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PETROLOGY OF DOLOMITIC LIMESTONES, KINGSTON, ONTARIO, CANADA

By J. E. GILLOTT

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Petrology of Dolomitic Limestones, Kingston, Ontario, Canada

Abstract: Dolomitic limestone from Kingston, Ontario, has importance as a type example of rock which, when used in concrete, produces an alkali-aggregate reaction different from reported alkali-silica reaction. The expansive reaction causes deterioration and cracking of the concrete. A petrographic, mineralogical, and chemical comparison was made between rock which showed high expansion in concrete and that which displayed no reactivity, and also between the same material before and after use in concrete (referred to throughout as "rock" and "aggregate," respectively).

The reactive rocks are fine-grained dolomitic limestones or calcitic dolomites that contain about 10 per cent of acid-insoluble material. The groundmass is composed of fine calcite, which shows little evidence of grain-growth or recrystallization, together with larger crystals of clear sparry calcite which appear to be original. The rock has been extensively dolomitized. Several techniques were employed to make concentrates of the minerals present. These were identified microscopically and examined by X-ray diffraction and differential thermal analysis.

The composition of the carbonates was deter-

mined in terms of mole percentage Ca-Mg by relating lattice constants to composition. The clay minerals are described and minerals present in various residues identified.

The total solid and acid-insoluble fractions were chemically analyzed. To facilitate comparison the oxides were recombined by computation into minerals to which were arbitrarily assigned the same composition. An attempt to relate oxide or computed mineral content to reactivity was unsuccessful.

Rocks that are similar petrographically expand to markedly different degrees. The reason for this is unknown but petrographic character, distribution of clay minerals, dolomite composition, and dolomite-calcite ratio may well be connected interdependently with the cause of reactivity. Although it is not possible on petrographic criteria alone to predict the degree of reactivity which a particular rock may show if used as aggregate, it is possible to recognize rocks of this petrographic type. Such rocks should be subjected to more detailed examination, such as the concrete prism test, before being used as aggregate with high-alkali cement.

CONTENTS

Introduction	760
Acknowledgments	760
Reactivity	760
Treatment of samples	761
Petrography	762
Mineralogy	763
General statement	763
Carbonates	763
Clay minerals	765
Residues	769
Chemistry	770
General statement	770
Computational procedure	770
Discussion of chemical results	771
Conclusions	773
References cited	777

Figure

1. Comparison of linear expansion of concrete beams containing samples of Kingston aggregate 760
2. Expansion of Kingston rock prisms in alkaline solution 761
3. X-ray diffractometer patterns of Kingston 6- to 7-foot rock, acid-insoluble fraction . . . 766

4. X-ray diffractometer patterns of Kingston 24- to 30-foot rock, acid-insoluble fraction . . 767
5. X-ray diffractometer patterns of Ottawa Valley rock, acid-insoluble fraction 768
6. Thermograms of acid-insoluble fractions . . . 769
7. Oxide content of acid-insoluble fraction of rock versus expansion of concrete beams . . . 772
8. Silica content of acid-insoluble fraction of rock versus expansion of concrete beams . . . 773
9. Computed mineral composition expressed as total carbonate 774
10. Computed mineral-composition variation in acid-insoluble fraction 775

Plate Facing

1. Photomicrographs of thin sections of reactive rocks, Kingston, Ontario, Canada 764
2. Photomicrographs of thin sections of unreactive rocks, Ottawa Valley, Ontario, Canada . . . 765

Table

1. Mole percentage Ca-Mg in dolomite and calcite determined by X-ray analysis 764
2. $\text{SiO}_2/\text{Al}_2\text{O}_3$ in acid insoluble 774
3. $\text{K}_2\text{O}/\text{H}_2\text{O}$ in acid insoluble 774
4. Classification of rocks and aggregates 776

INTRODUCTION

Argillaceous dolomitic limestone from Kingston, Ontario, shows an expansive reactivity when used in concrete as coarse aggregate with high-alkali cement. This expansion results in cracking and general deterioration of concrete (Swenson, 1957; Swenson and Gillott, 1960; Swenson and Legget, 1960).

Some of the most expansive rock in the Kingston area is assigned to the Pamela Formation of the Ordovician system (I. D. Maycock, 1959, M.S. thesis, Queen's University, Kingston, p. 208). In early work no attempt was made to restrict the source of aggregate to specific beds; it was made from a series of beds in the top 24 feet of a quarry near the city of Kingston. This aggregate produced high expansion in concrete and it was thought probable that this reflected the combined effect of both more and less expansive rocks, as petrographic examination showed rims surrounding only some of the aggregate particles enclosed in concrete. It was evident that restriction of the source of aggregate to individual beds would enhance the possibility of correlation between expansive behavior and texture, mineralogy, or chemistry of the rock. At this particular quarry fairly well-defined beds occur at depths of 6-7, 10½-12, 20-21, and 24-30 feet, the last mentioned weathering a characteristic green.

This paper describes the petrography, mineralogy, and chemistry of these beds and the differing degrees of reactivity they display when used individually as coarse aggregate in concrete. For comparison a similar description is included of a nonexpansive limestone from a source in the Ottawa River Valley near Ottawa.

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The author is particularly indebted to E. G. Swenson who supervised the fabrication of the concrete beams and the subsequent measurements of length changes on these beams. Thanks are also due to Drs. S. A. Forman and J. E. Brydon and the other staff members of the clay mineralogy group of the Canadian Department of Agriculture, and to Dr. L. D. Calvert, Division of Applied Chemistry, National Research Council, Canada. The author wishes to thank Prof. J. R. Goldsmith, Associate Dean of the Division of Physical Sciences, University of Chicago, for his valuable assistance, and Drs. J. A. Maxwell and B. A. Liberty of the

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REACTIVITY

For each type of aggregate two pairs of concrete beams measuring 3 by 4 by 16 in. were fabricated. High-alkali cement was used in one pair and low-alkali cement in the other. The

Figure 1. Comparison of linear expansion of concrete beams containing samples of Kingston aggregate

high-alkali cement was similar to that previously reported (Swenson and Gillott, 1960, p. 21), and the low-alkali cement contained 0.18 per cent sodium oxide and 0.28 per cent potassium oxide, giving total alkali content computed as Na_2O to be 0.36 per cent. An additional pair of high-alkali beams was made in which the 10½- to 12-foot bed was ground and used as sand fraction. These concrete beams were continuously conditioned at nearly 100 per cent relative humidity and 73°F., and length-change was recorded by measurement as described by Swenson and Gillott (1960, p. 21). The rate and degree of linear expansion are shown in Figure 1. The 20- to 21-foot bed produced by far the highest rate of expansion and indeed the highest rate of any of the rocks from the two main quarries in the area. The 10½- to 12-foot bed produced expansion at a

slower rate, which was approximately the same as that produced by the composite 0- to 24-foot series. The rate of expansion decreased in the order: 6- to 7-foot bed, 24- to 30-foot bed, and Ottawa Valley limestone, which had little reactivity. There was no apparent increase in

at a rate similar to a previously reported sample of this rock (Swenson and Gillott, 1960, p. 30). This suggests that whatever causes the expansive reactivity varies not only from bed to bed but also within different parts of the same bed.

Figure 2. Expansion of Kingston rock prisms in alkaline solution

expansion from the use of the 10½- to 12-foot bed as sand-size fraction. None of the beams made with low-alkali cement expanded deleteriously.

Prisms measuring 1½ by 5¾ in. were cut from the rock and placed in alkaline solutions made up to simulate those likely to be present in high-alkali cement (Swenson and Gillott, 1960, p. 28). The prism cut from the 20- to 21-foot bed, which produced the highest expansion when used as aggregate in concrete, expanded at a slower rate than the prism cut from the 6- to 7-foot bed (Fig. 2); this expanded

TREATMENT OF SAMPLES

The concrete beams were removed from test after several months under conditions of 100 per cent relative humidity and 73°F., when linear measurements on the beams indicated that the expansive reaction had occurred in the aggregate.

Petrographic examination was carried out on hand specimens, thin sections of the rocks, the rocks after they had soaked in alkali, and on the concrete.

Both rock and aggregate particles separated

from concrete were analyzed mineralogically and chemically in order to ascertain whether the expansive reaction led to changes in the composition of the rock. Dedolomitization was the major change observed when these rocks were treated with alkali (Hadley, 1961, p. 468; Swenson and Gillott, 1960, p. 26).

On smoothed surfaces of concrete made with composite aggregate, rims were observed surrounding some of the aggregate; these were removed with a dentist's drill. Cores of rimmed aggregate and nonrimmed aggregate were separated in a similar way, but the process was tedious and gave only small yields. In later work when aggregate was made from restricted beds of rock, the concrete was broken up and the aggregate separated from the adhering cement paste by hammer and chisel. The final clean-up was accomplished with a dentist's drill; each piece of aggregate was held in a vise and treated separately. This process was also lengthy and only a few grams of aggregate were obtained from which it could be certain that cement paste had been completely removed. The sampling, therefore, was probably less representative than that for the rock. From this point, both rock and aggregate were treated in a similar manner. Concentrates were made of the dolomite, the acid-insoluble fraction, the residue heavy in liquid of specific gravity 3.0, and the residue from the acid-insoluble fraction which was light in a liquid of specific gravity 2.4. Fractions separated by means of a Frantz isodynamic magnetic separator were also examined.

The dolomite was concentrated in five stages. Rock was carefully ground by hand with repeated sieving through a battery of sieves to prevent overgrinding. Much of the clay material passed into the fines and was discarded. The material retained on 325 mesh was fractionated by sink-float in heavy liquids, the dolomite being relatively concentrated in the fraction, which was heavy in liquid of specific gravity 2.77 and light in liquid of specific gravity 3.0. This residue was dried and passed through a Frantz isodynamic magnetic separator. The nonmagnetic fraction was stirred for $\frac{1}{2}$ hour in 4-molar ammonium acetate at 70°C; this led to a further enrichment in dolomite as this is less soluble than calcite, some of which had passed through the other separation procedures. The dolomite concentrate was then sufficiently pure for analysis by X-ray diffraction. The dolomite required for chemical

analysis was further purified by dispersion of the clay minerals in Calgon with repeated sedimentation of the carbonate. It was found beneficial to repeat each step of the procedure for best results.

Clay minerals were concentrated for X-ray examination by two procedures. In the early stages of the work the technique adopted was a modification of that described by Ray, Gault, and Dodd (1957, p. 683) in which Rohm and Haas commercial resin IRC-50 was employed. In the later stages of the X-ray work and for the material analyzed chemically, the acid-insoluble fraction was separated by stirring the rock for 16 hours in cold 4-molar acetic acid. The residue was washed with distilled water and concentrated by centrifuging. In some instances the clay minerals were converted to a calcium system by flocculation with calcium chloride.

PETROGRAPHY

In hand specimens the 6- to 7-foot, 10½- to 12-foot, and 20- to 21-foot beds are medium gray and show thin impersistent bedding on the millimeter or centimeter scale. There is evidence of small-scale disturbance and current action as beds are broken into lenses, and a few overlying beds erode and truncate underlying disturbed layers. The 24- to 30-foot bed is massive and greenish but shows faint lighter clots on polished surfaces. Ottawa Valley limestone is a dark rock with graphitic material coating some of the fracture surfaces. There are veinlets and vugs of clear calcite, and fossil fragments are common.

In thin section the 6- to 7-foot, 10½- to 12-foot and 20- to 21-foot beds have similar features, although there is probably rather less dolomite at the 20- to 21-foot level. The dolomite occurs as euhedra of about 0.05-mm cross section and is sometimes zoned by inclusions (Pl. 1, fig. 1). The matrix in all three beds consists of fine cryptocrystalline or microcrystalline, dusky-brown calcite, which occurs in clot-like aggregates shot through by veinlets of dolomite. The undolomitized clots, of the order of 1/5–1/10 mm across, are in many cases a palimpsest fabric, as anastomosing off-shoots from clots can be followed meandering among dolomite euhedra. On the other hand there are aggregations of fine calcite that are quite evidently a sedimentary feature of the rock and existed prior to dolomitization. These are the "intraclasts" of Folk's terminology (Folk,

1959, p. 4) and are most clearly seen in the 10½- to 12-foot bed, where thin bands of "intrasparrite" are interbedded with murky finer-grained layers with evenly distributed dolomite. The clear sparry calcite of the intrasparritic layers shows the plane grain boundaries, which according to Bathurst (1958, p. 16) are characteristic of granular cement, and the distribution of the fragments or intraclasts is consistent with mutual, three-dimensional contact. Many of the intraclasts are speckled with black graphitic material and some have scattered dolomite euhedra. These are more concentrated in the surrounding matrix (Pl. 1, fig. 2) however, and apparently the intraclasts, although fine-grained, have been less susceptible to dolomitization than the fine-grained material of the darker layers with which the intrasparrite is interbedded. Within the dark layers are a few sack-shaped areas of the order of 1 mm across that consist of 0.05-mm pellets and spar and which may have been caused by boring organisms, the pellets possibly being faecal.

There are 1- to 2-mm irregular rhombs or rectangles visible in hand specimens in the 10½- to 12-foot bed; these apparently are single crystals of calcite. In section, however, they are found to consist of a calcite mosaic with some dolomite euhedra. It is thought that they may be pseudomorphs after a cubic material such as sodium chloride.

The 24- to 30-foot bed, when examined in thin section, is seen to be a silty argillaceous dolomite. The section is finely sprinkled with angular detrital quartz grains about 0.1 mm in diameter, although rounded grains about 1 mm in diameter also occur. Dolomite euhedra range downward in size from about 0.2 mm. Dusky argillaceous material is of rather irregular distribution and patches occur of purer carbonate.

The Ottawa Valley limestone employed as aggregate was not restricted to a single horizon, and when thin sections of the concrete were examined the rock fragments showed variation in petrographic character. In contrast with the Kingston samples, however, most of the Ottawa Valley material has a higher content of skeletal material and a smaller amount of dolomite. Nonetheless, the presence of more dolomitic beds is indicated, as at least one piece of aggregate contains a fairly high concentration of evenly distributed, well-formed dolomite euhedra of about 0.1-mm cross section. In another fragment the dolomite rhombs are preferentially concentrated, in many cases marginally,

in rounded or elliptical objects possibly of organic origin (Pl. 2, fig. 1). If so, the dolomite may result from the unmixing of magnesian calcite. Such a process has been suggested by Chave (1954, p. 596) who also discusses the occurrence and distribution of magnesian calcite in the shells of marine organisms. In some cases the magnesium occurs in a concentration which is metastable at normal temperatures (Goldsmith and others, 1955, p. 215), and an approach to equilibrium through recrystallization seems likely in the course of geological time. An alternative, though apparently less likely explanation, is that the objects are calcarenite grains which have been preferentially dolomitized.

Irregular, silt-size quartz grains vary in abundance among different fragments of aggregate. In some of the aggregate oölites occur and show both radial and concentric structures. In some cases, the core is composed of fine structureless calcite. In others, the outer rim of the oölit is embayed, which suggests recrystallization involving both matrix and oölit (Pl. 2, fig. 2). A consideration of fabric criteria (Bathurst, 1958, p. 18-20) indicates, however, that much of the abundant clear sparry calcite was formed as an original product of crystallization either as pore-filling cement or drusy mosaic.

MINERALOGY

General Statement

The residues and concentrates separated from the rock and aggregate were examined by microscopic, X-ray diffraction, and differential thermal methods of analysis.

Carbonates

Structural and compositional variation among the rhombohedral carbonates has been discussed by Goldsmith and Graf (1948a; 1958b). Calcite may contain magnesium in solid solution in the lattice, and dolomites, metastable at normal temperatures, occur which hold an excess of CaCO_3 above the ideal 50-mole per cent. This leads to three changes which may be detected by X-ray diffraction: an increase in the size of the unit cell, a weakening of the three-order reflections, and diffuseness in reflections with a strong c-axis component. Composition and lattice constants are connected by a straight-line relationship between dolomite and calcite (Goldsmith and Graf, 1958a, p. 680) and between magnesite and calcite (J. R. Gold-

smith, 1961, personal communication). Using the published values of a_0 and c_0 for hexagonal axes and the relationship

$$\frac{1}{d^2} = \frac{4}{3a^2}(h^2 + hk + k^2) + \frac{l^2}{c^2},$$

curves were constructed using the derived d -values as ordinate and per cent composition as abscissa. Indices for the lines used were taken from Howie and Broadhurst (1958, p. 1212) and the A.S.T.M. Powder Data File (American Society for Testing and Materials).

obtained from these samples generally showed strong lines due to dolomite in addition to those due to calcite. The dolomite compositions were in these cases derived both from the pattern obtained from the dolomite concentrate and also by measurement of the film taken primarily for the calcite compositions. Values of composition in terms of mole per cent are shown in Table 1 with confidence limits calculated for a probability of 95 per cent.

The results show that all the dolomite contains excess calcium, but this is at a minimum in

TABLE 1. MOLE PERCENTAGE Ca-Mg IN DOLOMITE AND CALCITE DETERMINED BY X-RAY ANALYSIS

Sample	Rock	Aggregate
CALCITE RESULTS*		
Kingston 6-7-ft. bed	Ca _{99.53} Mg _{0.47} ± 0.21	Ca _{99.67} Mg _{0.33} ± 0.35
Kingston 10½-12-ft. bed	Ca _{97.90} Mg _{2.10} ± 0.71	Ca _{98.34} Mg _{1.66} ± 0.52
Kingston 20-21-ft. bed	Ca _{96.17} Mg _{3.83} ± 0.94	Ca _{97.95} Mg _{2.05} ± 0.60
Kingston 24-30-ft. bed	Ca _{99.20} Mg _{0.80} ± 0.97	Ca _{98.61} Mg _{1.39} ± 1.31
Ottawa Valley limestone	Ca _{99.03} Mg _{0.97} ± 0.32	Ca _{97.87} Mg _{2.13} ± 0.80
DOLOMITE RESULTS*		
Kingston 6-7-ft. bed	Ca _{55.6} Mg _{44.4} ± 0.72	Ca _{56.68} Mg _{43.32} ± 0.60
Kingston 10½-12-ft. bed	Ca _{55.51} Mg _{44.49} ± 0.77	Ca _{56.34} Mg _{43.66} ± 0.58
Kingston 20-21-ft. bed	Ca _{55.14} Mg _{44.86} ± 0.65	Ca _{56.48} Mg _{43.52} ± 0.70
Kingston 24-30-ft. bed	Ca _{55.37} Mg _{44.63} ± 0.33	Ca _{54.85} Mg _{45.15} ± 0.73
Ottawa Valley limestone	Ca _{53.47} Mg _{46.53} ± 0.43	Ca _{54.80} Mg _{45.20} ± 0.73

* Precision expressed at the 95 per cent confidence level

The X-ray diffraction patterns of the dolomite concentrates from the Kingston rocks and Ottawa Valley limestone were recorded photographically with a Debye Scherrer powder camera and a Nonius Guinier quadruple-focusing camera. Copper radiation, in the latter case monochromatized by means of a bent quartz crystal, was employed. The films taken with the Guinier camera were used for measurement of d -value which was obtained from the average of three readings on each line. In some instances values of the lattice constants a_0 and c_0 were also derived. The dolomite and calcite lines measured were within the angular range from $\theta = 14^\circ$ – 25° , in which the focussing geometry of the camera leads to coincidence of the $K\alpha_1$ and $K\alpha_2$ components of the copper radiation. In one panel on every film, As_2O_3 was used as standard to correct for length changes in the film due to processing. Calcite lines were either very weak or absent on the films obtained from the dolomite concentrates, but, although an attempt was made to concentrate calcite by means of heavy liquids, the patterns

the Ottawa Valley sample which is therefore less imperfect than those from Kingston. These show some Ca-Mg disorder in addition to "disturbed layer sequences normal to the c -axis" (J. R. Goldsmith, 1961 personal communication).

The incorporation of considerable quantities of Fe or Mn in the place of Mg in the dolomite lattice could produce effects on the diffraction pattern similar to an excess of Ca (Goldsmith and Graf, 1958a, p. 678). For the Kingston 6- to 7-foot, 10½- to 12-foot, and 24- to 30-foot beds, therefore, a more careful separation was carried out; it gave small concentrates of purer dolomite in a quantity sufficient for partial chemical analysis. The results showed little or no Mn and only just over 1 per cent total iron. This is probably due to inclusions of clay within the crystals (Pl. 1, fig. 1). In these cases, therefore, the increase in lattice spacings above those for ideal dolomite is not due to substitution in the lattice of Mn or Fe for Mg, and it seems reasonable to assume excess Ca to be responsible in all cases.

Figure 1. Photomicrograph of Kingston 20- to 21-foot bed showing arrangement of inclusions in euhedral crystal of dolomite. Vickers projection microscope: 8-mm objective N.A. 0.5; 4× eyepiece; ×220

Figure 2. Photomicrograph of Kingston 10½- to 12-foot bed showing cal-cilutite fragments in dolomitized matrix of calcite. Vickers projection microscope: 16-mm objective N.A. 0.28; no eyepiece; ×65

PHOTOMICROGRAPHS OF THIN SECTIONS OF
REACTIVE ROCKS, KINGSTON, ONTARIO, CANADA

Figure 1. Photomicrograph of Ottawa Valley limestone showing organic material (algae?) with preferentially concentrated dolomite euhedra. Vickers projection microscope: 16-mm objective N.A. 0.28; no eyepiece; $\times 65$

Figure 2. Photomicrograph of Ottawa Valley limestone with oölite showing embayment due to recrystallization or grain-growth of calcite crystals. Vickers projection microscope: 8-mm objective N.A. 0.5; 4 $\times$ eyepiece; $\times 220$

PHOTOMICROGRAPHS OF THIN SECTIONS OF
UNREACTIVE ROCKS, OTTAWA VALLEY, ONTARIO, CANADA

Graphical recordings of the angular region, which includes the three dolomite reflections (11.0), (01.5), and (00.6), were made by means of a microdensitometer from the films taken with the Guinier camera. The higher order (00.12) was generally too diffuse to register a pattern. The (11.0) reflection, from planes parallel to the *c*-axis, is more intense than the (00.6) reflection from planes perpendicular to *c*. The order reflection (01.5) is also weak and in many cases broadened.

The lattice spacings of the calcite are generally somewhat less than those of the pure mineral which indicates the inclusion of small amounts of ions such as Mg with a smaller ionic radius than Ca.

Clay Minerals

An initial interest was focussed on the clay minerals in the Kingston rocks as the hypothesis was considered that the expansion in alkali might be due to the presence or formation of expanding lattice minerals (Caillère and others, 1949, p. 1742; White, 1950, p. 130). Some minerals of the montmorillonite type, *i.e.*, hectorite, are known to be acid-soluble, and experiments were performed to determine the minimum quantity of hectorite in powdered rock that could be detected after removal of the carbonates. It was found that a 1 per cent addition could be detected on glycolation of oriented slides examined by means of the X-ray diffractometer when the carbonates were removed by the use of ion-exchange resin. By this means separations were made from the rock, from the rock after treatment in alkaline solution (Swenson and Gillott, 1960, p. 28), and from the aggregate. In the last case, rims, cores and nonrimmed aggregate were treated separately. In all cases the dominant clay mineral found was illite, together with lesser amounts of chlorite, but no swelling clay minerals were detected.

In later work the clay minerals were separated from the carbonates by means of cold 4-molar acetic acid. A typical diffractogram recorded from an oriented sample of the acid-insoluble fraction of the reactive Kingston 6- to 7-foot bed is shown in Figure 3 and diffractograms recorded from similar samples of the Kingston 24- to 30-foot bed and the Ottawa Valley limestone are shown in Figures 4 and 5. The tailed form of the 10Å-illite peak in the air-dried condition suggests that this mineral is more hydrated in the Ottawa Valley rocks than in the Kingston samples.

In addition to diffractograms of oriented samples, Debye Scherrer powder photographs were taken in all cases. Measurement of the *d*-value of the (060) reflection showed that the illite belongs to the dioctahedral series (Brindley, 1951, p. 160).

A consideration of the other reflections on the powder patterns indicated that the illite has some of the characteristics of both 1M and 2M muscovite. A reflection with a *d*-value of about 2.7Å was taken as evidence in favor of a 1M-type structure (Smith and Yoder, 1956, p. 230; Yoder and Eugster, 1955, p. 243), but two other strong lines at 3.07Å and 3.62Å that characterize this polymorph were either very weak or absent. It is considered that this indicates disorder (Levinson, 1955, p. 47), but in these samples there were more lines present than characterize the 1Md polymorph. Most of the lines could be matched with the 2M pattern, and in particular quite strong lines were visible at about 3.48Å and 3.2Å that should be absent on a 1M pattern. In a 2M muscovite, however, powder lines should occur at 2.98Å, 2.86Å, and 2.78Å (Grim, 1953, p. 95) but they were absent on films of the acid-insoluble fraction of the Kingston 6- to 7-foot and 10½- to 12-foot beds. In the samples from the Ottawa Valley and Kingston 20- to 21-foot and 24- to 30-foot beds a very weak line did occur at 3.0Å. A quite strong line on the 2M pattern at 3.889Å (Yoder and Eugster, 1955, p. 247) was also absent in these samples. The Ottawa Valley material also differed somewhat from the Kingston samples in that there was not the same distribution of intensity between the lines.

A perfect match could not be made between *d*-values of these samples and published data for either 1M or 2M polymorphic forms of muscovite. It is concluded that the "illite" is composed of a mixture of both polymorphs which are to some extent disordered.

The illite is, in many cases, intergrown in variable proportions with organic material, and due to the consequent variation in physical properties it appeared in different fractions during the separations. The purest material was in the fraction light in liquid of specific gravity 2.4, and thermograms of the Kingston samples at the 6- to 7-foot and 10½- to 12-foot levels and Ottawa Valley sample are shown in Figure 6. The peaks due to illite are most clearly seen on the samples run in vacuum as then the exothermic peaks of the organic material are largely suppressed. The two Kingston samples

Figure 3. X-ray diffractometer patterns of Kingston 6- to 7-foot rock, acid-insoluble fraction. (a) Air dried; (b) treated with glycol; (c) heated 550°C 1 hour. Setting of North American Philips (Norelco) Diffractometer: counts full scale, 3000; time constant, medium; scanning rate, 1°/min; divergence slit, 1°; scatter slit, 1°; receiving slit, .003; X-radiation, Co; filter, Fe; KV, 30; mA, 15

Figure 4. X-ray diffractometer patterns of Kingston 24- to 30-foot rock, acid-insoluble fraction. (a) Air dried; (b) treated with glycol; (c) heated 550°C 1 hour. Setting of North American Philips (Norelco) Diffractometer: counts full scale, 3000; time constant, medium; scanning rate, $1^\circ/\text{min}$; divergence slit, 1° ; scatter slit, 1° ; receiving slit, .003; X-radiation, Co; filter, Fe; KV, 30; mA, 15

Figure 5. X-ray diffractometer patterns of Ottawa Valley rock, acid-insoluble fraction. (a) Air dried; (b) treated with glycol; (c) heated 550°C 1 hour. Setting of North American Philips (Norelco) Diffractometer: counts full scale, 3000; time constant, medium; scanning rate, $1^\circ/\text{min}$; divergence slit, 1° ; scatter slit, 1° ; receiving slit, .003; X-radiation, Co; filter, Fe; KV, 30; mA, 15

Figure 6. Thermograms of acid-insoluble fractions. D.T.A. apparatus: R. L. Stone and Co.; heating rate: 10° minute; full-scale deflection: 50 μ -volts

have much in common. An endothermic peak appeared as pressure was reduced, due presumably to evaporation of hygroscopic moisture, but the low-temperature endotherm in the 100°C range, common in illites, was absent. This peak showed up, however, as a low, poorly defined hump below 100°C when the sample was heated in air at atmospheric pressure. In the temperature range 500–600°C there is a broad endotherm with subsidiary peaks, most clearly seen on the curve of the Kingston 10½- to 12-foot sample. A broad peak indicates larger crystallites than does a sharp peak (MacKenzie, 1957, p. 177). It also seems probable that the peak owes its complex nature in this case to the presence of more than one polymorph of illite. The characteristic endotherm/exotherm around 900°C is developed in both Kingston samples, but is larger in the case of the 10½- to 12-foot bed.

The curve from the sample of Ottawa Valley material differs in details from the curves of the Kingston samples. The endotherm produced on evacuation is larger and is followed by a small sharp peak at about 80°C. The peak at 490°C is single and well defined and is followed by an endotherm at about 785°C, a temperature which is approximately 80 centigrade degrees lower than that at which the corresponding reaction occurs in the Kingston samples. This peak is followed by a small exotherm.

The differential thermal analysis further supports the conclusion based on X-ray diffraction data that the illite is not a single polymorph. It also confirms that there are differences between the Kingston illite and that from the Ottawa Valley limestone, which is probably more hydrated.

Residues

In the heavy residue varying quantities of the iron minerals limonite and pyrite were found, together with detrital grains of pale-green pleochroic tourmaline. In the residue from the Kingston 24- to 30-foot bed there were rare crystals of ceylonite. Differential thermal analysis was used to confirm the presence of organic material intergrown with illite in the magnetic fractions. This was only present in small amounts in the Kingston 24- to 30-foot rock.

Pale-brown, cryptocrystalline, detrital flakes were concentrated from the Kingston 20- to 21-foot rock in a magnetic fraction which was heavy in liquid specific gravity 2.77 and light in 3.0. The refractive index was found by the

immersion method to be 1.610, and a powder X-ray diffraction photograph indicated that the mineral belongs to the chlorite family. Similar flakes were not found in any of the other samples.

Many of the worst cases of alkali-aggregate reaction have been attributed to the mineral opal from which a swelling, alkali-silica gel is produced (Powers and Steinour, 1955, p. 498). At Kingston many of the features associated with this reaction were lacking (Swenson and Gillott, 1960, p. 30), and microscopic examination failed to detect much, if any, gel although its presence was suggested from a consideration of the form of adsorption isotherms (Feldman and Sereda, 1961, p. 211). The acid-insoluble fraction of the rocks was suspended in a liquid of specific gravity 2.4 in an attempt to concentrate opal by flotation. In each case, however, only a low percentage of the total rock floated, and microscopic examination, differential thermal analysis, and powder X-ray diffraction of the residue failed to detect it.

Illite and some quartz were identified in all samples except the Kingston 20- to 21-foot rock. The clay mineral was generally intermixed with organic material and thermograms run in air show large exothermic peaks (Fig. 6). The concentrate from the 24- to 30-foot bed gave a diffraction pattern similar to that of the other samples but differed in physical appearance and was green in color. The small amount of material available was mixed with calcined alumina in a proportion of 1:5 but failed to register peaks on a thermogram at 50μ volts.

CHEMISTRY

General Statement

Chemical analyses were carried out under the supervision of Dr. J. A. Maxwell of the Geological Survey of Canada on the total solid and the acid-insoluble fractions by methods modified from those introduced by Shapiro and Branock (1956). The results in tabulated form are available from the author. It was hoped that over short distances within single beds there would be constancy of chemical composition so that a direct comparison could be made between the selected horizons and between the rock and aggregate.

It was thought that a recombination of oxides into minerals might make easier the comparison of analyses if mineral phases with the same chemical compositions could be computed in each case. Imbrie and Poldervaart (1959) de-

scribed a scheme upon which the mineral calculation adopted here was based. It is emphasized that this procedure was adopted as an aid to comparison between samples and that the minerals computed may not be present in the actual rock. In particular, the computation of "montmorillonite" in this section does not indicate the presence of a swelling clay mineral in the sample, and the word is used only to comply with the scheme of Imbrie and Poldervaart (1959, p. 591) and the method of allocation of oxides.

Computational Procedure

Details of the calculation are available from the author. Section (i) of the Imbrie-Poldervaart scheme was followed except that L.O.I and H_2O^- were not available and calculations based upon these values were omitted. It was also assumed that only 20 per cent of the sulphur was combined as pyrite as it was found that for some of the rocks uncombined sulphur would remain even if all the divalent cations were used. The figure of 20 per cent is arbitrary but ensures that at least some ferrous iron remains after the calculation of pyrite in each case. The excess of sulphur indicates that this element is present in some compound other than pyrite, which is known to be present, and one may speculate that it is combined in some form of organic compound as the carbon content is quite high and sulphur is a biophile element (Gorham, 1960, p. 466). The group SO_3 was not included in the determination and gypsum was not calculated.

For the calculation of the clay-fraction the divalent ions, magnesium, iron, and calcium, were combined, and according to the proportions of each element the molecular weight of the clay minerals was adjusted. The same procedure was followed for Al_2O_3 and Fe_2O_3 , which were combined for chlorite and montmorillonite and the molecular weight computed on the basis of a weighted average for R_2O_3 . The molecular weight of illite was computed on the assumption that only Al_2O_3 was present, as for certain of the rocks there was insufficient Fe_2O_3 for even 1 mole per illite mole.

All the analyses of the acid-insoluble fraction, except that of the Ottawa Valley aggregate, fell within field C (ii) of the Imbrie-Poldervaart scheme. In many, however, the supply of R_2O_3 became exhausted prematurely; it was found that this difficulty could be overcome if picrophengite was calculated instead of sericite. When the "total rock" analyses were considered

it was found that if the chemical formulae assumed in this scheme were followed the samples fell within different clay-mineral fields. As comparison of the samples was the main purpose of the calculation the assumptions made as to chemical compositions were adjusted so that the same minerals could be computed in each case.

The structural formula adopted for illite was

The relationship of this formula to the Imbrie-Poldervaart formula can be seen by multiplying by 7.5:

(Imbrie-Poldervaart formula)

There is thus more K_2O , less H_2O , and less MgO . There is less replacement of silica by trivalent ions, and more vacancies had to be assumed in the tetrahedral layer. The formula adopted does not necessarily bear more than the most general relationship to the true composition of the mineral present in the rocks. In illite which actually occurs in Ottawa Valley limestone, for example, the degree of hydration is certainly greater than in the Kingston samples, although the same composition is assumed for both in the calculation.

The chlorite formula was also modified so that more divalent and less trivalent ions would be used. The structural formula in this case can be represented as

The amount of illite, chlorite, and montmorillonite computed depends on the amount of K_2O , $\text{R}''\text{O}$, and R_2O_3 , respectively, and these oxides are disposed of successively in this order. The moles of illite are given by $\text{K}_2\text{O}/3.5$, of chlorite by $\text{P}'/5$, and of montmorillonite by $\text{U}/2$, P' being the moles of $\text{R}''\text{O}$ remaining after illite and U the moles of R_2O_3 remaining after illite and chlorite. The ratio $1.9\text{H}/3.5\text{F}'$ had to be less than unity in every case if there was to be $\text{R}''\text{O}$ available for the formation of chlorite after illite, and the values 1.9 and 3.5 were arbitrarily arrived at to ensure that some $\text{R}''\text{O}$ would remain unused after illite. Similarly, the amount of R_2O_3 used in each mineral was derived in such a manner as to ensure an excess after the computation of illite and chlorite

and the balance was computed as montmorillonite. In the case of both silica and water a deficit occurred for certain of the analyses, and while it does not affect the value of the computation it emphasizes for comparative purposes that the compositions assumed for the minerals are hypothetical.

Discussion of Chemical Results

Various graphical plots were made of the oxide and computed mineral percentages but in no case was a clear-cut relationship found between reactivity and composition. Figures 7 and 8 show the percentage of oxide plotted against per cent linear expansion in concrete beams that had been kept for the same time under high-humidity conditions. The SiO_2 curve is plotted separately in view of the important role assigned to this material when amorphous as causative agent in the alkali-silica reaction (Powers and Steinour, 1955, p. 498). There is no maximum in the percentage curve of any oxide that corresponds with the most expansive concrete.

A consideration of the analyses of the Kingston rocks in order of increasing depth from the surface of the quarry shows that there is an increase in the percentage of SiO_2 , Al_2O_3 , K_2O , and FeO . The Ottawa Valley rock is in many ways similar in composition to the Kingston 10½- to 12-foot bed, but it differs from the Kingston samples in that it has a lower content of MgO and a higher carbon and sulphur content. Analysis of the acid-insoluble fraction shows that all of the beds have a similar content of CaO and FeO . The MgO percentage is also similar in the Kingston samples, but there is a smaller percentage in the acid-insoluble sample from the Ottawa Valley, as was found in the analysis of the total solid. Computed values for the $\text{SiO}_2/\text{Al}_2\text{O}_3$ oxide ratio are shown in Table 2.

The maximum in the Kingston 24- to 30-foot bed suggests that this sample contains the largest proportion of free silica, which is probably present as the silt-size quartz referred to under Petrography. It is also interesting to note that there is apparently more uncombined silica in the Ottawa Valley sample than in the three reactive Kingston rocks.

A similar computation of the $\text{K}_2\text{O}/\text{H}_2\text{O}$ -oxide ratio gave the values shown in Table 3.

The value of the ratio is similar in the three reactive beds; it has a maximum for the slightly reactive Kingston 24- to 30-foot bed and a minimum for the nonreactive Ottawa Valley

sample. This low value is probably a reflection of increased hydration in the mineral illite, which was inferred from the tailed form of the $10\text{\AA}$ peak on the diffractogram (Fig. 5).

Rims similar in appearance to those sur-

not be discerned in the analysis of the acid-insoluble fraction, but the significance of this is obscure.

The dolomite and calcite content expressed as a percentage of the total carbonate is shown

Figure 7. Oxide content of acid-insoluble fraction of rock versus expansion of concrete beams

rounding Kingston aggregate have been described by Bisque and Lemish (1960, p. 45) who have shown silicification to be the cause, the silica being derived from the cement paste. In the analysis of total solid the aggregate from the three reactive Kingston beds contains more silica than the rock, but this trend is reversed in the Ottawa Valley and Kingston 24- to 30-foot aggregates which, however, generally show little or no rim formation. This trend can-

graphically in Figure 9. Calcite content is high in Ottawa Valley limestone and low in the Kingston 24- to 30-foot bed. On the other hand the Ottawa Valley sample contains the smallest amount of dolomite whereas it reaches its maximum in the Kingston 24- to 30-foot bed. In each of the three reactive beds there is less dolomite and more calcite in the aggregate than in the rock. This effect could be produced by the dedolomitization reaction (Swenson and

Gillott, 1960, p. 26), which leads to an increase in the amount of calcite at the expense of dolomite.

In the computation based on the analysis of the acid insolubles the recombination of K_2O , Al_2O_3 , and SiO_2 into illite is reflected in the re-

shows little apparent sign of recrystallization. There are also thin bands composed of intracasts or fragments of fine lime-mud (Pl.1, fig. 2) surrounded by a matrix of larger crystals of clear sparry calcite, which fabric criteria indicate were an original cement rather than the

Figure 8. Silica content of acid-insoluble fraction of rock versus expansion of concrete beams

lationship between the percentages of these oxides and the form of the curve for the mineral (Fig. 10). A similar control is also apparent in the relationship between per cent Na_2O and albite, and divalent cations and chlorite.

CONCLUSIONS

The three reactive beds are dolomitic limestones or calcitic dolomites, the microfacies characteristics of which are similar. The matrix consists of fine lime-mud composed of microcrystalline or cryptocrystalline calcite, which

product of grain-growth from smaller crystals. Although much of the calcite shows little evidence of recrystallization or grain-growth, the rock has been extensively dolomitized. Dolomite crystals occur as well-formed euhedra, some of which contain inclusions arranged symmetrically (Pl. 1, fig. 1). On the megascopic scale the dolomite is evenly distributed but shows some irregularity on the microscale. Quartz as small detrital crystals and murky argillaceous material occurs in all three beds.

The reactive rock differs from the unreactive

Ottawa Valley rock and the very slightly reactive 24- to 30-foot layer. The latter is a silty argillaceous dolomite, which differs from the reactive beds most markedly in that it is more dolomitic and contains more detrital quartz. It is also more porous. The Ottawa Valley aggregate contains rock of variable petrographic character. In general, however, it differs from the reactive beds in that there is less dolomite; when this mineral is present, the distribution

TABLE 2. $\text{SiO}_2/\text{Al}_2\text{O}_3$ IN ACID INSOLUBLE

Sample	Rock	Aggregate
Kingston 6-7-ft. bed	3.12	2.83
Kingston 10½-12-ft. bed	3.46	3.40
Kingston 20-21-ft. bed	3.55	3.15
Kingston 24-30-ft. bed	4.69	4.20
Ottawa Valley limestone	3.78	3.64

sometimes differs (Pl. 2, fig. 1). The rock is more fossiliferous and there are indications of partial recrystallization of the matrix. The reactive beds appear to have formed in shallow water under fairly quiet conditions when the

bands of microcrystalline calcite and clay were deposited. Currents were sometimes active and produced the sedimentary features characteristic of scouring and erosion. When quiescence was resumed the thin sheets of loosely deposited

TABLE 3. $\text{K}_2\text{O}/\text{H}_2\text{O}$ IN ACID INSOLUBLE

Sample	Rock	Aggregate
Kingston 6-7-ft. bed	0.956	1.0
Kingston 10½-12-ft. bed	0.963	0.987
Kingston 20-21-ft. bed	1.16	1.05
Kingston 24-30-ft. bed	1.317	1.336
Ottawa Valley limestone	0.70	0.825

intraclasts accumulated. Subsequent filling of pores resulted in lithification by clear sparry calcite and formation of intraspartic bands (Pl. 1, fig. 2) within the more homogeneous layers of microcrystalline calcite. The very slightly reactive 24- to 30-foot bed formed under different sedimentary conditions. The larger crystals of quartz and the much higher content of argillaceous material imply stronger currents of greater transporting power. What-

Figure 9. Computed mineral composition expressed as total carbonate

ever the cause it seems likely that this bed was deposited under conditions of higher energy than were the reactive beds. The oörites in some of the Ottawa Valley rocks (Pl. 2, fig. 2) also imply more disturbed sedimentary condi-

tion in the cell of ions such as Mg, Fe, or Mn which are smaller than calcium.

Illite and chlorite are the clay minerals. Swelling clay minerals are absent and no evidence could be found that soaking the rock in

Figure 10. Computed mineral-composition variation in acid-insoluble fraction

tions than prevailed when the reactive beds were laid down at Kingston.

Mineralogical investigation showed that the dolomite is of the nonideal metastable type, which has an enlarged unit cell from the incorporation of an excess of calcium above 50-mole per cent in the lattice. The reactive Kingston rocks contain dolomite that is somewhat more abnormal than the dolomite contained in the nonreactive rocks.

The calcite has lattice constants smaller than those of pure calcite because of the incorpora-

alkali brought about formation of expanding from nonexpanding lattice minerals. The illite appears to be a mixture of disordered 1M and 2M muscovite; although its powder pattern could not be matched perfectly with either polymorph it has affinities with both. The form of the 10A peak suggests that the illite in the Ottawa Valley rock is more hydrated than that in the Kingston samples. This is supported by differential thermal analysis.

An attempt was made to concentrate by flotation any opal present in the rocks. In all

cases the light fraction amounted to a very small percentage of the rock, and mineralogical examination of this residue failed to detect opal. Active silica is unlikely to account for the alkali reactivity of these rocks.

All the rocks contain less than 50 per cent acid insoluble and they are, therefore, properly classified as carbonate rocks (Carozzi, 1960, p. 193). There are in the literature detailed discussions of the chemistry of limestones and dolomites and there are various suggested classifications for them based on chemical composition (Pettijohn, 1956, p. 417). According to one such scheme and the mineral computations followed in this report the rocks and aggregates fall into the fields shown in Table 4.

The three most reactive rocks are intermediate in composition between limestone and dolo-

series. Rocks from other areas petrographically similar to those from Kingston, however, are known to react expansively with strong alkali. It therefore seems reasonable to assume that a connection exists between the alkali reactivity and petrography of the rocks. It seems that certain features found during the analyses of these rocks are likely to be significant in elucidating the mechanism of expansion and are, therefore, of utmost importance to the petrographer concerned with the assessment of usable aggregate. These features are:

- (1) The rocks are calcitic dolomites or dolomitic limestones in which there is a fairly uniform distribution of euhedral dolomite crystals, some of which contain inclusions of argillaceous material.
- (2) The groundmass by which the dolomite

TABLE 4. CLASSIFICATION OF ROCKS AND AGGREGATES

	<i>Rock</i>	<i>Aggregate</i>
Ottawa Valley	limestone	magnesian limestone
Kingston, 6-7-ft. bed	calcitic dolomite	dolomitic limestone
Kingston, 10½-12-ft. bed	calcitic dolomite	dolomitic limestone
Kingston, 20-21-ft. bed	dolomitic limestone	dolomitic limestone
Kingston, 24-30-ft. bed	calcitic dolomite	dolomite

mite, and according to several authorities are relatively less common than the end members of the series (Pettijohn, 1956, p. 417; Steidtmann, 1917, p. 436). On the other hand, the analyses of the acid-insoluble fraction of the limestones are quite similar to one given by Clarke (1924, p. 631) for an "average shale." It seems clear that the sulphur content is too high in certain of the rocks for pyrite to be the only sulphur-containing material, and it appears most likely that the sulphur is combined with carbon. The K_2O/H_2O ratio is a chemical reflection of mineralogical differences in the illite.

The computation adopted shows that, if the same minerals are to be calculated for each sample, rather drastic modifications have to be made to the formulae for the clay minerals proposed by Imbrie and Poldervaart (1959, p. 589), despite the fact that other techniques have shown the rocks to be similar mineralogically.

It is not known why expansion is produced by aggregate made from rocks of the Pamela formation, Kingston, nor is it possible on petrological criteria to predict the degree of reactivity that will result from the use of aggregate made from a particular bed of rock in a

euhedra are enclosed is dusky-brown cryptocrystalline calcite. Although there are layers in which the cement consists of larger crystals of clear calcite these are subordinate to the bands of microcrystalline calcite. The over-all picture is one of sedimentation under relatively quiet conditions—the lithified products of a low-energy environment of deposition (Folk, 1959, p. 12).

- (3) The calcareous ooze seems not to have recrystallized since sedimentation.
- (4) There is a widespread distribution of argillaceous material which consists of illite and chlorite together with some quartz, which gives the rock an acid-insoluble content of about 10 per cent.
- (5) The dolomite is of the metastable type, which contains an excess of calcium above the 50-mole per cent of an ideal dolomite.
- (6) The rocks are of low porosity.

Although the mechanism is obscure by which the rocks expand in strong alkali, it is likely that some or all of these features are significant. Therefore, if a dolomitic limestone or calcitic dolomite is recognized petrographically to be similar to the reactive Kingston rocks, it should

not be used as aggregate with high-alkali cement unless measurement of length-changes on concrete beams under controlled conditions (Swenson and Gillott, 1960, p. 20) or some other test procedure (under development) has proved the rock to be nonexpansive.

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