Note on datolite and other minerals in a contact-altered limestone at Chapel quarry, near Kirkcaldy, Fife.[1]

(With Plate IX.)

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THE Chapel, or Charlestown Main, Limestone horizon lies in the Lower Limestone Group of the Carboniferous Limestone Series. At Chapel quarry the limestone is underlain by a thick sill of quartz-dolerite. The contact, however, is not exposed but probably lies only a few feet below the quarry floor.[2] Other intrusions are seen in the vicinity: a very thin dike of decomposed basalt, which connects with a small 'float' of white trap, cuts the limestone about 30 yards north-eastwards of the point where the quarry crosses the Chapel road, and a small sill of olivine-dolerite or teschenite occurs at a horizon about 100 feet above the top of the limestone. In the quarry the limestone is about 60 feet thick. The contact-alteration described in this paper is most evident near the base of the limestone face and its extension laterally in the quarry has no relation to the outcrop of the thin dike. The alteration is therefore considered to be essentially connected with the underlying, unexposed, sill of quartz-dolerite, and, as will be shown in the following pages, has been effected by the action of hot solutions rather than by thermal reconstruction.

The specimens in which datolite was first observed were collected by Mr. D. Haldane, through the courtesy of Mr. William Black, during excavations undertaken by the Charlestown Lime Co., Ltd., with a view to renewing operations at the lime-works. The excavations, now abandoned, were made at the base of the quarry face, 150 yards north-eastwards of the point where the quarry crosses the Chapel road. Subsequently Dr. A. G. MacGregor and Mr. Haldane made a fuller collection of material to determine the extent of the alteration. The minerals determined in the altered limestone are datolite, garnet (mainly grossular), pectolite, talc, and apophyllite. In the limestone forming the walls of the thin dike mentioned above much pyrite and a small quantity of blende are present. Working of the limestone at Chapel quarry was stopped because fluxing took place in the kiln.

The limestone.—The Chapel Limestone is a grey compact crinoidal limestone of a type common in the Lower Carboniferous. Towards the base of the face in Chapel quarry mineralogical change is made manifest by the development of a white or pale green lustreless aggregate as flecks and wisps disseminated through the grey limestone, as concentrations of irregular size and shape, and as thin layers parallel to the bedding. Some of the last are sharply bounded against the grey limestone, others show passage into the limestone either by gradual change

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[1] Communicated with the permission of the Director, Geological Survey and Museum.

[2] J. K. Allan and J. Knox, The economic geology of the Fife coalfields, Area II. Mem. Geol. Surv. Scotland, 1934, p. 24.

in the proportions of carbonate and aggregate, or by way of a zone of veins and strings of aggregate. In one specimen, S 34826,[1] replacement of crinoid columnals and shell fragments by the aggregate can be readily observed in hand-specimen. Thus the macroscopic relations suggest that the aggregate may partly represent shaly intercalations, but certainly is in part a metasomatic introduction.

Where the grey limestone is free of the white or pale green aggregate it has usually a slightly indurated appearance suggesting contact-metamorphism of a very low order. Under the microscope a wide range of texture and mineral composition is observed. In some specimens the shapes of fossils are well preserved (S 34819), but the matrix of fine calcareous debris and the walls of the more delicate fossils may be recrystallized in finely granular calcite. In others the limestone is converted to a mosaic of calcite grains with complete destruction of fossil shapes. There is every stage between these extremes of texture, but there is no regular relation of the degree of recrystallization to the proportion of new silicates nor to the vertical distance from the quartz-dolerite sill. For example, granoblastic recrystallized limestone occurs up to at least 30 feet from the base of the quarry face, and recrystallized limestone of similar texture occurs in patches near the base of the quarry face among fossiliferous limestone in which there is little textural change. These granular patches usually show by transmitted light a brown coloration which is discharged when the powdered rock is roasted and is probably due to disseminated bituminous or carbonaceous particles. Similar brown calcite mosaic occurs as infilling in the chambers of foraminifera and polyzoa and within ostracod bivalves (S 34444). These observations perhaps indicate that calcite is easily recrystallized at comparatively low temperatures in the presence of hydrocarbons residual from the decay of organisms, the less volatile hydrocarbons being trapped in the growing calcite.

In thin sections of the limestones the fine aggregate is seen in all proportions from small interstitial tufts of fibres or flakes to extensive accumulations. Some of the latter, as already recognized in hand-specimen, appear in thin section also to represent layers of calcareous shale. The aggregate in these is extremely finely divided and is almost opaque and enwraps numerous organic remains including crinoid columnals, often showing trabecular structure, ostracod and other shells, and Productid spines (S 34445). In other specimens where the fine aggregate is abundant it is distributed in irregular manner through the limestone and replaces the fossils. The crinoid columnals are usually replaced peripherally, but the alteration has sometimes proceeded throughout the whole plate (S 34826 and pl. IX, fig. 3). The fine aggregate is sometimes pectolite, but in most specimens is steatite. Distinction in thin section is based mainly on the different appearance of the straight, sharply bounded fibres of pectolite and the softer, less regular laminae of steatite. The individual crystals of the two minerals are, however, often so small that distinction in thin section is not possible. Pectolite has been observed as the main constituent of two very thin, sinuous, and impersistent veins (S 34445A) in one of which the pectolite is locally replaced by apophyllite.

Garnet (pl. IX, fig. 4) is the next most abundant silicate and occurs in variable proportion through the limestone up to 24 feet above the quarry floor. It shows a tendency to be abundant where there is much fine aggregate, both of the pectolite

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[1] The numbers refer to specimens and corresponding slides in the Sliced Rock Collection of the Geological Survey and Museum.

and steatite types. Datolite is more abundant in the purer calcite portions (pl. IX, fig. 4). Its vertical extension seems to be much less than that of garnet, and it has not been observed at a level more than 8 or 10 feet above the quarry floor. Both garnet and datolite are present abundantly in the type of limestone which retains perfect fossil shapes as well as in the granoblastic limestone. Even within the limits of a thin section the content of these minerals is, however, very variable. The mode of occurrence of the datolite and garnet in the texturally little altered limestone is interesting. They appear indifferently in the fine matrix, inside the casts of shells, within the single crystals composing the crinoid columnals, and seem to have developed with special ease along the axial canal of the columnals. A remarkable phenomenon is the replacement of gastropod shells by aggregates of minute garnet (pl. IX, fig. 2), and it is suspected that the walls of foraminiferal and polyzoan chambers are replaced in like manner, but the structures, being very delicate, have been too greatly obscured for definite identification (S 34445B).

The observations recorded above prove the permeation of the rock by solutions carrying silica, boric oxide, and soda (perhaps also alumina and magnesia) in a condition chemically active but productive of little physical reconstruction. The reason for the fixation of silica and alumina as garnet in the shells is not apparent. Analyses of the hard parts of marine organisms show that in general the sum of silica and alumina is less than one per cent.,[1] and these oxides must therefore have been carried into the shells. Local concentration of alumina and silica leading to the initiation of centres of garnet crystallization may perhaps have been produced if a film of clay were adherent to the shell. Garnet occurs also as a narrow rim surrounding small nodules of chert (S 34445B2).

Apophyllite is a later mineral than those already described and occurs in veins and irregular patches (pl. IX, fig. 5) indefinitely bounded against the rest of the rock. The relations indicate that apophyllite has replaced both calcite and fine aggregate, the grossular and datolite remaining as inclusions in the plates of apophyllite. Though apophyllite is the scarcest of the minerals under description it is the only one of the suite which has already been recorded from Chapel quarry. Heddle states that it is found as fine crystals associated with elaterite and dolomite.[2]

In the following paragraphs notes on optical and other properties of the minerals are given. As already stated the fine aggregate is in some specimens pectolite, but in most it is steatite.

Pectolite is found most abundantly in the small extension of the quarry west of the Chapel road, where it occurs disseminated through the limestone, and also as white earthy-looking masses several centimetres in length, and as coatings on joint-planes. The mineral forms microscopic bundles of sharp-edged fibres which are sufficiently parallel to afford a biaxial positive figure from cross-sections of the bundle (S 34812). The individual fibres are about 0·005 mm. thick, but larger crystals reaching 0·03 mm. in thickness are fairly common. The refractive indices determined by immersion in oils are $\alpha \leqslant 1{\cdot}595$, $\gamma = 1{\cdot}632$. When heated in 1 : 1 HCl, the mineral is partly soluble, partly replaced by opaline silica, and partly unaffected. It gives a strong sodium coloration to the Bunsen flame. Mr. F. A. Bannister confirmed the identification by an X-ray powder photograph which proved identical with that of pectolite from Paterson, New Jersey.

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[1] F. W. Clarke and W. C. Wheeler, Prof. Paper U.S. Geol. Survey, 1922, no. 124.

[2] M. F. Heddle, Mineralogy of Scotland, 1901, vol. 2, p. 82.

Talc (*steatite*).—Optical tests on the fine aggregate from the specimens in the main quarry proved very difficult. The aggregate is softer and more matted, the individual fibres less clearly defined than is the case for the pectolite aggregate of the quarry west of the road. Highest and lowest refractive indices of approximately 1·580 and 1·535 were determined for the principal constituent of the aggregate in several specimens, and in view of these values and of the mode of occurrence of the material in the manner of argillaceous intercalations it was thought the mineral was a kind of sericite or illite, possibly a sodic variety, since the analysis of the limestone shows Na_2O 1·82%. Mr. F. A. Bannister, to whom we then appealed, proved by X-ray methods that the aggregate is steatite, the powder photograph proving identical with that of crystallized talc. A partial chemical analysis by Mr. C. O. Harvey showed SiO_2 58·0, MgO 29·4, CaO 1·2, Na_2O+Li_2O 0·2, and K_2O 0·1 %, yielding $SiO_2:MgO = 4:3$ as required for talc. The aggregate, nevertheless, is not greasy to the touch like talc, perhaps on account of the considerable quantity of minute garnet included in it. Though talc is the main constituent of the earthy-looking material in the main quarry, pectolite is present in some specimens, and in a slide, gelatinized and stained primarily for the detection of apophyllite, pectolite is observed to be stained slightly (S 34445B).

The two main constituents of the fine aggregates have thus been identified, but it is certain that other silicates are present in smaller quantity. They can be detected when the powder is immersed in suitable oils and also in certain small areas of the thin sections where apophyllite has replaced the most of the fine aggregate. Tiny straight-extinguishing fibres of a mineral, perhaps antigorite, perhaps pectolite though the birefringence seems too low, persist in the apophyllite. The minor constituents perceived by immersion in oils include pyroxene as tiny prisms with high refractive index and highly inclined extinction, and small sharp-edged prisms showing straight extinction, positive elongation, and refractive indices approximately 1·605 and 1·625, properties which would agree with anthophyllite.

Garnet forms more or less perfect dodecahedra of diameter usually about 0·05 to 0·1 mm., rarely as great as 0·2 mm., and often very minute. Most of the crystals have a turbid appearance which appears to be due to the presence of a broken, or otherwise discontinuous coating of andradite, of refractive index ranging upwards from 1·825 to over 1·845. The main garnet is grossular with n 1·740 on the average, but varying slightly above and below this value. Mr. F. A. Bannister kindly offered to make a powder photograph of this mineral also and found as the unit-cell dimension 11·85±0·02Å., a figure agreeing well with other measurements for grossular, notably G. Menzer's measurement of 11·84Å. The andradite skin produced no effect in the X-ray photograph. It is worth noting that there is no transitional zone between the grossular and the andradite and that the relation is therefore not one of zoning in the usual usage of this term. It was observed also that the garnets which show perfect crystal faces are entirely grossular without any andradite coating.

Datolite forms rounded grains which are usually about 0·1 to 0·2 mm. in diameter but reach up to 0·4 mm. (S 34821) and they tend to occur in small groups (pl. IX, fig. 5). The grains sometimes have an irregular outline as if corroded. The mineral is scarcely attacked by cold dilute HCl and is therefore easily concentrated by dissolving away the calcite matrix. In hot acid, however, it is readily gelatinized and the powder moistened with HCl yields a pronounced boron reaction to the flame test. The optical constants were determined as α 1·625, β 1·653, γ 1·670, 2V 73·5°, negative, and are almost identical with those measured by McLintock on datolite from the Lizard.[1]

The refractive indices were determined by immersion in oils, and the optic axial angle is the average of values ranging from 72° to 74·5° measured on five different grains by use of the universal stage. Mr. F. A. Bannister has made an X-ray powder photograph of the mineral and states that the pattern is identical with that of datolite from Seisser Alpe, Tirol.

Apophyllite was first detected in an unfinished section which had been covered with glycerine. The perfect cleavage, which is not seen in balsam mounts, was then conspicuous. The mineral gelatinizes readily with acid and its fairly wide distribution is most easily observed by treating the thin section with acid and staining the gelatinized mineral. Refractive indices were determined on an uncovered section, washed clean of balsam, as α (or ω) 1·535, γ (or ϵ) 1·537 or 1·538. The mineral is optically positive, but we were unable to decide whether the figure observed with the conoscope was that of a uniaxial or of a biaxial mineral. The extinction is straight, α is parallel to the cleavage and the latter is normal to sections which give the highest polarization colour (an anomalous brownish-grey).

Chemical composition.—By courtesy of the Director, Macaulay Institute for Soil Research, we are able to reproduce an analysis made by Mr. A. Muir of a composite sample of the limestone, prepared from two datolite-bearing specimens. One of these was collected near the level of the quarry floor, and the other at a level of approximately 6 or 8 feet above the floor. The material analysed is thus not a bulk-sample of the quarried rock.

SiO_2	21·72	MgO	1·90	H_2O-	0·41
TiO_2	0·40	CaO	38·82	P_2O_5	0·08
Al_2O_3	4·26	Na_2O	1·82	CO_2	26·68
Fe_2O_3	1·00	K_2O	0·25	B_2O_3	2·00
MnO	0·09	H_2O+	n.d.	FeS_2	0·21
				Total	99·64

The amounts of B_2O_3, K_2O, and MgO shown by this analysis would correspond respectively to datolite 9·15, apophyllite 4·8, and steatite 6·0 %, figures which agree for apophyllite, but seem rather high for datolite and low for steatite relative to the evidence of the thin sections. The content of soda would correspond to 19·5 % of pectolite, the only soda-bearing mineral we have been able to identify. While such a proportion is probably realized in parts of the limestone in the small quarry west of the road, it appears to be very high when referred against the evidence of the thin sections of limestones from the part of the main quarry which supplied the specimens for analysis. Either some other soda-bearing mineral is present or a local concentration of pectolite must have occurred in the sample analysed. A rational analysis in terms of the minerals calcite, datolite, grossular, talc, pectolite, and apophyllite cannot be calculated from the analysis with a reasonable degree of approximation.

Summary and Conclusions.—In the limestone at Chapel quarry the minerals

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[1] W. F. P. McLintock, Min. Mag., 1910, vol. 15, p. 412.

datolite, grossular, and steatite are of contemporaneous formation, pectolite is partly contemporaneous with datolite, and garnet partly somewhat later, while apophyllite is definitely later than these minerals, enclosing or replacing them. These silicates occur as constituent minerals of a limestone which is texturally little altered; garnet and datolite appear among the calcite infilling well-preserved shells and in the canals and plates of crinoids; shells are replaced by garnet, and pectolite replaces shell fragments and crinoid columnals without destruction of the fossil shapes. It is evident therefore that the minerals were developed through the agency of fluids capable of intimate penetration through the rock, chemically very active but productive of only a low degree of physical reconstruction.

With the exception of steatite, the silicates are richly calcic, and with the exception of grossular they contain the elements of water. For their formation introduction of much silica has been necessary and minor but notable amounts of boric oxide, soda, potash, and probably fluorine have been required to form datolite, pectolite, and apophyllite. It is possible that the alumina and subordinate ferric oxide of the garnet were originally present in the limestone in the form of slightly ferruginous clay substance. The metasomatizing fluids must therefore have been essentially solutions of alkali silicates, initially sodic and finally potassic, and containing in the early stages boric oxide.[1] The temperature of the solutions was probably low, since they reacted to form hydrous minerals and effected little physical reconstruction. The development of grossular under such circumstances[2] affords striking confirmation of the fact, already well recognized, that this species of garnet forms with ease in aluminous limestones subjected to thermal metamorphism.

The formation of a pectolite-like mineral from calcite by the action of hot sodium silicate solution was realized experimentally by Lemberg in 1885, and the formation of apophyllite by the action of potassium silicate solution on calcite was carried out by Churin between 1913 and 1918.[3] Lemberg recorded the pectolite-like mineral as formed also by the action of sodium silicate on datolite. This transformation, though suspected in specimen S 34812, was not convincingly observed in the Chapel quarry limestone.

Datolite is best known as a mineral from cavities and veins in basic igneous rock and in mineral deposits and hydrothermal veins associated with the contacts of limestone and igneous rock.[4] As a constituent of the altered country-rock it has, however, been recorded only rarely, and in Britain up to the present only from a calc-flinta associated with greenstone in the aureole of the Dartmoor granite.[5] In the Caucasus low-temperature metasomatic replacement of limestone by datolite occurs at Pyatigorsk,[6] where Senonian limestone is changed by the action of hot springs, of temperature 46°C. and containing 0·00344 gm. per litre of B_2O_3, into compact datolite-bearing hornstone in which foraminifera are preserved.

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[1] The presence of boron in the residual glass of tholeiite has been stated by Dr. F. Walker as conclusively proved but to an amount under 1%, Min. Mag., 1930, vol. 22, p. 375.

[2] Cf. J. H. Taylor, Min. Mag., 1940, vol. 25, p. 545. (Grossular associated with analcime.)

[3] See G. W. Morey and E. Ingerson, Econ. Geol., 1937, Supplement to vol. 32, no. 5; Bibliography items nos. 31 and 86.

[4] For example, E. V. Shannon, Proc. U.S. Nat. Mus., 1924, vol. 66, art. 2, and 1925, vol. 66, art. 28. [M.A. 3-204.]

[5] K. Busz, Neues Jahrb. Min., 1899, Beil.-Bd. 13, p. 126.

[6] P. Tchirwinsky, Zeits. Krist., 1929, vol. 70, pp. 260–261. [M.A. 4-181.]

From the Caucasus also datolite-bearing rocks have been described as a result of the action of boron-rich emanations or solutions on limestone in contact with laccoliths of trachyliparite.[1] Pseudomorphs after datolite in lenses of limestone within gneiss altered by diabase have been found at Hopewell mine, Pennsylvania.[2] The paragenesis of datolite at Chapel quarry indicates that here also the conditions of formation were of comparatively low-temperature hydrothermal type.

The occurrence of steatite in the limestone is puzzling. Like the other silicates present it is a hydrothermal mineral but, in contrast to them, it is a purely magnesian silicate. Three hypotheses of its formation may be mentioned: (i) the steatite may represent argillaceous material into which magnesia has been introduced and from which alumina has been removed; (ii) and (iii) the steatite may represent magnesium carbonate present (ii) as dolomite in dolomitized limestone or (iii) as a primary constituent of the limestone. The first hypothesis involves metasomatic introduction of magnesia which is of all the oxides the one least likely to be effluent from a quartz-dolerite. Moreover, there is no indication of the secondary formation of an aluminous mineral from the alumina removed according to this hypothesis. If, as seems more probable, the magnesia were present in the limestone prior to its hydrothermal alteration one of the hypotheses (ii) and (iii) should be true. Though dolomitized limestone is certainly present in Chapel quarry it appears to be resistant to the hydrothermal changes and none of the new minerals of the limestone appears in the dolomite. It is, moreover, a compact granular rock showing scarcely a trace of fossil structures, but these are abundantly present in the steatitic limestone. It would appear, therefore, that the steatite has developed from magnesia present as a primary constituent of the limestone. According to Clarke magnesia may occur in this manner either as a direct precipitate from shallow marine waters in which organisms are decomposing or as a constituent of the hard parts of organisms. These have been proved to contain varying proportions of magnesium carbonate up to 11, 11, 14, and 8·6 % in the case of polyzoa, foraminifera, echinoderms, and brachiopods respectively.[3] If the magnesia of the steatite has been so derived there is no indication of the fact in the distribution of the steatite, for this mineral shows no tendency towards preferential replacement of, for example, crinoid remains. Again, if the steatite represents primary magnesium carbonate the dense, almost monomineralic aggregates which it forms show that wide redistribution of magnesia and lime must have been involved at the time when hydrothermal solutions were active. A further hypothesis may be mentioned. Stevensite, a mineral similar to talc in composition, has been recorded as due to the action of magnesian solutions, derived from weathered basalt, on pectolite.[4] In the Chapel limestone, however, there is no reason to believe that the steatite is derived from pectolite nor that it owes its formation to weathering solutions.

In conclusion, we wish to express our thanks to Mr. F. A. Bannister, Deputy Keeper, Natural History Museum, for his confirmatory X-ray work on datolite, pectolite, and grossular, and for his identification of steatite.

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[1] I. M. Kurman and Z. M. Usacheva, 1937. [M.A. 7–442.]

[2] G. Vaux, jr., Proc. Acad. Nat. Sci. Philadelphia, 1927, vol. 78, p. 17. [M.A. 3–121.]

[3] F. W. Clarke, Data of geochemistry. 4th edition, Bull. U.S. Geol. Surv., 1920, no. 695, p. 565.

[4] M. L. Glenn, Amer. Min., 1916, vol. 1, p. 44. [M.A. 1–31.]

EXPLANATION OF PLATE IX.

Datolite, grossular, pectolite, and apophyllite in the Chapel Limestone, Chapel quarry, Kirkcaldy, Fife.

The numbers refer to the Rock Slide Collections of the Geological Survey and Museum. All photographs are taken by ordinary light.

FIG. 1. S 34444A3. *Right: a normal type of limestone showing a crinoid columnal and shell* fragments in turbid fine-grained calcareous debris with patches of granular calcite stained deep brown. Left: a more granular limestone showing relics of organic structures, numerous clear grains of datolite, and many individual crystals and small aggregates of turbid grossular which appear in the photographs as black spots. × 12.

FIG. 2. S 34445B3. A small gastropod shell replaced by grossular. Clear datolite grains are numerous among granular calcite and form a cluster just to the right of the centre. × 25.

FIG. 3. ED 1269. A longitudinal section of a crinoid columnal is completely, and a transverse section is peripherally and axially, replaced by fibrous pectolite. Small crystals of grossular occur in the pectolite felt at the bottom of the photograph. × 10.

FIG. 4. S 34444A2. Clear roundish grains of datolite and small semi-opaque dodecahedra of grossular are numerous among granular calcite. × 46.

FIG. 5. S 34445B. The dark vein and patches are apophyllite (gelatinized and deeply stained). The patches of apophyllite in the top left quadrant of the photograph are associated with an unusually large aggregate of clear grains of datolite. × 22.

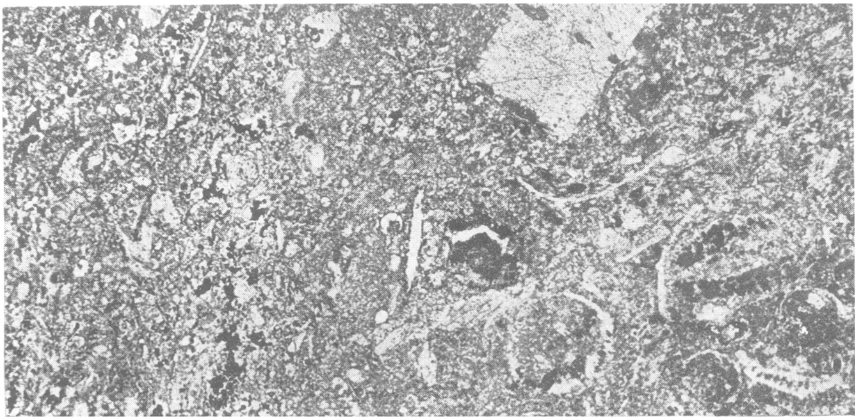

Fig. 1

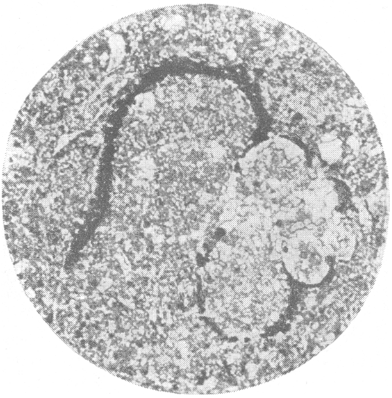

Fig. 2

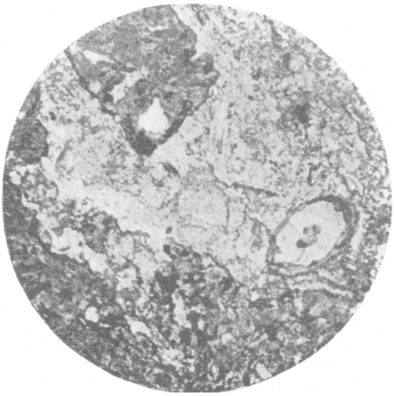

Fig. 3

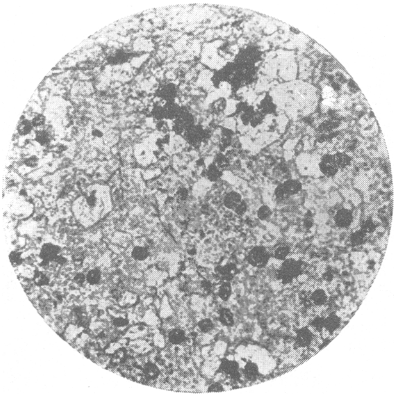

Fig. 4

Fig. 5

J. Phemister and A. G. MacGregor: Contact-altered limestone.