, light gray, massively bedded, oolitic limestones. All aggregate from this quarry passes acceptance tests and has an excellent service record.

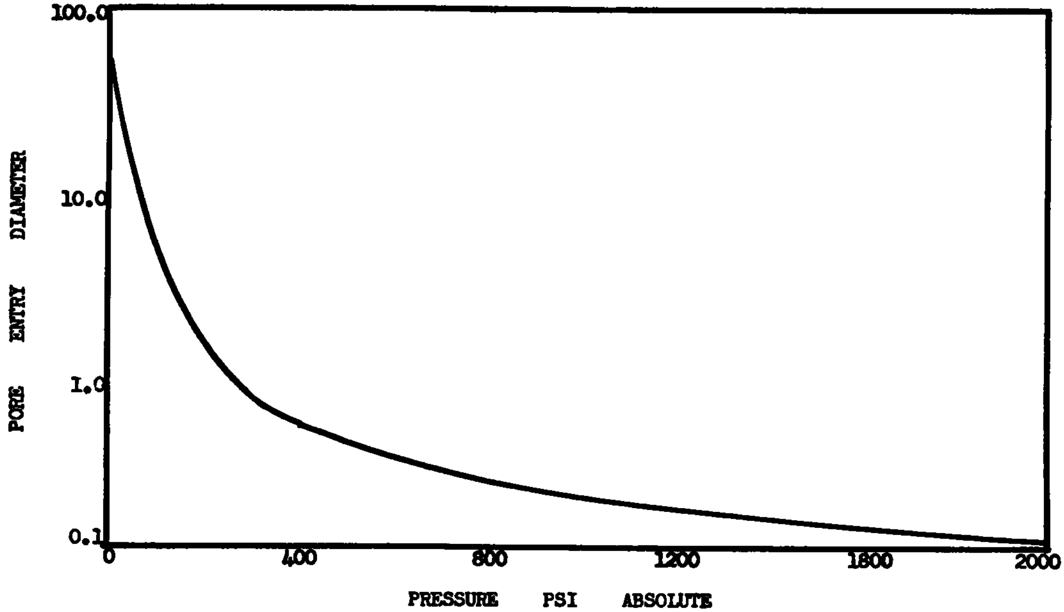


Figure 5. Equilibrium pore diameter in microns vs absolute pressure in psi.

EXPERIMENTAL METHODS

Pore-Size Distribution

The determination of pore volume is usually calculated by taking the difference between two specific volumes. Thus, the total internal pore volume is found by subtracting powder volume from bulk volume. McBain (14) presents a complete discussion of terms and procedures involved in determinations of this kind.

In situations where rates of diffusion or availability of internal surfaces are deciding factors, pore sizes present and pore-size distribution could be more important than total pore volume. It might be assumed that such a situation does exist when carbonate rocks are utilized as aggregate in concrete.

Pore space characteristics are important because of their direct influence on other physical and chemical properties. Pore characteristics determine the flow of moisture into and out of an aggregate, its water retentivity, and the development of pressure during a freezing-and-thawing cycle. In addition, the interior surface of the pores provides an area on which surface-chemical reactions can proceed between mobile substances and the constituents of the rock.

Considerations of this nature have led Blanks (3), Rhoades and Mielenz (17), and Lewis and Dolch (11) to state that characteristics of the pore space in concrete aggregates are among the most important of all aggregate physical properties.

Relation of Pore Size to Pressure. Washburn (22) originally presented the idea that the size of a pore in a porous medium could be determined by measuring the pressure that must be applied to some non-wetting liquid (for example, mercury) to cause that liquid to enter the pore against the repelling force of the capillary pressure which it exhibits.

The relation (quoted from Washburn) giving the pressure required to force liquid into a pore of a given size is

$$pr = -2\sigma \cos\theta \qquad (1)$$

where p is the pressure, r the pore radius, σ the surface tension, and θ the contact angle. It may be derived as follows·

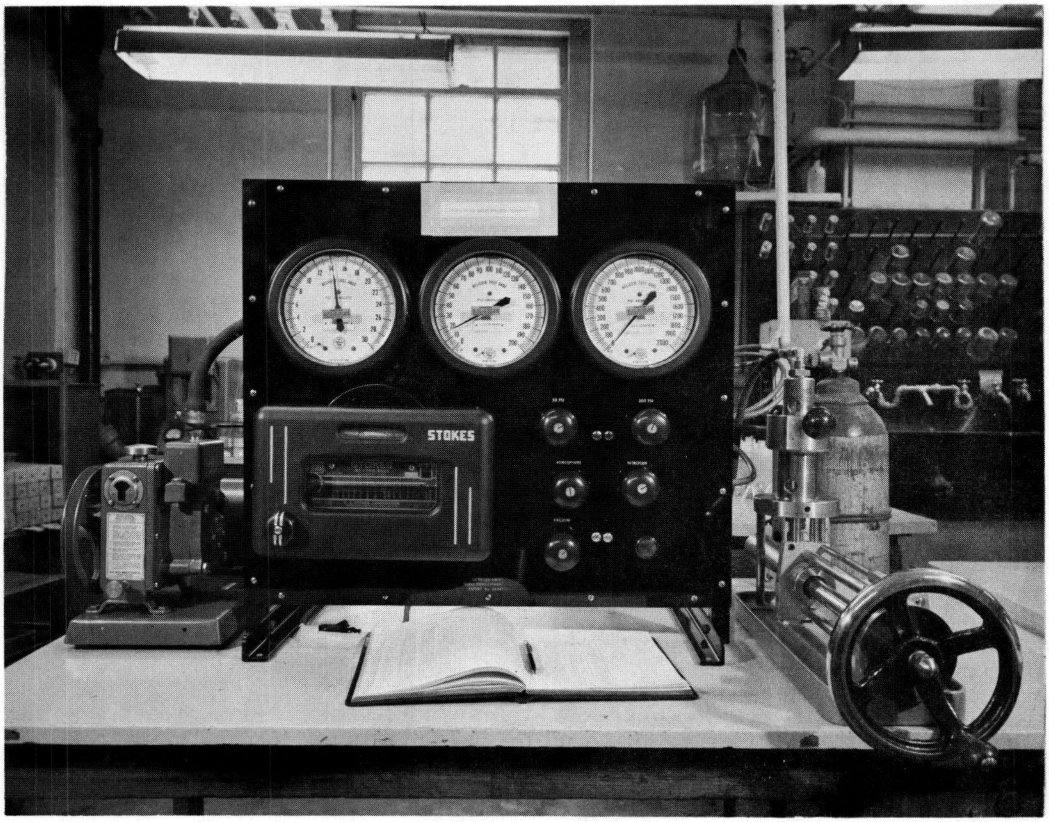


Figure 6. Mercury capillary pressure apparatus manufactured by Ruska Instrument Corporation, Houston, Texas.

In a pore of circular cross-section, the surface tension acts along the circle of contact over a length equal to the perimeter of the circle. This force is $2\pi r \sigma$. Normal to the plane of the circle of contact, the force tending to squeeze the liquid out of the pore is $-2\pi r \sigma \cdot \cos\theta$. (The negative sign arises from the fact that the angle between the direction of action of the surface tension and the positive normal to the plane of contact is $\pi - \theta$. Since $\theta > 90$ deg, the term $-2\pi r \sigma \cdot \cos\theta$ is intrinsically positive.) Opposing this force is the applied pressure acting over the area of the circle of contact with a force equal to $\pi r^2 p$. At equilibrium these opposing forces are equal: $-2\pi r \sigma \cdot \cos\theta = \pi r^2 p$, whence Eq. 1 follows immediately.

If Eq. 1 is solved for 4,

$$r = -\frac{2\sigma \cdot \cos\theta}{p} \qquad (2)$$

and the general form of this relationship is shown graphically in Figure 5.

Mercury Capillary Pressure Apparatus. Ritter and Drake (18) made use of the dilatometer for measuring the relatively small changes in volume of mercury imbibed as porous materials were subjected to pressures ranging from 25 to 10,000 psia.

Purcell (16) developed less complicated equipment for measuring pores of such size that their capillary pressures would range between 20 and 2,000 psia. This mercury capillary pressure apparatus (Fig. 6) is manufactured and distributed by Ruska Instrument Corporation, Houston, Texas, under license of the Shell Development Company.

The apparatus consists of a modified 100 cc volumetric pump with attached sample

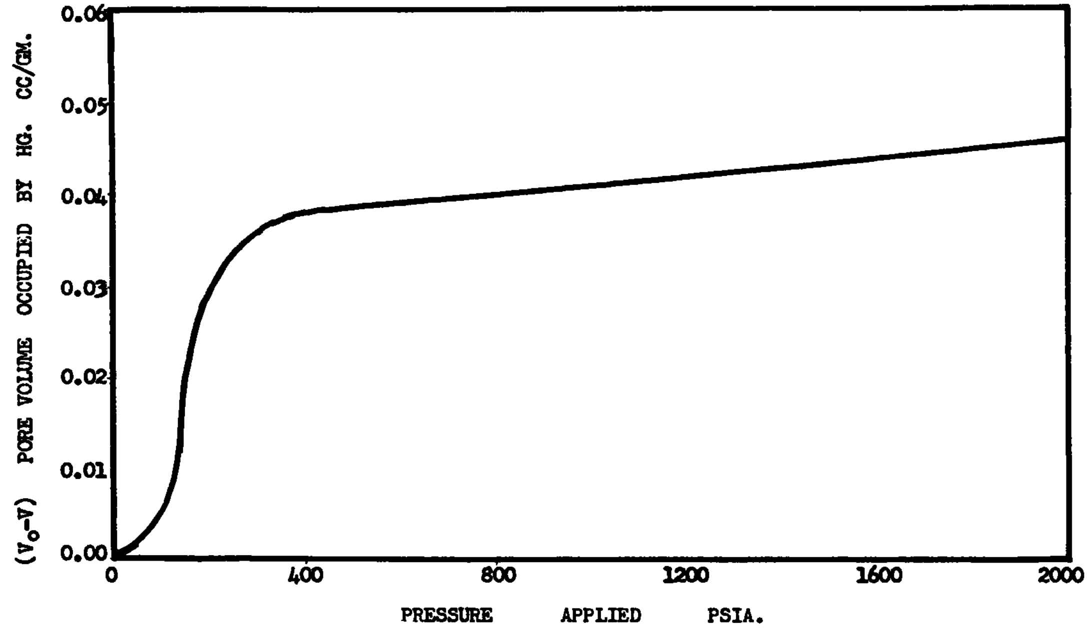


Figure 7. Pressuring curve for sample number 14 from the Rapid member of the Glory Quarry.

chamber which is connected to a manifold system with manometer and pressure gauges. The pump has a special scale arrangement for reading volume changes in the sample chamber to an accuracy of 0.01 cc. The chamber has a cylindrical cavity 2 in. long and 1 in. in diameter provided with two windows for observing reference levels. The manifold system is mounted on a bakelite panel and permits evacuating the apparatus with a vacuum pump and building up pressure with bottled nitrogen.

After the apparatus has been evacuated to a pressure of between 10 and 20 microns of mercury, the sample chamber with the core is filled with mercury by advancing the pump plunger until the mercury level reaches the reference mark in the upper window. The bulk volume of the core is read directly on the scale of the pump. The pressure in the mercury-filled sample chamber is then increased step-by-step by releasing nitrogen into the system and the amount of mercury forced into the core is determined by a reading on the pump upon returning the mercury level to the reference mark.

By measuring the change in the volume of mercury absorbed at corresponding changes in pressure applied, a distribution curve for the existing pores within the rock is determined.

Derivation of Distribution Function. The mercury capillary pressure apparatus was used to collect the pore-size reported in this paper. The relationships through which one can go from the data collected to the final distribution curve were derived by Ritter and Drake (18) and were arrived at in the following manner:

Let the total volume of all pores having radii between r and r + dr be

$$dV = D(r)\,dr \tag{3}$$

where D(r) is the non-normalized distribution function for pore size. From Eq. 1, assuming constant σ and θ,

$$p\,dr + r\,dp = 0. \tag{4}$$

Eliminating r and dr from Eq. 1, 3, and 4 gives

$$dV = D(r)\frac{2\sigma\cos\theta}{p^2}\,dp = -D(r)\frac{r}{p}\,dp. \tag{5a}$$

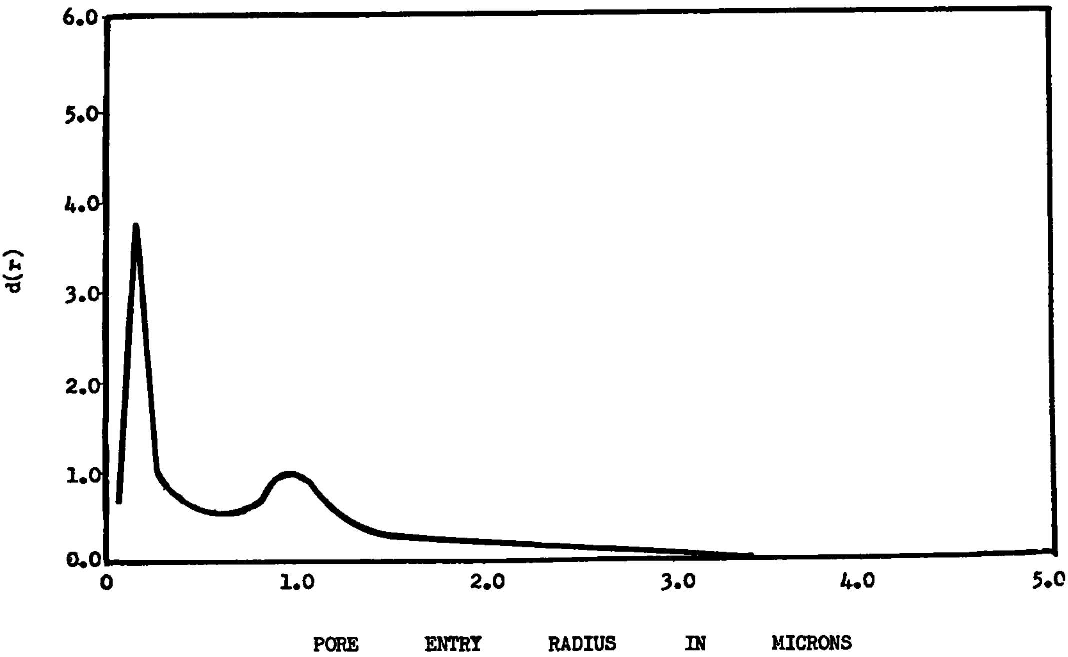


Figure 8. Normalized distribution curve. Sample number 14 from the Rapid member of the Glory Quarry.

The volume measured by the mercury pump is the volume of all pores having radii greater than r; that is, the total pore volume, V_0, decreased by the volume, V, or pores smaller than r. Thus, the pressuring curves (Fig. 7) plot $V_0 - V$ as a function of p. The slope of the pressuring curve, $\frac{d(V_0 - V)}{dp} = -\frac{dV}{dp}$, is then an experimentally determinable quantity and Eq. 5a may now be rewritten in the form

$$D(r) = \frac{p}{r}\,\frac{d(V_0 - V)}{dp} \qquad (5b)$$

in which all the terms on the right are known or determinable.

Values of the derivative in Eq. 5b, required to evaluate D(r) are readily obtained by graphical differentiation. For a number of values of p the pressuring curve is differentiated to obtain $d(V_0 - V)/dp$, r is calculated from Eq. 1, and D(r) is calculated from Eq. 5b.

Plotting D(r) against r gives the non-normalized distribution curve.

If f(x) is defined and everywhere continuous for all $a \leq x \leq b$ and is a normalized distribution function of x, then

$$\int_a^b f(x)\,dx = 1.$$

Therefore in order to normalized D(r) it must be multiplied by a normalizing factor N, where

$$\int_a^b N\,D(r)\,dr = 1.$$

Taking the constant factor outside the integral sign

$$N\int_a^b D(r)\,dr = 1,$$

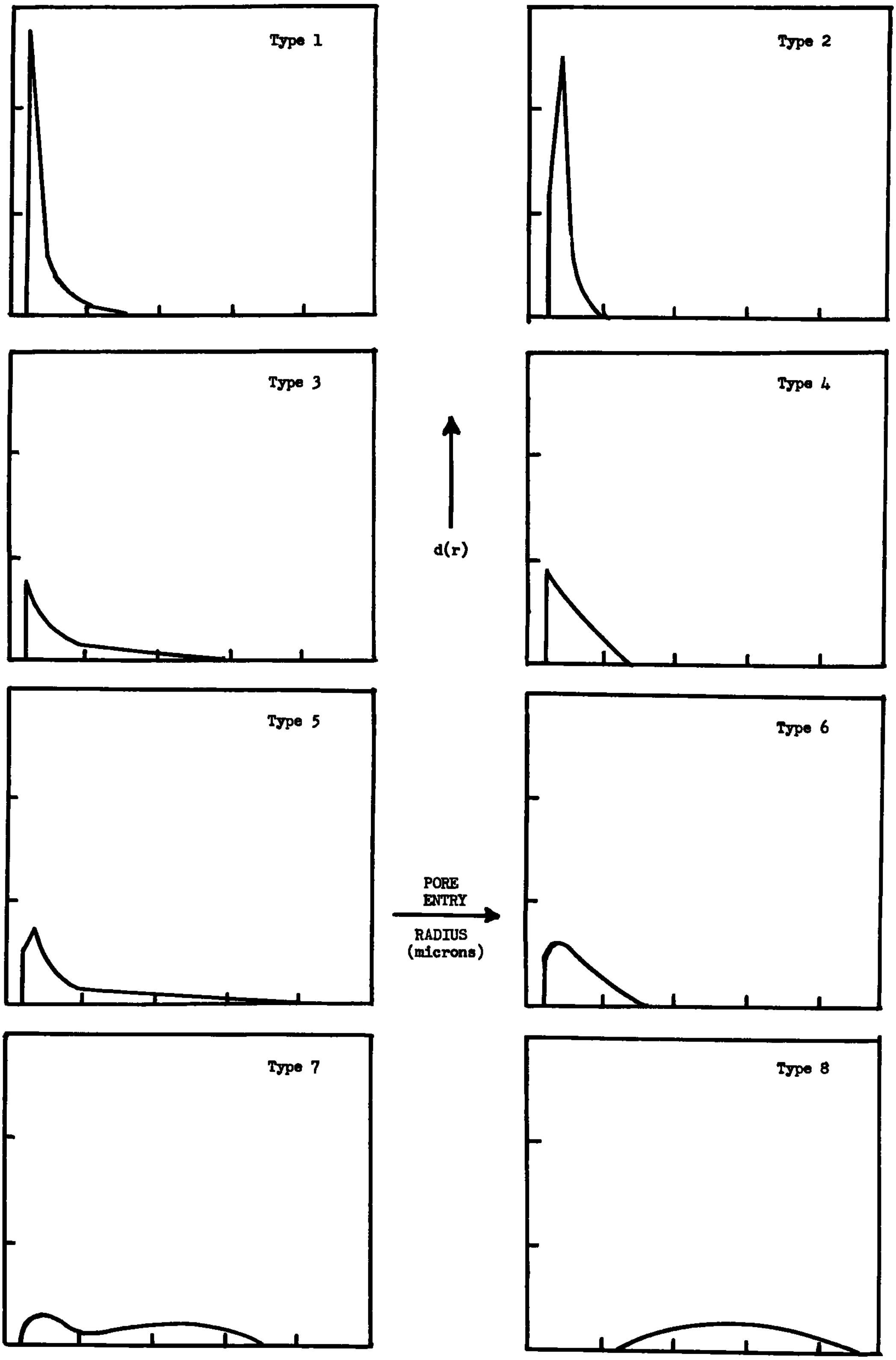
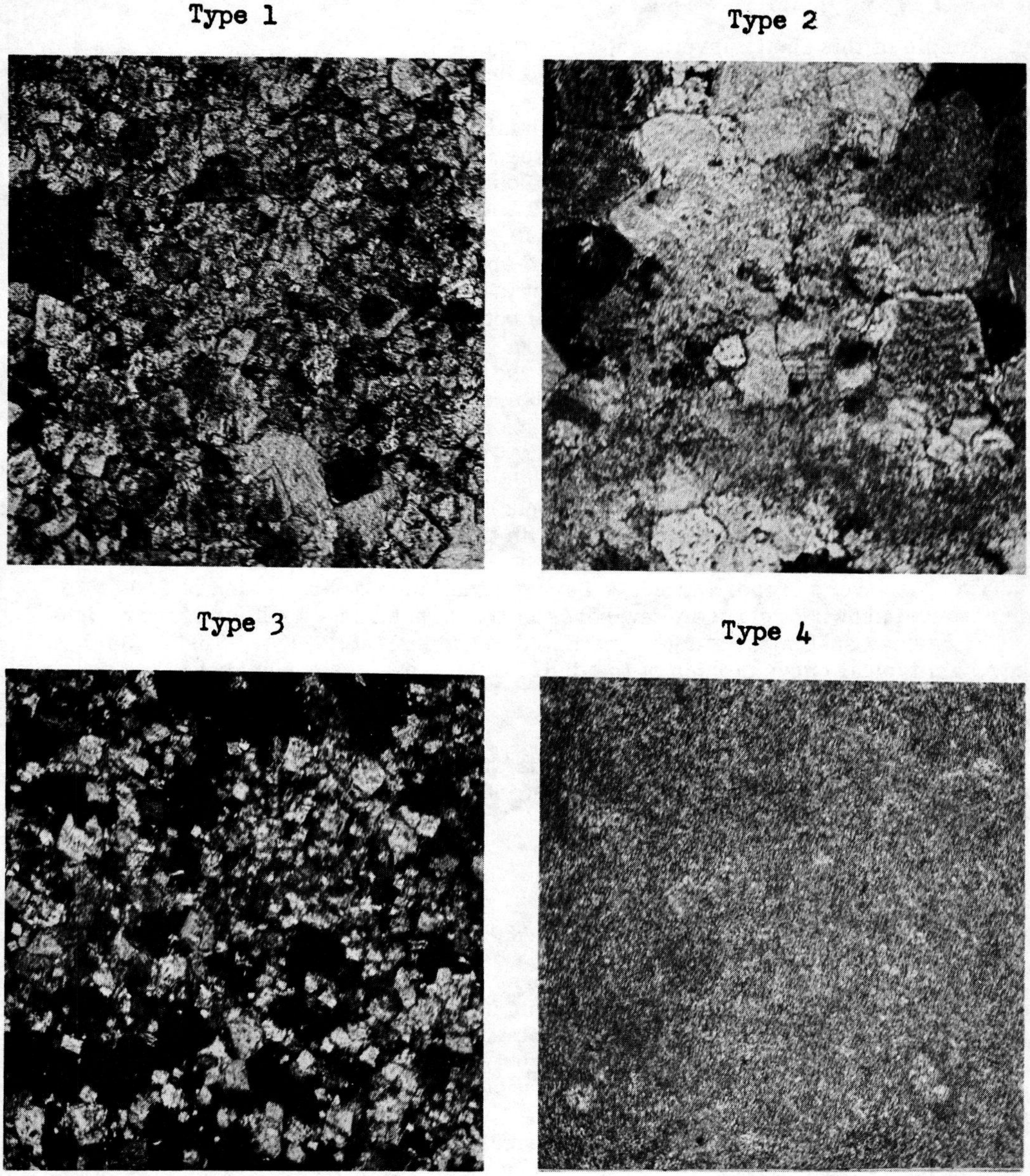


Figure 9. Sketch showing a typical curve of each type distribution.



Type 1

Type 2

Type 3

Type 4

Figure 10. Photomicrographs of typical thin sections representing each of the four categories.

or

$$N = \frac{1}{\int_a^b D(r)\,dr}$$

But

$$\int_a^b D(r)\,dr = V_0 .$$

Therefore, the normalized distribution function becomes

$$d(r) = \frac{p}{rV_0} \frac{d(V_0 - V)}{dp}.$$

An example of this final curve is shown in Figure 8.

The advantage of plotting the normalized function is that in each sample the area under the curve becomes unity and thus the d(r) graphs, for the various samples, can be compared at a glance since the one remaining diagnostic feature is shape.

Type Curves. Each of the normalized distribution curves which has been experimentally arrived at, conveniently falls into one of eight categories. These categories are characterized as follows. Types 1 and 2 are very steeply rising, sharp curves having maxima equal to, or greater than, 10. Curves of type 1 show an increase in d(r) as they progress to the left (that is, to smaller and smaller pores). Contrasted to this, type 2 reaches a maximum at about 0.05 micron and then declines to the left.

Types 3 and 4 have maxima less than 10 but are still relatively sharp curves and each tends toward a maximum as the pore radius becomes smaller and smaller. Type 3 has a significant area under the curve to the right of 2 microns. Type 4 does not.

Types 5 and 6 have maxima less than 10 and possess broader and less well defined peaks. They reach a maximum in the neighborhood of 0.05 to 0.2 micron and then decline as smaller pore sizes are reached. Type 5 is differentiated from 6 by having a significant area under the curve to the right of 2.0 microns.

Type 7 is a broad curve, definitely bimodal in character. It possesses one maximum in the very small pore size range (that is, in the neighborhood of 0.05 micron) and another farther to the right in the neighborhood of 2.0 microns.

Type 8 is a very broad, flat curve having a maximum in the region of 2.0 to 4.0 microns and showing relatively few pores in the radius range of 1.0 micron or less.

The various specimens studied are listed in Tables 1-8. Figure 9 is a sketch showing a typical curve of each of the distribution types described above.

Porosity

Effective Porosity. Effective porosity is that part, expressed in percent, of the bulk volume of a rock which is composed of interconnected voids. Thus, it can be calculated from the following equation:

Effective porosity

$$(\%) = P_e = \frac{\text{effective pore volume}}{\text{bulk volume}} \times 100, \qquad (6)$$

where it is understood that the effective pore volume is the volume of interconnected pores per gram of material and the bulk volume is that volume of mercury which is displaced by one gram of the material in the bulk.

Effective porosities reported in this paper were calculated by the above equation and both effective pore volume and bulk volume were obtained directly from the mercury capillary pressure apparatus.

Total Porosity. Total porosity is that part of the semiporous solid which is composed of inter-connected as well as non-connected voids. It, too, is expressed in percentage and can be calculated by the following formula

$$\text{Total porosity } (\%) = P_t = 100\left(1 - \frac{\gamma_b}{\gamma_p}\right) \qquad (7)$$

where γ_b is the bulk density and γ_p is the powder density.

The total porosities reported in this paper were calculated by the above equation and powder densities were determined by displacement of an acetone-water solution in a volumetric flask according to the method of Rush (20).

Calcium/Magnesium Ratio

Calcium and magnesium were determined according to the method of Cheng et at (4).

This method allows the direct titration of these two constituents, with versenate at appropriate pH levels, without separation. Calcium/magnesium weight ratios were calculated directly from these data.

Insoluble Residue

The method used to determine the insoluble residue is that described by Lemish, et al (10). A 100-g sample of chip-size material was dissolved in 3 N hydrochloric acid, the residue was filtered, dried, and weighed. The percent of insoluble residue was based on the weight of sample originally taken.

Clay Mineral Present

To concentrate and collect clays present in limestone samples a straightforward method was developed by Bisque (1), Rush (20), and Lemish, et al (10). In this procedure, approximately 100 g of powdered limestone with 50 ml of dispersing agent and several hundred ml of H_2O are placed in a drink mixer and mixed for 5 min. This mixture is then transferred to a 1,000-ml graduated flask and diluted to volume. After standing 7 hr, a 50 ml aliquot is taken, at a depth of 6 cm from the surface, and is centrifuged for 15 min. The supernatant liquid is decanted and then about 4 ml of H_2O is added to make a thick slurry. This slurry is placed on a 25-mm by 46-mm glass slide and oven-dried at 60 C. The samples, thus prepared, were subjected to X-ray analysis and the clay mineral present was determined.

The only clay mineral observed in any of the samples was illitic in nature. Data under this heading will indicate either the presence of significant amounts or of trace amounts or the absence of illite. (The term illite was proposed by Grim, Bray, and Bradley (6) as a general term, not as a specific clay-mineral name, for the mica-like clay minerals.)

Freezing-and-Thawing Results

The freezing-and-thawing tests were conducted by the Materials Laboratory of the Iowa State Highway Commission according to the procedure outlined in the Standard Specifications Manual, Iowa State Highway Commission, Ames, Iowa (1956). Samples were prepared by crushing the material in a jaw crusher, to pass a 3/4-in. sieve and subsequently removing all particles passing a No. 4 sieve. A 2,500-g sample was weighed, placed in the appropriate test vessel and evacuated to 1.0 in. of mercury. The sample was then covered with 0.5 percent by weight alcohol-water solution, without breaking the vacuum, and was subsequently frozen for a 2-hr period at -5 to -15 F. It was then thawed for 30 min in liquid of the same composition, which was maintained at a temperature of 65 to 75 F.

Those aggregates having a freezing-and-thawing loss during 16 cycles in excess of 6 percent are considered to be unacceptable for use in highway concrete.

Service Records

Of the five quarries in rocks of Devonian age, only the Glory quarry has a service record which can be evaluated at this time. Rock from that quarry was widely used throughout northeastern Iowa as long ago as thirty years. That length of time has been quite sufficient to prove the unsuitability of Glory stone for concrete highway construction. It is generally agreed to be unacceptable and is used here as a basis for comparison.

Service records for the aggregate from the other four Devonian quarries are indeterminate due to the relatively short time these aggregates have been used in highway construction.

Considering the quarries in Mississippian rocks, the Alden has proved itself excellent in service; the "basal" oolitic Chapin of the LeGrand has had a good service record, but rocks from the overlying Hampton formation are unsatisfactory; and the service record of rocks from the Ferguson quarry is indeterminate.

Textural Studies

Rock specimens from most of the beds for which pore-size distributions were determined were studied both in hand specimens and in thin section. In order that texture could be considered as another parameter, each specimen had to be categorized according to the textural characteristics which it possessed. It was felt essential that the basis for the categories should be of such a nature that the specimens, once categorized, could be re-categorized into those same groups by visual observation at a later date.

Those familiar with the extreme variability of carbonate rock textures will realize the difficulty in accomplishing such a task. The texture which a carbonate rock exhibits is a function of grain size, variability of grain size, the shape of the grains and their arrangement with respect to one another. Texture is a result not only of the environment in which the rock was deposited but also of any diagenetic or metasomatic changes which it experienced.

To categorize some 120 rock specimens into mutually exclusive classes is very difficult if not impossible. Simply due to the nature of rocks, regardless of the categories chosen, there will be some specimens which will exhibit characteristics diagnostic of two, or more, different classes. Being fully aware of these difficulties and of the need for reproducibility, it is believed that some interesting correlations might be forthcoming if the specimens were classified, as well as possible, and relationships sought with other data.

The categories chosen are the following and are, for the most part, based on observation in thin section.

Rocks in category number 1 are characterized by paurograined (that is, 0.1 to 0.01 mm) calcite and dolomite crystals densely, but evenly, distributed throughout and solidly cemented in a clear, crystalline, mesograined (that is, 1.0 to 0.1 mm) matrix of calcite. There are no fossils and relatively few holes.

Rocks of class 2 are characterized by a matrix of clear, crystalline, mesograined calcite with dolomite crystals distributed throughout. This group, however, possesses abundant fossils. Again, these rocks appear dense with few holes.

Rocks of class 3 are characterized by the presence of a great many holes and channels. Rocks of this group are most easily recognized. They have the appearance of having undergone solution which resulted in leaching of the more soluble material and leaving behind a randomly packed mass of the more insoluble crystals. These randomly packed, paurograined crystals are not set in a matrix of calcite. The absence of such a matrix and the presence of abundant holes and open channels are outstanding features of this type.

Category number 4 is reserved for those rocks best described as lithographic. They are micrograined (that is, 0.01 to 0.001 mm) very dense, possess no fossils and no holes.

Photomicrographs of typical thin sections representing each of the four categories are shown in Figure 10. Textural classifications are catalogued in Tables 1-8.

DEFINITION OF FREQUENCY OF OCCURRENCE

To give tentative answers to questions concerning the data collected, frequencies of occurrence were observed. In this paper, frequency of occurrence means nothing more than the fraction formed by placing in the denominator the total number of rocks with a given set of characteristics and in the numerator the number of rocks within the total which meets an additional requirement or additional requirements.

The frequencies of occurrence reported in this paper are based only on those specimens for which data are listed in Tables 1-8.

Experimental Results

The experimental results obtained are listed in Tables 1-8. For the most part, the tables and column headings are self-explanatory. However, the following should be noted.

The specimen numbers are arranged so that if one reads from the bottom to the top of any table he is reading the data obtained for specimens going up the face of the quarry.

The specimens on Table 7 (Ferguson Quarry) are numbered in the reverse order of the other seven. This is inconsequential. In that table, as in all others, up the table is up the quarry face.

The notations in the clay column indicate either the presence of significant amounts, or the presence of trace amounts, or absence of illite

In some instances a single number is used to describe several specimens. This indicates that a particular parameter was determined for those specimens from a composite sample. A short horizontal dash in the tables indicates a parameter that was not determined because of one of several possible reasons.

Frequencies of Occurrence

With a set of data such as that given here, the frequency with which certain characteristics occurred can be observed and tentative answers can be given to questions concerning possible relationships between certain characteristics. A few questions will be considered here in the hope of shedding some light on the problem of what measurable properties affect the suitability of rocks as aggregates for highway construction.

It must be kept in mind that the frequencies of occurrence calculated here apply only to that collection of specimens for which data are presented in the tables.

TABLE 1

CARBONATE ROCK PROPERTIES, GLORY QUARRY

Stratigraphic Units		Sample No.	Insoluble Residue	Clay Mineral Present	Total Porosity	Effective Porosity	Freeze-Thaw % Loss	Pore-Size Distribution	Texture	Ca/Mg Ratio	Miscellaneous
CEDAR VALLEY FORMATION	CORALVILLE	24	2.0	-	2.5	1.2	6.9	6	-	63.0	
		23	50.0	-	19.0	-	-	6	-	-	
		22	1.0	-	2.2	0.3	1.8	8	-	59.0	
		21	8.0	-	2.2	0.9	10.0	6	1	19.5	
		20	2.0	-	1.5	0.3	5.4	8	4	77.7	
		19	12.0	illite	5.0	1.1	20.0	6	1	8.5	
		18	50.0	illite	12.8	-	-	-	-	-	
	RAPID	17	22.0	illite	21.4	-	95.0	-	-	2.5	Unsatisfactory Service Record
		16	16.0	illite	13.9	13.0	68.0	5	1	2.7	
		15	6.0	illite	1.8	0.1	-	8	1	8.7	
		14	16.0	illite	12.2	10.9	27.0	7	1	2.5	
		13	21.0	illite	14.2	11.5	30.0	7	1	2.4	
		12	22.0	illite	11.7	-	-	-	-	-	
		11	12.0	illite	10.4	-	42.0	-	-	3.4	
		10	10.0	-	7.6	5.8	25.0	2	2	3.3	
		9	15.0	illite	9.6	6.4	42.0	1	2	3.8	
		8	39.0	illite	4.8	4.6	29.0	2	2	-	
		7	12.0	illite	7.6	2.8	32.0	1	2	4.8	
		6	19.0	illite	8.0	4.2	33.0	2	2	3.8	
		5	15.0	illite	7.3	3.2	36.0	1	2	4.5	
		4	10.0	illite	5.2	3.4	42.0	1	2	4.0	
		3	22.0	illite	5.2	4.2	30.0	6	2	4.7	
		2	49.0	-	8.1	4.8	27.0	5	2	5.8	
		1	7.0	-	8.3	5.6	11.0	6	2	8.3	

TABLE 2

CARBONATE ROCK PROPERTIES, NEWTON QUARRY

Stratigraphic Units		Sample No.	Insoluble Residue	Clay Mineral Present	Total Porosity	Effective Porosity	Freeze-Thaw % Loss	Pore-Size Distribution	Texture	Ca/Mg Ratio	Miscellaneous
CEDAR VALLEY FORMATION	CORALVILLE	20	1.0	none	3.7	-	19.0	-	-	1.4	
		19	14.0	illite	2.2	0.9	4.7	6	1	1.7	
		18	6.0	none	5.2	3.0	49.0	2	1	7.1	
		17	2.0	none	1.1	-	11.0	-	-	10.0	
		16	10.0	none	8.2	5.0	38.0	4	4	1.8	
		15	2.0	none	6.8	4.4	5.5	1	1	2.3	
		14	14.0	none	-	-	-	-	-	2.0	
		13	6.0	illite	11.5	-	7.5	-	-	1.9	
		12	3.0	none	8.3	-	1.6	-	-	2.3	Concrete Aggregate Ledge Service Record Indeterminate
		11	5.0	none	11.6	10.2	3.2	4	1	2.6	
		10	2.0	none	6.2	-	1.0	-	-	5.5	
		9	2.0	none	10.6	10.5	0.9	7	-	2.5	
		8	4.0	none	11.1	11.0	1.6	4	-	3.3	
		7	5.0	none	7.4	-	-	-	-	11.1	
		6	3.0	none	5.5	5.4	1.9	4	1	6.0	
		5	3.0	none	5.5	5.3	1.5	3	1	6.5	
		4	2.0	none	4.6	4.1	2.8	6	1	3.4	
		3	4.0	none	9.5	-	1.2	-	-	2.6	
		2	13.0	illite	7.9	1.5	31.0	2	1	2.1	
	RAPID	1	10.0	none	6.2	4.3	16.0	6	1	2.1	

TABLE 3

CARBONATE ROCK PROPERTIES, PINE QUARRY

Stratigraphic Units		Sample No.	Insoluble Residue	Clay Mineral Present	Total Porosity	Effective Porosity	Freeze-Thaw % Loss	Pore-Size Distribution	Texture	Ca/Mg Ratio	Miscellaneous
CEDAR VALLEY FORMATION	CORALVILLE	15	1.7	none	9.8	8.4	1.7	5	3	3.2	Concrete Aggregate Ledge Service Record Indeterminate
		14	1.6	trace	10.8	-	3.6	-	-		
		13	1.7	trace	10.2	8.3	1.0	5	1		
		12	3.4	none	13.0	-	1.9	-	-		
		11	2.4	none	8.5	3.8	2.3	4	1		
		10	2.3	none	9.8	6.1	1.7	6	1		
		9	2.7	none	10.9	9.1	1.5	7	2		
		8	1.9	trace	4.7	-	3.7	-	-		
	RAPID	7	9.0	trace	6.2	-	-	-	-	2.0	
		6	0.8	none	3.6	-	2.3	-	-		
		5	18.9	illite	20.1	-	Weathered High Losses	-	-		
		4	16.7	illite	21.2	-		-	-		
		3	22.5	illite	17.8	-		-	-		
		2	17.5	illite	18.2	-		-	-		
		1	15.8	illite	25.4	21.6	58.0	7	3		

Question 1.—Among those specimens that have a pore-size distribution of type 1 or 2, what is the frequency of occurrence (F of O) of specimens having freezing-and-thawing losses in excess of 6 percent?

F of O = 19/31.

This can be interpreted as meaning: if one selects at random from that group of rock specimens listed in the tables, any specimen which has a pore-size distribution of type 1 or 2, then 19 out of 31 times it will exhibit freezing-and-thawing losses which make it unacceptable for concrete aggregate.

Question 2.—Suppose three additional criteria are enforced on those rocks which exhibit distribution curves of type 1 or 2, namely that they have Ca/Mg ≤ 12.0, insoluble residue ≥ 10 percent and texture of type 2 or 3, then what is the frequency of occurrence of specimens having freezing-and-thawing losses equal to or in excess of 6 percent?

F of O = 15/15.

In other words, of the specimens with pore-size distribution curves of type 1 or 2, all of those which also had the other three characteristics mentioned here, also had freezing-and-thawing losses of 6 percent or higher.

TABLE 4

CARBONATE ROCK PROPERTIES, BURTON AVENUE QUARRY

Stratigraphic Units		Sample No.	Insoluble Residue	Clay Mineral Present	Total Porosity	Effective Porosity	Freeze-Thaw % Loss	Pore-Size Distribution	Texture	Ca/Mg Ratio	Miscellaneous
CEDAR VALLEY FORMATION	CORALVILLE	28	8.8	illite	14.0	9.1	2.8	6	1	2.4	
		27	1.6	trace	7.3	1.6	7.1	4	1	62.7	
		26	27.5	illite	16.1	8.2	-	6	1	3.7	
		25	0.7	none	7.5	1.7	2.8	2	4	4.3	
		24	11.2	illite	12.2	10.4	57.0	6	1	2.6	
	RAPID	23	12.7	illite	14.4	12.1	28.0	7	3	2.8	
		22	47.6	illite	10.3	1.7	17.0	1	3	2.0	
		21	17.0	none	21.2	18.0	32.0	5	3	2.0	
		20	20.4	illite	14.6	9.1	75.0	6	3	2.2	
		19	35.0	illite	14.1	6.4	75.0	2	3	2.0	
		18	33.0	illite	14.1	7.1	81.0	2	3	2.4	
		17	34.0	illite	13.9	4.3	51.0	2	3	8.8	
		16	23.0	illite	8.6	3.4	90.0	1	3	2.4	
		15	23.0	illite	10.8	3.3	56.0	1	3	3.0	
		14	24.0	illite	12.2	4.5	56.0	2	3	2.5	
		13	16.0	illite	5.7	0.7	61.0	4	1	12.5	
	SOLON	12	5.2	none	5.1	1.1	2.5	4	2	20.5	Concrete Aggregate Ledge Service Record Indeterminate
		11	7.0	none	5.1	0.6	4.2	3	2	22.1	
		10	4.0	none	7.1	1.3	3.0	2	2	30.4	
		9	2.8	none	9.4	1.6	1.9	1	2	39.6	
		8	4.3	none	5.4	0.4	1.8	3	2	23.6	
		7	2.9	none	6.5	1.2	2.9	1	2	41.5	
		6	12.8	none	9.7	3.3	6.7	6	2	27.8	
		5	2.9	none	5.8	1.7		6	2	31.3	
		4	4.5	none	6.5	2.0		2	1	33.0	
		3	5.5	trace	6.1	1.4	3.8	6	1	12.1	
		2	8.0	trace	7.8	0.7		3	1	9.7	
		1	14.4	illite	9.7	1.9		1	1	8.5	

Question 3.—Consider now a specimen whose service record is known to be unacceptable. Rocks from the Rapid member of the Glory Quarry and from the Hampton formation of the LeGrand fulfill that requirement. What is the frequency of occurrence of specimens belonging to textural type 1 or 2, possessing clay in quantities designated as illite in the charts, having Ca/Mg $\leq$ 9.0, having 3.0 $\leq$ total porosity $\leq$ 23.0, and having 2.5 $\leq$ effective porosity $\leq$ 15.0 among those specimens having unacceptable service record?

F of O = 15/17.

Question 4.—Among those specimens whose service record is poor, what is the frequency of occurrence of specimens whose texture is type 1 or 2 and whose clay content is present in amounts designated at illite?

F of O = 18/18.

Question 5.—It is given that the service record is good (that is, consider the Chapin of the LeGrand and Gilmore City of the Alden). Among those specimens, what is the frequency of occurrence of textures of types 1 or 2?

F of O = 16/17.

Therefore, textural type 1 and 2 do not appear to be diagnostic of poor service record, and there remains the following question.

TABLE 5

CARBONATE ROCK PROPERTIES, RIVER PRODUCTS QUARRY

Stratigraphic Units		Sample No.	Insoluble Residue	Clay Mineral Present	Total Porosity	Effective Porosity	Freeze-Thaw % Loss	Pore-Size Distribution	Texture	Ca/Mg Ratio	Miscellaneous
CEDAR VALLEY FORMATION	CORALVILLE	32	-	none	-	-	-	-	-	-	Concrete Aggregate Ledge Service Record Indeterminate
		31	-	none	-	-	7.9	-	-	-	
		30	1.0	none	4.8	0.7	5.3	5	4	657.0	
		29	1.0	none	8.8	4.0	2.7	6	2	163.0	
		28	3.0	-	6.7	1.8	1.6	5	-	10.4	
		27	1.0	none	6.7	4.4	-	3	-	64.6	
		26	4.0	none	5.4	2.0	-	5	-	212.0	
		25	4.0	none	5.5	2.5	-	3	2	127.0	
		24	2.0	none	7.0	2.0	-	5	1	162.0	
		23	3.0	none	5.5	1.2	4.5	2	2	214.0	
		22	6.0	none	7.5	2.5	4.5	2	-	99.6	
	RAPID	21	5.0	trace	5.5	0.9	53.0	3	2	3.9	
		20	12.0	illite	12.6	6.7	-	6	2	4.1	
		19	11.0	illite	7.0	3.2	31.0	2	2	6.2	
		18	12.0	-	7.5	2.9	42.0	2	2	3.5	
		17	10.0	illite	7.1	4.0	35.0	2	-	4.9	
		16	8.0	trace	3.5	1.1	31.0	7	2	36.3	
		15	5.0	-	4.8	0.8	47.0	5	2	8.4	
		14	13.0	-	5.6	0.9	47.0	5	2	7.4	
		13	8.0	trace	6.4	0.8	47.0	7	2	19.5	
		12	10.0	none	6.0	0.9	-	7	-	13.5	
		11	13.0	illite	5.3	0.8	-	7	2	12.3	
	SOLON	10	12.0	illite	6.0	0.6	68.0	7	2	8.6	
		9	15.0	illite	10.0	0.5	63.0	5	-	7.3	
		8	14.0	illite	7.2	0.5	59.0	5	-	9.0	
		7	10.0	trace	7.0	0.9	50.0	5	2	10.3	
		6	13.0	trace	5.5	0.7	56.0	5	-	15.4	
		5	16.0	trace	6.8	0.4	56.0	7	4	11.1	
		4	12.0	trace	5.0	1.0	46.0	7	2	30.2	
		3	16.0	-	5.5	0.4	61.0	7	4	68.3	
		2	10.0	-	-	-	43.0	-	-	197.0	
		1	15.0	trace	6.0	0.4	3.8	7	-	24.0	

Question 6.—Among those specimens whose clay content = illite, what is the frequency of occurrence of poor service record?

F of O = 21/21.

This result appears to indicate a significant trend.

Question 7.—Among those specimens whose service record is good, what is the frequency of occurrence of clay content = illite?

F of O = 0/20.

The above points out the fact that clay is absent from those beds which are good in service. Returning to Question 3 the next question follows logically.

TABLE 6

CARBONATE ROCK PROPERTIES, LEGRAND QUARRY

Stratigraphic Units	Sample No.	Insoluble Residue	Clay Mineral Present	Total Porosity	Effective Porosity	Freeze-Thaw % Loss	Pore-Size Distribution	Texture	Ca/Mg Ratio	Miscellaneous
HAMPTON FORMATION	7	1.4	illite	22.8	10.5	5.8	6	2	7.8	Service Record Unsatisfactory
	6	6.1	illite	6.3	5.3	4.0	6	2	18.5	
	5	1.6	illite	6.7	5.5	3.8	3	2	2.9	
	4	1.4	illite	22.5	14.3	1.9	7	2	2.1	
	3	1.1	illite	21.5	9.7	3.0	7	2	3.1	
	2	1.3	illite	13.5	9.2	4.3	6	1	4.5	
	1	2.2	illite	10.0	9.7	3.3	7	2	2.2	
CHAPIN FORMATION	Top.	1.5	none	8.5	5.5	3.5	2	-	∞	Service Record Good
	Mid.	1.2	none	12.5	12.5	3.5	6	-	∞	
	Bot.	1.5	none	10.3	7.3	3.5	4	-	∞	

TABLE 7

CARBONATE ROCK PROPERTIES, FERGUSON QUARRY

Stratigraphic Units	Sample No.	Insoluble Residue	Clay Mineral Present	Total Porosity	Effective Porosity	Freeze-Thaw % Loss	Pore-Size Distribution	Texture	Ca/Mg Ratio	Miscellaneous
	1	1.2	illite	10.1	9.4	5.6	5	-	2.0	
	2	1.1	trace	13.3	11.0	3.9	6	-	8.9	
	3	1.5	illite	10.0	15.8	3.5	5	-	7.0	
FORMATION	4	7.7	illite	3.3	1.6	1.0	5	-	2.1	Concrete Aggregate Ledge Service Record Indeterminate
	5	1.8	illite	6.1	6.5	7.7	3	-	2.3	
	6	0.5	illite	5.3	3.6	1.6	1	-	32.6	
	7	3.3	illite	7.0	2.4	1.1	3	-	1.9	
	8	0.3	none	9.5	7.9	1.5	6	-	81.2	
	9	9.6	trace	4.5	4.4	3.4	5	2	2.8	
	10	4.4	illite	8.5	6.8	2.4	5	2	2.7	
	11	16.9	illite	3.5	2.9	8.5	5	2	2.4	
HAMPTON	12	1.5	trace	14.5	7.7	4.6	6	2	∞	
	13	2.9	illite	3.4	3.3	3.8	6	1	2.8	
	14	1.9	illite	5.5	3.0	1.0	4	1	5.2	
	15	2.0	illite	10.3	10.2	1.0	5	2	2.8	
	16	1.1	illite	11.5	10.1	2.5	6	-	1.2	
	17	3.1	illite	15.5	14.5	1.3	8	3	1.4	
	18	2.7	illite	18.2	15.3	3.2	7	-	7.9	
	19	1.5	illite	14.5	13.7	1.2	7	-	4.7	
	20	2.3	illite	14.5	14.1	1.2	8	-	1.9	

TABLE 8

CARBONATE ROCK PROPERTIES, ALDEN QUARRY

Stratigraphic Units	Sample No.	Insoluble Residue	Clay Mineral Present	Total Porosity	Effective Porosity	Freeze-Thaw % Loss	Pore-Size Distribution	Texture	Ca/Mg Ratio	Miscellaneous
GILMORE CITY FORMATION	20	0.2	none	2.8	0.8	2.0	5	4	53.7	Service Record Excellent
	19	0.2	none	6.1	1.2		1	2	42.9	
	18	0.4	trace	8.6	4.3		6	2	26.6	
	17	0.5	trace	11.4	8.2		6	2	49.2	
	16	0.2	none	9.7	6.0		6	2	71.7	
	15	0.3	trace	6.5	2.7		6	2	100.3	
	14	0.1	none	8.6	3.6		6	2	78.4	
	13	0.3	none	11.5	6.9		3	2	64.1	
	12	1.6	trace	12.7	11.4		5	2	87.7	
	11	0.1	none	15.1	12.2		7	2	99.3	
	10	1.3	none	17.3	15.3		7	2	99.5	
	9	0.7	trace	17.3	16.1		5	2	88.4	
	8	0.6	none	17.6	15.2		5	2	70.5	
	7	0.4	none	14.8	11.3		7	2	58.5	
	6	0.1	trace	16.6	10.7		7	2	101.7	
	5	0.1	none	15.5	12.9		7	2	102.1	
	4	0.1	none	8.4	7.8	2.0	5	2	100.7	
	3	-	-	-	-	-	-	-	-	
	2	-	-	-	-	-	-	-	-	
	1	-	-	-	-	-	-	-	-	

Question 8.—Among those specimens whose Ca/Mg $\leq$ 9.0, what is the frequency of occurrence of poor service record?

F of O = 21/21.

Questions 6, 7, and 8.—Indicate that clay content and Ca/Mg ratio are equally good indicators for poor service record.

Question 9.—Among those specimens whose service record is poor, what is the frequency of occurrence of Ca/Mg $\leq$ 9.0?

F of O = 21/22.

Question 10.—Among those specimens whose service record is poor, what is the frequency of occurrence of clay content = illite?

F of O = 21/21.

Thus, it appears that the presence and absence of illite may be diagnostic of unacceptable and acceptable concrete aggregate, respectively.

Question 11.—Among those specimens whose service record is good, what is the frequency of occurrence of specimens which have pore-size distribution curves of type 5 or 6 or 7, have 0 $\leq$ effective porosity $\leq$ 17, have 2 $\leq$ total porosity $\leq$ 18, have Ca/Mg $\geq$ 42, have insoluble residue $\leq$ 1.6, have clay content $\leq$ trace, have freeze-thaw $\leq$ 4.0, have texture = 2?

CONCLUSIONS

It must be kept in mind that the frequencies of occurrence given here are based only on data provided in the section titled "Experimental Results". Extrapolation of any one of these frequencies of occurrence to relate to another group of rocks would not be valid. However, it is felt that the techniques described are applicable and the frequencies of occurrence indicate a direction which would be worthy of further investigation. Furthermore, it is believed that the frequency-of-occurrence method provides a means for other investigators to compare their results with these as well as the results of others.

Rock specimens from two different systems (Devonian and Mississippian) are discussed here. Unfortunately, the service record for only three of these eight can be evaluated. On the basis of this incomplete evidence, Questions 1 and 2 of the preceding section give an idea as to how the frequency-of-occurrence method can be used to describe a potentially poor aggregate. The result of Question 2 indicates that among those specimens which are dolomitic, have an insoluble residue equal to or greater than 10 percent and have a certain texture in addition to possessing a characteristic type distribution of pores, the frequency of occurrence of specimens having poor freezing-and-thawing characteristics is high. It is this type of statement which may be employed to describe a rock whose service record may be unsatisfactory. At present, the decision as to whether or not a rock is acceptable for use as concrete aggregate in highway construction in Iowa is largely based on freezing-and-thawing tests. It can be seen from the tables that specimens with poor service record also failed in the freezing-and-thawing test as long as only the Devonian section was considered. However, Mississippian rocks from the Hampton formation of the LeGrand quarry fail in service but pass freezing-and-thawing tests. The question then naturally arises: What criterion, or set of criteria, occur more frequently with poor service record than does failure in the freezing-and-thawing tests? Questions 3 through 10 were asked and frequencies of occurrence observed in an attempt to get some idea of the answer to that question. The frequencies of occurrence which are the answers to Questions 6, 7, and 10 show that the presence and absence of illite were observed with poor and good service record, respectively.

Why is it that specimens from the Hampton formation of the LeGrand fail in service and yet pass freezing-and-thawing tests? Perhaps a clue to the answer of this question can be learned by reference to Question 2 again. Since any specimen from the Hampton formation of the LeGrand quarry fails to fulfill the four given requirements, one might expect it to pass freezing-and-thawing tests and it does.

Pore-size distribution alone does not appear to be related closely with either freezing-and-thawing or service record when both Devonian and Mississippian rocks are considered. On the other hand, combined with certain other variables, it does appear to be a factor in determining freezing-and-thawing characteristics. In Question 11 pore-size distribution, along with seven other characteristics, is related to good service record. Considering that nine variables are being related simultaneously, a F or O = 13/19 appears interesting. Perhaps a more detailed study and careful analysis of pore-size distribution would lead to a closer relationship between distribution curve type and service record.

It might be postulated that pore-size distribution and texture are somehow related. That relationship is not obvious, however, from the work done to date. Perhaps a textural classification other than the one employed here would make such a correlation apparent. In any case, further sampling and investigation both with the mercury injection apparatus and with the microscope is to be conducted in an attempt to prove or disprove that postulate.

ACKNOWLEDGMENTS

The authors wish to express appreciation to those persons who contributed to the completion of this work.

The project was financed by the Iowa Highway Research Board, with funds supplied by the Iowa State Highway Commission.

The late Bert Myers, Head of Materials Department, Iowa State Highway Commission, took a special interest in the project since he had long been concerned with the aggregate problem. Others of the Highway Commission, Mark Morris, Director of Research; Theodore Welp, Chief Geologist; Howard Dixon, former geologist; and Arthur Myhre, District Materials Engineer; provided valuable information, and also made it possible to have many tests performed in their laboratories.

C.J. Roy and staff of the Geology Department, Iowa State College, are thanked for their interest and cooperation.

Richard Handy, geologist for the Engineering Experiment Station, granted permission to use the Station's X-ray equipment.

REFERENCES

1. Bisque, R.E., "Limestone Aggregate as a Possible Source of Chemically Reactive Substances in Concrete." Unpublished M.S. Thesis, Iowa State College Library, Ames, Iowa (1958).
2. Bisque, R.E. and Lemish, J., "Chemical Characteristics of Some Carbonate Aggregates as Related to the Durability of Concrete." HRB Bull. 196, (1958).
3. Blanks, R.G., "Modern Concepts Applied to Concrete Aggregates." Am. .Soc. Civil Engrs., Proc., Vol. 75, pp. 441-446 (1949).
4. Cheng, K.L, Kurtz, T., and Bray, R.H., "Determination of Calcium, Magnesium and Iron in Limestone." Anal. Chem., Vol. 24, pp. 1640-1641 (1952).
5. Dorheim, F.P., "Petrography of Selected Limestone Aggregates." Unpublished M.S. Thesis, Iowa State College Library, Ames, Iowa (1950).
6. Grim, R.E., Bray, R.H., and Bradley, W.F., "The Mica in Argillaceous Sediments." Am. Mineral., Vol. 22, pp. 813-829 (1937).
7. Hiltrop, C.L., "Relation of Pore-Size Distribution to the Petrography of Some Carbonate Rocks." Unpublished M.S. Thesis, Iowa State College Library, Ames, Iowa (1958).
8. Iowa State Highway Commission, "Standard Specifications for Construction on Primary Farm to Market and Secondary Roads." Iowa State Highway Commission, Ames, Iowa (1956).
9. Laughlin, G.F., "Usefulness of Petrology in Selection of Limestone." Rock Products, March 17, pp. 50-59 (1928).
10. Lemish, J., Rush, E. and Hiltrop, C.L., "Relationship of the Physical Properties of Some Iowa Carbonate Aggregate to the Durability of Concrete." HRB Bull. 196 (1958).
11. Lewis, D.W., and Dolch, W.L., "Porosity and Absorption." ASTM Spec. Tech. Pub. No. 169, pp. 303-313 (1955).
12. Mather, B., Callan, E.J., Mather, K., and Dodge, N.B., "Laboratory Investigation of Certain Limestone Aggregates for Concrete." Tech. Memorandum No. 6-371, Waterways Experiment Station, Corps of Engineers, U.S. Army (1953).
13. Mather, K., "Crushed Limestone Aggregates for Concrete." Transactions AIME, TP 3616H, Mining Engineering, pp. 1022-1028 (Oct. 1953).
14. McBain, J.W., "Sorption of Gases and Vapours by Solids." George Routledge and Sons, London (1932).
15. Mielenz, R.C., Greene, K.T., and Benton, E.J., "Chemical Test for Reactivity of Aggregates with Cement Alkalies, Chemical Processes in Cement Aggregate Reaction." Jour. Am. Concrete Inst., Vol. 19, pp. 193-221, (1947).
16. Purcell, W.R., "Capillary Pressures—Their Measurement Using Mercury and the Calculation of Permeability Therefrom." Trans. AIME Vol. 186, pp. 39-48 (1949).
17. Rhoades, R., and Mielenz, R.C., "Petrographic and Mineralogic Characteristics of Aggregates." Am. Soc. Test. Mats. Symposium on Mineral Aggregates, pp. 20-47 (1948).
18. Ritter, H.L., and Drake, L.C., "Pore Size Distribution in Porous Materials." Ind. Eng. Chem., Anal. Ed., Vol. 17, pp. 782-786 (1945).
19. Roy, C.J., Thomas, L.A., Weissmann, R.C., and Schneider, R.C., "Geologic Factors Related to Quality of Limestone Aggregates." HRB Proc., Vol. 34, pp. 400-415 (1955).
20. Rush, F.E., "Petrography and Physical Properties of Some Devonian Limestones of Iowa." Unpublished M.S. Thesis, Iowa State College Library, Ames, Iowa (1957).
21. Sweet, H.S., "Research on Concrete Durability as Affected by Coarse Aggregate." ASTM, PROC, Vol. 48, pp. 988-1016 (1948).

22. Washburn, E.W., "Note on a Method of Determining the Distribution of Pore Sizes in a Porous Material." Nat. Acad. Sci., Proc., Vol. 7, pp. 115-116, (1921).