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Metamorphic Processes at High Temperature and Low Pressure: The Petrogenesis of the Metasomatized and Assimilated Rocks of Carneal, Co. Antrim

P. A. Sabine and B. R. Young

Phil. Trans. R. Soc. Lond. A 1975 **280**, 225-269
doi: 10.1098/rsta.1975.0101

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METAMORPHIC PROCESSES AT HIGH TEMPERATURE AND LOW PRESSURE: THE PETROGENESIS OF THE METASOMATIZED AND ASSIMILATED ROCKS OF CARNEAL, CO. ANTRIM

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(*Communicated by Sir Kingsley Dunham, For.Sec.R.S. – Received* 21 *October* 1974)

[Plates 12 and 13]

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 [Published 27 November 1975

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The Tertiary dolerite plug at Carneal cuts basalt lavas and incorporates blocks of chalk and flint from underlying Cretaceous rocks. Assimilation by the dolerite of the pure limestone and flint took place under the very rare highest-temperature, low-pressure conditions. Only about twenty-five examples of the resulting metamorphosed and metasomatized rocks are known in the world, few fully described. The rock suite enables the conditions and mechanisms of assimilation to be deduced. The pressure, about 200×10^5 Pa (200 bar), and the temperature, estimated as 1050–1100 °C, produced an exomorphic suite of larnite, spinel, merwinite, spurrite, scawtite and related assemblages, with wollastonite, quartz, plagioclase, hydrogrossular, xonotlite and related minerals representing flint. Complementary desilication of the igneous rocks gives the endomorphic suite of pyroxene-rich dolerite, pyroxenite, titanaugite-melilite-rock and aegirine- and nepheline-bearing types. Chemical analyses of titanaugite, sahlite, melilite, wollastonite and the main rock types are provided and optical and other properties of the minerals.

Two related mechanisms of limestone assimilation occurred. Most of the rocks resulted from the incorporation of the chalk in the olivine-dolerite magma: paradoxically, the addition of 18–26 % CaO to the dolerite magma so lowers the silica ratio that 17–18 % additional SiO_2 is required to produce the endomorphic hybrids, with the complementary exomorphic suite. The second mechanism, a metasomatic replacement, preserves existing mineral (and fossil) textures. Mineral textures and the preservation of a cyclostome bryozoan now composed of wollastonite (by silicification of calcite) show the metasomatism to have been a tranquil process despite the high temperature. To produce the vein assemblage of merwinite, hydrogrossular and melilite, this mechanism required 63 % CaO and 23 % H_2O, an addition which is only slightly more hydrous than calcium hydroxide. These mechanisms are evidence for the production of peralkaline rocks by limestone assimilation but only on a very small scale. The retrograde phase of metamorphism produced minerals in order of approximately increasing water content, including xonotlite, bicchulite (a new mineral), thomsonite, tobermorite, tacharanite and plombierite.

1. Introduction

The contact rocks associated with the Tertiary volcanic plug of Carneal, Co. Antrim, Northern Ireland, form one of the very rare but scientifically unusually interesting examples of the results of highest-temperature metamorphism at low pressures. Only about twenty-five occurrences are known in the world, and few have been adequately described. At depth the plug has traversed the pure flint-bearing limestone of the underlying Chalk, blocks of which have been carried up and are now preserved largely as calc-silicates. The rocks demonstrate clearly the intense but very limited effects of contact metamorphism and metasomatism of limestone (and flint), and of the resulting desilication of the igneous rocks when sufficient heat is available.

Two related mechanisms of limestone assimilation are revealed by the rock and mineral suites. The progressive incorporation of chalk (and flint) in the olivine-basalt magma results in the production of two complementary sets of rocks. The *exomorphic* suite consists of the country rocks now represented by assemblages including larnite–spinel rocks, complex associations containing melilite, hydrogrossular, merwinite, and spurrite, and hydrated and alteration products including xonotlite-scawtite-rock. Flint, still recognizable macroscopically, is represented by assemblages of wollastonite, quartz, plagioclase and hydrogrossular, with common hydrated calcium silicates.

As the chalk and flint are transformed into calcium silicates, silica is successively abstracted from the olivine-basalt magma to produce the complementary *endomorphic* suite of pyroxene-

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rich dolerite and pyroxenite, the rare, striking titanaugite-melilite-rock (the norm of which contains 11.3 % larnite), and other alkaline types.

Paradoxically, in the desilication of the magma to yield the hybrid rocks, the addition of considerable quantities of lime so increases its proportion that the lime must be balanced by the addition of silica itself, or the final rocks are too low in silica. The amounts of material involved in these reactions range probably from grams to kilograms or perhaps tonnes.

The results of a metasomatic replacement mechanism are seen on a small or microscopic scale in the precise direct substitution of one mineral by another. The preservation of a pre-existing microscopic texture of a cyclostome bryozoan shows that this was a tranquil process, despite the very high temperature, probably 1050–1100 °C.

These mechanisms of limestone assimilation provide conclusive evidence for the production of moderate to extreme alkaline rocks at this scale only. They are equally important for the (negative) evidence of what they do not show: the mechanisms are evidence against the processes on a large scale which have long since been postulated as the major source of peralkaline rocks.

Calculation of the chemical changes that accompany a metasomatic merwinite vein in dolerite shows that only restricted amounts of water were involved in the chemical reactions. Lime and water in the proportions of 63:23 parts by mass were required to produce the metasomatic minerals from 100 parts by mass of the olivine-dolerite. These proportions, fixed in the metasomatic minerals, are only slightly more hydrous than calcium hydroxide (portlandite).

The plug was emplaced under low overburden from which it may be concluded that the load pressure was about 200×10^5 Pa only. The basaltic magma, flowing through the neck, was the source of a considerable quantity of heat at a high temperature (1050–1100 °C), much more than is normally found at simple contacts of igneous rocks where a lava feeder is not present.

This combination of conditions, which is rarely encountered in limestone metamorphism, is reflected in the presence of the rare mineral suite, although these minerals are also indicative of the chemical constraints of the metamorphic reactions, especially the pressure of the CO_2. It is likely that the rarity of the suite of hydrated secondary minerals is related to the fact that they are dominantly lime-rich species which were formed under the unusual conditions of a long-continued high geothermal gradient near the plug.

The plug of Carneal (figure 5, plate 12) occurs on the south side of Glenoe Water at its confluence with Carneal Water, 1.2 km NNW of Carneal Bridge some 9 km NNW of Carrickfergus (J 389 959). The plug is roughly circular in plan, 150 m in diameter, adjacent to the Carneal Fault that trends NNW on its eastern side. It is probable that the fault bifurcates and occurs on both sides of the plug which is composed of olivine-dolerite and intrudes the Tertiary Lower Plateau Basalts of the Thulean province. The Carneal Fault is flanked by Chalk about 1 km to the south. The plug is located on the line trending about N 10° W through the Carrickfergus Castle Plug and the dyke-like Ballymoney Townland intrusion. Several other plugs in Northern Ireland occur on lines of parallel trend which probably define basement structures. The Carneal Plug also lies on the N 60° W, approximately linear trend through the Doonbought, Skerry Rock and Slemish intrusions (Patterson & Swaine 1957; Wilson 1972).

The general geology is described in the Geological Survey Memoir by Cobbing, Griffiths, Manning & Wilson (in preparation) in which the present author has provided a fuller account of the normal igneous rocks; brief reference has been made to the plug by Robbie (1960) and

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to some aspects of the mineralogy by Sabine (1968) and Henmi and others (1973). There are considerable similarities between the minerals found (and their paragenesis) and the suite described from Scawt Hill, also in Antrim, in the classic papers by Tilley (especially 1929*b*, and with H. F. Harwood, 1931).

(*a*) *Nomenclature of the metamorphic rocks, abbreviations and conventions*

In this work the two concepts of exomorphism and endomorphism are used in the sense in which they were originally introduced by Fournet (1847, p. 243), and especially as the corresponding adjectives. Thus the endomorphic rocks are the igneous rocks that are themselves modified in the process of metamorphism, sometimes very severely, and the exomorphic rocks are those produced from the country rocks which at Carneal are the chalk (limestone) and flint brought up as fragments by the intrusion, now represented by the assemblages including rare minerals. Numbers in parenthesis preceded by 'NI' or 'I' are those of thin sections in the Petrographical collections of the Institute of Geological Sciences. The identities of all the minerals here described have been confirmed or established by X-ray powder methods. However, the X-ray film numbers, given in parenthesis and preceded by 'X', are generally quoted in running text only when there might be doubt about the identification without the X-ray evidence. Where the results of proximate and similar chemical calculations are shown to two decimal places it is to avoid rounding errors, and high precision is not implied. Refractive index measurements are for sodium light. Pressure is quoted in the works cited in several different systems of units. To avoid confusion here, they have been converted to pascals*; the small differences between comparable units may be ignored for the purposes of this paper, but the original sources should be referred to for precise figures.

2. Mineralogy

The minerals may generally be assigned to the complementary exomorphic and endomorphic assemblages, but in the absence of recognizable field relations, or with the occurrence of enclaves not in equilibrium with the host rocks, this is not always so, particularly with the hydrated products.

The principal minerals of the exomorphic suite, including hydrated and alteration products, are as follows:

calcite	melilite	bicchulite
chalcedony	plagioclase	gyrolite
quartz	magnetite	portlandite
spurrite	chlorospinel	hydrogrossular (hibschite)
larnite	perovskite	tobermorite
(bredigite)	hematite	plombierite
wollastonite	sphene	scawtite
merwinite	xonotlite	calcium magnesiosilicate

Typical exomorphic assemblages are set out below. The main minerals are followed by the less common ones including alteration products which may be present; minor amounts of the other exomorphic minerals also occur.

* 1 pascal = 10^{-5} bar.

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larnite-spinel (chlorospinel, magnetite); hydrogrossular, spurrite, portlandite	derived from chalk
xonotlite-scawtite; tobermorite, hydrogrossular, melilite, calcite	
spurrite-merwinite-calcite-melilite; hydrogrossular, scawtite	
calcite-wollastonite; magnetite (marble)	
spurrite-calcite-melilite-spinel; xonotlite (marble)	
hibschite-calcite-spinel; plagioclase, tobermorite	
wollastonite-plagioclase-hydrogrossular; sphene, gyrolite, tobermorite	derived from flint
wollastonite; melilite, hibschite, bicchulite	
wollastonite-quartz; scawtite, xonotlite	
wollastonite-xonotlite	
quartz; calcite	

Among the minerals of the endomorphic suite the following are recorded

olivine	sahlite	sphene
plagioclase	nepheline	thomsonite
(albite-bytownite)	aegirine	(hydrogrossular)
augite	wollastonite	calcite
magnetite	marcasite	tacharanite
apatite	pyrrhotine	allophane
ilmenite	pyrite	gismondine
chlorite	melilite (humboldtilite)	tobermorite
olivine, fayalitic	merwinite	chabazite
titanaugite	scawtite	analcime

The endomorphic assemblages are

olivine-augite-plagioclase-magnetite (olivine-dolerite)	
sahlite-plagioclase; wollastonite, melilite, fayalitic olivine, thomsonite, calcite, tacharanite (pyroxenite and pyroxene-rich dolerite and gabbro)	derived from olivine-dolerite
titanaugite-melilite-plagioclase; magnetite, marcasite, thomsonite, tobermorite	
melilite; magnetite, wollastonite	
aegirine(-augite)-nepheline; apatite	
spurrite-merwinite-larnite-magnetite (metasomatized rock)	
merwinite-melilite-magnetite-hydrogrossular	metasomatized vein in dolerite

The phases in the system CaO–MgO–SiO_2 are shown in figure 1. Those in the system CaO–SiO_2–H_2O–CO_2 are shown in figure 2, revealing some of the progressive stages of decarbonation and hydration. Attention may be drawn to the clustering of the hydrous phases along the join wollastonite-water (with portlandite on the lime-water join).

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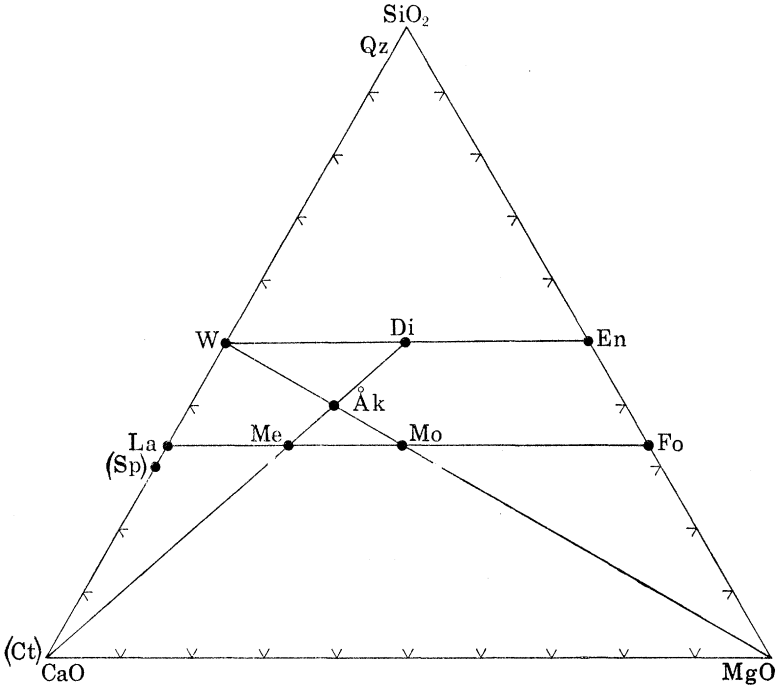

FIGURE 1. Phases in the system CaO–MgO–SiO_2 (after Bowen). Åk, åkermanite; (Ct), calcite (projected onto plane); Di, diopside; En, enstatite; Fo, forsterite; La, larnite; Me, merwinite; Mo, monticellite; Qz, quartz; (Sp), spurrite (projected onto plane); W, wollastonite.

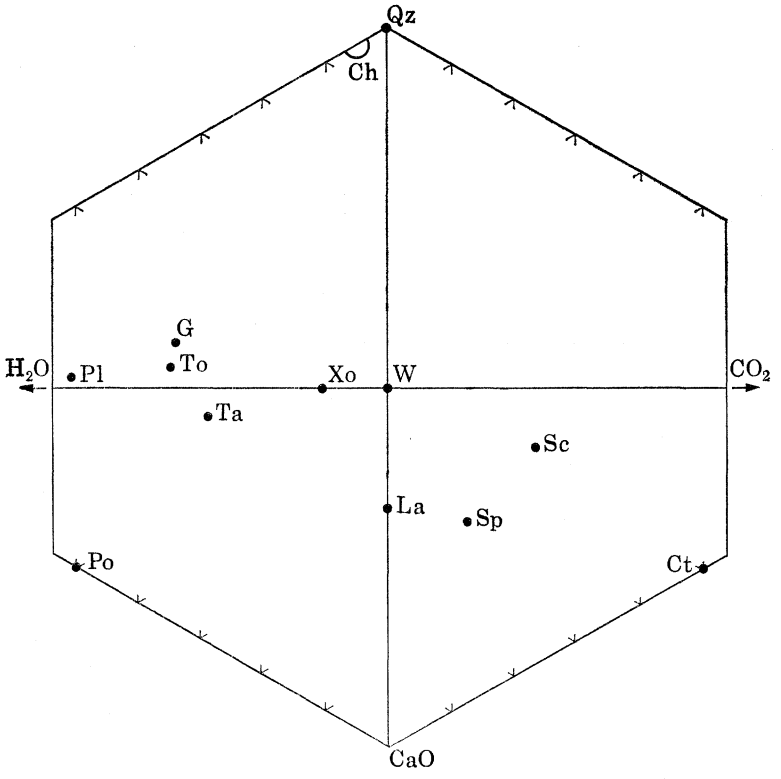

FIGURE 2. Phases in the system CaO–SiO_2–H_2O–CO_2. Ch, chalcedony (flint); Ct, calcite; G, gyrolite; La, larnite (bredigite); Pl, plombierite; Po, portlandite; Qz, quartz; Sc, scawtite; Sp, spurrite; Ta, tacharanite; To, tobermorite; W, wollastonite; Xo, xonotlite.

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(*a*) *Primary minerals of the reaction processes*

(i) *Pyroxenes*

In the endomorphic rocks two distinctive pyroxenes occur: titanaugite, mainly in melilite-titanaugite-rock, and sahlite in pyroxenite and pyroxene-rich hybrid dolerite and gabbro. There is also aegirine or aegirine-augite in small quantities in interstitial alkaline patches.

A. Titanaugite. This pyroxene is well developed in titanaugite-melilite-rock. It also occurs in much smaller quantity in hybrid pyroxenite as the pyroxene of late crystallization, and there is some also at the boundary between hybrid gabbro (in which the pyroxene is sahlite) and xonotlite-scawtite-rock. In the titanaugite-melilite-rock it forms deeply coloured hypidiomorphic prisms up to 5 mm or more in length moulded upon melilite and thomsonitized plagioclase, and also enclosed by the melilite. Hour-glass structure is only rarely present. The mineral shows strong pleochroism, and the colours in similar orientation may be variable (NI 1302). It has the following properties and dominant colours:

$\alpha = 1.734$; deep purplish vinaceous (69''' RV–R of Ridgway 1912) [purplish-brown],
$\beta = 1.746$; snuff brown (15''k Y–O) [brown],
$\gamma = 1.774$; pale brownish drab (9'''' OR–O) [pale greyish-mauve].

These figures may include some small range of values from within the crystals. 2V moderate, positive; $\gamma : c$ 53°. Absorption strong, $\alpha > \beta > \gamma$. Dispersion very strong $r > v$ on an optic axial section. Numerous (and markedly discrepant) schemes are quoted for both pleochroism and absorption of titanaugite from various sources, but these now presented agree reasonably well with those given by Tilley (in Tilley & Harwood 1931) for Scawt Hill and by Holzner in his careful study of German titanaugites (1934).

A sample was separated and analysed with the results shown in table 1, column I. The pyroxene is seen to be rich in alumina, ferrous iron, lime and titania, but low in silica and magnesia. The analysis may be recalculated in terms of standard molecules, as shown in the table. There is a small silica deficiency or excess of lime (0.47 %) which is revealed in the norm by the presence of nepheline (2.52 %). The norm also contains wollastonite, while most of the alumina enters anorthite (23.45 %). It has been pointed out by Tilley that lime- and alumina-rich augites are common in alkali-rocks containing feldspathoids, although the pyroxenes are not themselves rich in alkalis.

The atomic formula is given in table 1, column IA. Because of the tenor of aluminium, it is not necessary for titanium to enter the tetrahedral (Si, Al) group.

B. *Sahlite.* This pyroxene, black in hand specimen, occurs generally in hypidiomorphic to idiomorphic, commonly stout, prisms. In some rocks it shows weak pleochroism and in others the pyroxene has pale greenish to pale brown centres with deep greenish-grey or reddish brown margins, reflecting the advance of the process of assimilation. Some hour-glass zoning occurs. In many specimens the sahlite is zoned sharply to deeply coloured rims (e.g. NI 1574); the shades of colour prevent ready determination of the pleochroism at the rims, but the hue and probably also degree of absorption appear to remain the same as in the central parts of the crystals, so that the pleochroism is masked by the lower transmissivity.

The range of grain size is generally 0.3–2 mm but crystals may reach 1 cm or more across in the coarser pyroxenites (NI 1591); where the size is variable in the same rock the colour of the coarser prisms is commonly deeper at the margins and, as might be expected, this more

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TABLE 1. CHEMICAL ANALYSES AND COMPOSITION OF PYROXENES

	I	A	II	B	C
SiO_2	39.9	40.28	49.3	49.31	49.81
Al_2O_3	9.5	10.30	5.5	4.60	6.42
Fe_2O_3	3.9	5.35	2.1	2.38	1.25
FeO	12.9	7.92	4.2	5.57	1.03
MgO	4.6	7.78	13.2	14.02	15.22
CaO	22.7	23.57	24.7	22.71	25.92
Na_2O	0.55	0.36	0.21	—	—
K_2O	n.d.	tr	0.01	—	—
$H_2O > 105\,°C$	0.20	0.19	0.11	—	0.13
$H_2O < 105\,°C$	n.d.		n.d.	—	0.06
TiO_2	5.2	3.85	0.73	1.26	0.14
MnO	0.25	—	0.05	—	0.06
totals	99.70	99.60	100.11	99.85	100.19*
Cr(s)	tr		tr		
V(s)	n.d.		n.d.		
diopside	24.7	41.30	70.9	72.79	
hedenbergite	45.4	26.98	14.7	17.43	
acmite	4.1	2.74	1.6	—	
wollastonite	4.9	7.91	3.9	—	
$CaTiO_3$	8.9	6.46	1.2	—	
Al_2O_3	9.5	10.19	5.5	4.60	
Fe_2O_3	2.5	4.42	1.5	2.38	
remainder	—	—	0.7	2.65	

IA			IIA		
Ca	0.963	1.005	Ca	0.982	0.997
Na	0.042		Na	0.015	
			K	0.0004	
Mg	0.271	1.000	Mg	0.730	1.010
Fe^{+2}	0.427		Fe^{+2}	0.130	
Fe^{+3}	0.116		Fe^{+3}	0.058	
Ti	0.154		Ti	0.020	
Mn	0.008		Mn	0.002	
Al	0.024		Al	0.070	
Al	0.419	2.000	Al	0.170	2.000
Si	1.581		Si	1.830	
O	6.000		O	6.000	

(s), spectrographic determination; tr, trace; n.d., not detected; * $CO_2 = 0.15$.

I, Titanaugite. From melilite-titanaugite-rock. Quarry on east side of Carneal Plug. 1840 m. N 56° E of Ballyvallagh Bridge, Co. Antrim. NI 1302 (Lab. no. 1913. Anal. G. A. Sergeant; spectrographic work by K. L. H. Murray).

A, Titanaugite. From melilite-nepheline-dolerite, Scawt Hill, Co. Antrim (Tilley & Harwood 1931, p. 454).

II, Sahlite. From hybrid pyroxenite. Small pit on east face of Carneal Plug. NI 1681 (Lab. no. 1912. Anal. G. A. Sergeant; spectrographic work by K. L. H. Murray).

B, Pyroxene. From pyroxenite, Scawt Hill, Co. Antrim (Tilley & Harwood 1931, p. 451).

C, Diopside. From contact skarn, Kilbride, Skye (Tilley 1951*b*, p. 635).

IA, Titanaugite as column I, on basis of 6 oxygens.

IIA, Sahlite as column II, on basis of 6 oxygens.

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deeply coloured pyroxene occurs more commonly in association with wollastonite (e.g. NI 1612).

In rocks showing the effects of local metasomatism more than one species of pyroxene may occur in the various zones.

The sahlite (NI 1681) has the optical properties

α = 1.683; Glaucous, (29‴, GG–Y of Ridgway) [very pale green],
β = 1.698; Glaucous, (29‴, GG–Y) [very pale green],
γ = 1.717; Artemisia Green, (33⁗ GY–G) [grey-green].

Absorption, $\alpha < \beta < \gamma$. Dispersion strong, $r > v$ on an optic axial section. 2V moderate, positive; $\gamma : c$ 43°.

This pyroxene was separated and analysed with the results shown in table 1, column II.

The atomic constitution of the pyroxene is shown in table 1 column IIA. These figures are for the anhydrous composition. If hydroxyl is included the tetrahedral and other cationic groups also balance reasonably well.

In the triangle of Poldervaart & Hess (1951) and others the molecular proportions are $CaSiO_3$ 55.8, $MgSiO_3$ 35.8, $FeSiO_3$ 8.4.

In contrast to the titanaugite, the sahlite is moderately aluminous, even though there are some similarities between the two pyroxenes.

As shown in the recalculation to standard molecules, the sahlite consists dominantly of diopside-hedenbergite. Lime is still in slight excess (0.62 %) or there is corresponding silica deficiency; in the norm anorthite only amounts to 14.0 % and there are small quantities of leucite (0.05 %) and nepheline (0.96 %). A little forsteritic olivine appears and wollastonite, with which olivine is normatively incompatible, is of course absent.

(ii) *Melilite*

Melilite occurs in various parageneses, its composition reflecting the nature of the formational process. As the dominant variety humboldtilite it forms abundant coarse prisms with titanaugite in a dark hybrid rock that represents an advanced stage in the results of assimilation, while as a generally minor constituent in hybrid dolerites and related rocks, or in the exomorphic rocks, it is gehlenitic or rarely åkermanitic. Although the compositional range from humboldtilite to gehlenitic species is probably not large (and is reflected by the small range of refractive indices, ω, mainly from 1.641 to 1.650) the distinction is of some importance as an index of the degree of assimilation.

The humboldtilite prisms are up to about 25 mm in length, and enclose titanaugite as well as bytownite somewhat altered to thomsonite. The melilite (NI 1302) is practically colourless with a barely detectable pleochroism, ω very pale yellow, ϵ very pale pink. It is uniaxial negative and has the refractive indices ω = 1.641, ϵ = 1.632. The crystals have a stronger birefringence at their borders and some are more deeply coloured or pleochroic. Some marginal peg structure is present.

The mineral was separated by physical methods followed by hand picking and chemically analysed with the results shown in table 2.

The composition of the unit cell of melilite has been discussed by various authors, to whom reference is given by Louisnathan (1971) in a paper refining the crystal structure of gehlenite. If (OH) is accounted for as hydroxyl (balancing the substitution Na for Ca) the X and Y

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groups total 3.91_1 and 1.89_2 respectively. These figures are to be compared with 3.98_3 and 1.99_5 for the calculation without water, supporting the argument of Goldsmith (1948) that H_2O represents absorbed water and not hydroxyl. The figure for $H_2O > 105°$ for melilite from the comparable paragenesis of Scawt Hill is 1.59 %, among the highest recorded for any melilite, but the figure at Carneal reaches only 0.83 %.

TABLE 2. CHEMICAL ANALYSES AND COMPOSITION OF MELILITE

		I	A			IA		
SiO_2		41.60	37.84	Si	3.87		4.00	
Al_2O_3		7.60	12.89	Al	0.13	0.82		
					0.69			5.99
Fe_2O_3		0.64	1.13	Mn	0.008			
FeO		5.60	5.66	Ti	0.004		1.99	
MgO		5.85	3.12	Fe^{+3}	0.04			
CaO		34.15	33.19	Fe^{+2}	0.43			
Na_2O		3.20	3.93	Mg	0.82			
K_2O		0.07	0.25	K	0.008			
$H_2O > 105$ °C		0.83	1.59	Na	0.57		3.98	
$H_2O < 105$ °C		0.07	0.07	Ca	3.40			
TiO_2		0.06	0.66					
MnO		0.10	—					
totals		99.77	100.33					
Ba*		tr						
Sr*		n.d.						
gehlenite	$Ca_2Al_2SiO_7$	6.43						
åkermanite	$Ca_2MgSi_2O_7$	40.57						
ferro-åkermanite	$Ca_2Fe^{+2}Si_2O_7$	24.30						
sodium-melilite	$NaCaAlSi_2O_7$	27.34						
ferri-gehlenite	$Ca_2Fe_2^{+3}SiO_7$	1.36						

* spectrographic determination; n.d., not detected; tr, trace.

I, Melilite (Humboldtilite) from melilite-titanaugite-rock. Quarry on east side of Carneal Plug. 1840 m N 56 ° E of Ballyvallagh Bridge, Co. Antrim. NI 1302. Lab. no. 1910. Anal. G. A. Sergeant; spect. work by K. L. H. Murray.

A, Melilite (Humboldtilite) from melilite-titanaugite-rock. Scawt Hill, Co. Antrim (Tilley 1929*a*).

IA, Carneal melilite. Atoms on basis of 14 oxygens.

The refractive index (ω) agrees reasonably well with the calculated index 1.634 arrived at by using the indices for pure end-members given by Nurse & Midgley (1953). The refringence also agrees excellently with the test, rather coarse in this instance, of applying the Gladstone & Dale rules of specific refractive energies (Larsen & Berman 1934, pp. 30–1). The value calculated from the chemical analyses is 0.212; that from the determined refractive indices 0.213. It thus seems likely that no substantial mineral impurity was included among the chemical constituents.

On the basis of 70 oxygens, the Carneal analyses yield 19.9 (Ca, Na, K), and 30.0 (Si, Al, Mg, Fe, etc.). These are almost exactly the mean figures for melilite analyses originally provided by Berman (1929) and used by Smith (1953) for the construction of histograms.

In terms of mineral species the analysis (table 2) may be considered close to

åkermanite	64.87
gehlenite	6.43
pseudo-sarcolite	28.70

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In various hybrid or metasomatized gabbros, dolerites or pyroxenites, rocks that are the result of less advanced assimilation processes, melilite is present in smaller quantities; and with somewhat different derivation it also occurs in the exomorphic calcium-silicate rocks. In hybrid pyroxenite, optically uniaxial negative melilite, with $\omega = 1.642$ (NI 1607) occurs in small quantity forming coarse prisms. Another example (NI 1681), yielded $\omega = 1.648$, $\epsilon = 1.640$, uniaxial negative, nearer gehlenite than the analysed specimen.

Uniaxial negative melilite (gehlenitic) with $\omega = 1.650$, and low birefringence occurs as a plentiful though very fine-grained constituent of zones of a metasomatic vein in dolerite (I 1313). In a fine-grained melilite-rock (NI 1590) the mineral is also uniaxial negative, with $\omega = 1.649$, low birefringence; and cell dimensions $a = 7.724$ Å,† $c = 5.056_5$ Å, properties in accord with a gehlenitic species, a conclusion supported by X-ray powder data (film X3728). This melilite is partly altered to cebollite.

Melilite is a constituent of a variety of calcium-silicate-rocks. In a scawtite-xonotlite-rock (NI 1589), showing the effects of metasomatism, well-formed uniaxial negative tablets about 0.1 mm or less across set with wollastonite and magnetite have the refractive indices $\omega = 1.678$, ϵ about 1.670. These probably indicate a gehlenitic species with some iron content that accounts for the high refringence. A melilite (gehlenite) having a refringence distinctly above that of the humboldtilite, with ϵ somewhat greater than 1.660, was noted by Tilley (1929*b*) from the larnite-spurrite assemblages of Scawt Hill, particularly those with merwinite and spinel. In the Carneal rock, spinel is present, and there are pseudomorphs, probably after merwinite.

In a comparable rock (NI 1588) some melilite is probably uniaxial positive, corresponding to åkermanite, and showing that magnesium (from the dolerite) has been fixed in a metasomatic zone.

(iii) *Wollastonite*

Wollastonite is a common constituent in various rocks at Carneal, occurring in the normal anhydrous form, and also as a variety that displays abnormally low refringence and is probably hydrous. In its normal anhydrous form the wollastonite occurs in metasomatized flint that is represented by wollastonite-xonotlite, wollastonite-quartz or wollastonite-plagioclase assemblages. It is also commonly developed as prisms in hybrid pyroxenite, gabbro or dolerite, when it has both higher refringence and ferrous iron content. Wollastonite is present to a minor extent in other rock types. The lowly refringent wollastonite occurs in a wollastonite-rock with hibschite and other minerals. Associated with these various occurrences, apart from the other rock-forming minerals of the Carneal suite, are the hydrated silicates, xonotlite, gyrolite, tacharanite and tobermorite; and bicchulite (Henmi *et al.* 1973), previously only reported in synthetic assemblages but found almost simultaneously in Japan and at Carneal.

The wollastonite of the metasomatized flints has normal optical properties. In metasomatized flint (NI 1301) the wollastonite forms abundant stumpy prisms up to 0.03 mm long, having $\alpha = 1.622$, $\gamma = 1.633$ with simple twinning parallel to (100). The prisms are apparently rounded, but this is no doubt due to the development of subordinate pinakoids and terminal faces that sometimes characterize this species, but cannot be discerned readily in the very small crystals.

The wollastonite of the hybrid pyroxenites and related rocks is a ferruginous variety, with correspondingly somewhat higher refringence and also birefringence. The wollastonite from

† 1 Å = 0.1 nm = 10^{-10} m.

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a hybrid pyroxenite (NI 1681) was separated and analysed with the results shown in table 3, column I. It has the refractive indices $\alpha = 1.623$, $\beta = 1.632$, $\gamma = 1.638$. Closely comparable figures ($\beta = 1.631$, $\gamma = 1.640$) were found in wollastonite from a hybrid pyroxene-rich gabbro (NI 1612). The analyses may be computed (table 3, column IB) on the basis of 18 (O, OH), when the tetrahedral group = 6.00, octahedral group: 5.84; or on the basis of 18 oxygens, when the tetrahedral group = 6.00, octahedral group = 5.97 (column IA). The better correspondence of the two groups in this latter case, without (OH), suggests that water is not constitutional, and this fits with the relatively high refringence. The mineral contains a higher percentage of FeO, 1.8%, than is normally present in naturally occurring wollastonite, although iron-wollastonite with much higher FeO has been recorded from the contact skarn of granite and dolomite in Skye (Tilley 1948; table 3, column II), and the refringence and other characters of wollastonite from the hybrid titanaugite-bearing rocks at Scawt Hill suggest that it is high in $FeSiO_3$ (Tilley 1937).

TABLE 3. CHEMICAL ANALYSES AND COMPOSITION OF WOLLASTONITE

	I	II	IA			IB	
SiO_2	51.9	50.00	Si	5.95	6.00	5.89	6.00
Al_2O_3*	0.1	—	Al	0.05		0.11	
Fe_2O_3	0.3	nil		0.09		0.02	
FeO	1.8	9.29	Fe^{+3}	0.03		0.03	
MgO	1.0	nil	Fe^{+2}	0.17	5.97	0.17	5.84
CaO	44.8	38.86	Mg	0.17		0.17	
Na_2O*	n.d.	—	Mn	0.01		0.01	
K_2O*	n.d.	—	Ca	5.50		5.44	
$H_2O > 105\,°C$	0.5	nil	O		18.00	17.62	18.00
$H_2O < 105\,°C$	0.1		OH			0.38	
TiO_2	n.d.*	tr					
MnO	0.1*	1.22					
insoluble in HCl	—	0.45					
totals	100.6	99.82					
Ba*	**tr**						
Sr*	**n.d.**						
$CaSiO_3$	93.9	80.6					
$MgSiO_3$	2.5	—					
$FeSiO_3$	3.4	17.1					
$MnSiO_3$	0.2	2.3					

* spectrographic determination; n.d., not detected; tr, trace.

I, Wollastonite from hybrid pyroxenite. Small pit on east face of Carneal Plug. 1-in 29, 6-in 46NE. NI 1681 (Lab. no. 1911. Anal. G. A. Sergeant; spect. work by K. L. H. Murray).
II, Iron-wollastonite from hedenbergite skarn, Camas Malag, Skye (Tilley 1948).
IA, Carneal wollastonite. Atoms on basis of 18 O
IB, Carneal wollastonite. Atoms on basis of 18 (O, OH).

In the Carneal rocks the margins of the pyroxene are commonly deeply coloured, although many have centres that are pale, and the ferruginous wollastonite occurs in association with this pyroxene. The association of the more extreme iron-wollastonite with deeply coloured pyroxene was noted by Tilley (1948).

A pale grey wollastonite-rock (NI 1597) in which the mineral has abnormally low refringence, is composed of a compact aggregate of bladed or plumose to radiating prisms, the majority of which have closely interlocking margins showing mosaic texture. They show some lamellar

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twinning apparently parallel to (010). The size is very variable, the larger prisms reaching 1 mm in length. The smallest are difficult to define owing to granulitization but are commonly 0.02 mm across. The wollastonite poikiloblastically encloses other minerals including birefringent to isotropic highly refringent vermicular hibschite which occurs elsewhere in the rock. Also associated with the wollastonite are tobermorite, some xonotlite, carbonate (probably calcite), opaque spinel and scattered small crystals probably perovskite. The texture of the rock does not resemble that of the other (normal) wollastonite-rich types. The wollastonite has the refractive indices $\alpha = 1.606$, $\gamma = 1.616$, with $2V_\alpha$ very approximately 36°. This figure implies $\beta = 1.615$ but the angle is very sensitive to changes in refringence and from imprecision of the determinations no close correlation can be made.

Wollastonite associated with hibschite and having low refractive indices $\alpha = 1.609, \gamma = 1.624$, was described from western Georgia by Beliankin & Petrov (1939), who ascribed the low refringence to water content. The association of comparable wollastonite at Carneal with the presence of hydrogrossular and xonotlite lends support to the suggestion that the low refringence is due to the presence of (OH) in the structure. From the low refractive indices of the Georgian wollastonite, Beliankin & Petrov (1939) suggested that water was present to the extent of 0.5 % by mass. Using Tilley's figures (1922) for specific refractivity, and a mean refringence of 1.612, the Gladstone & Dale rules give a possible water content for the Carneal mineral of about 1.8 % by mass. Neither this figure nor that for the Georgian mineral can be very accurate in view of the several assumptions that must be made in the calculations, but the refringences and water contents are in reasonable relation with each other and with the figures for the anhydrous mineral.

(iv) *Larnite*

The larnite, (β-Ca_2SiO_4) is present mainly in larnite-spinel-rocks representing metasomatized chalk. It generally comprises a granular aggregate of crystals commonly 0.05–0.15 mm across, which may show marked preferred orientation. The larnite shows a prominent cleavage, very fine polysynthetic twinning, and has $\alpha = 1.707$, $\beta = 1.714$, $\gamma = 1.730$, which are almost identical figures to those given by Tilley (1929) in his original description of the mineral.

The larnite was separated from the rock by Mr J. Dangerfield by physical methods followed by hand picking. Chemical separation was not attempted since there was a possibility that it might affect the analytical results. Small quantitites of impurity, principally spinel, remained in the separated mineral and from point counts Mr Dangerfield estimated these to amount to about 2.1 % by volume, equivalent to 2.5 % by mass. Optical and X-ray evidence did not reveal any aluminous mineral contaminating the larnite, apart from spinel. From the analysis (table 4, column I) the amount of spinel, taken to have the composition of the analysed ferrian chlorospinel (table 4, column III) can be calculated at 1.4 %, and other spinel 0.6 %, leaving pure larnite as 87.8 %. In this allotment, the remaining alumina then builds 9.6 % gehlenite (in which the proportion of $CaO : SiO_2$ is 2 : 1, as in the larnite). Anything approaching this amount of melilite would certainly have been detected in the analysed rock. The available, although somewhat secondary, evidence thus points clearly to the occurrence of most of the aluminium in the larnite lattice. The composition of the unit cell of larnite has been shown to be $4[Ca_2SiO_4]$ by Midgley (1952) whose crystallographic data were refined by Cruickshank (1964). On this basis of 16 O the formula is shown in table 4, column IV.

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Table 4. Chemical analyses of larnite and spinel

	I	II	A	B	III
SiO_2	32.3	33.33	31.00	34.0 (±2.1)	1.4
Al_2O_3	4.5	4.29	1.12	0.02	42.2
Fe_2O_3	0.46	—	tr	0.011 (±0.003)	29.0
FeO	0.31	—	0.64	—	4.0
MgO	0.30	—	0.69	0.05	20.2
CaO	60.6	62.30	64.98	65.5 (±1.8)	2.5
Na_2O	0.15	—	—	0.25 (±0.06)	—
K_2O	0.04	—	—	0.037 (±0.015)	—
$H_2O > 105\,°C$	0.9	—	0.61	—	n.d.
$H_2O < 105\,°C$	0.3	—	—	—	0.03
TiO_2	0.10	—	—	0.029 (±0.019)	0.20
P_2O_5	0.12	—	—	0.48 (±0.07)	—
MnO	0.01	—	—	0.074 (±0.028)	0.60
CO_2	—	—	0.82	—	—
SO_2	—	—	—	0.057 (±0.005)	—
allow for minor constituents	0.09	—	—	—	0.15
SrO	—	0.08	—	—	—
totals	100.18	100.00	99.86	—	100.28
Ba*	<10 p.p.m.				—
Co*	—				130 p.p.m.
Cr*	—				80 p.p.m.
Cu*	—				50 p.p.m.
Ga*	—				15 p.p.m.
Ni*	—				460 p.p.m.
Sr*	650 p.p.m.				—
V*	60 p.p.m.				—
Zn*	—				400 p.p.m.
Zr*	—				20 p.p.m.

	IV	
Ca	7.58	7.93_6
Sr	0.006	
Al	0.35	
Al	0.22	4.00
Si	3.78	
O	16.00	16.00

* spectrographic determination; n.d., not detected; tr, trace.

I, Larnite. From larnite-spinel-rock, east face of plug, quarry beside laneway, Carneal, Co. Antrim. 1-in 29, 6-in 46NE. NI 1608A (Lab. no. 2024. Anal. G. A. Sergeant; spect. work by C. Park).

II, Analysis I, recalculated to remove impurities (principally ferrian chlorospinel, column III) to accord with structural formula.

A, Larnite, contact-rock, Scawt Hill, Co. Antrim (containing very small amounts of spurrite). Tilley, 1929*b*, p. 80.

B, Larnite, contact-rock, Scawt Hill, Co. Antrim. Electron probe analysis (Midgley & Bennett 1971). Figures in parentheses are stated to be one standard deviation.

III, Ferrian chlorospinel. From larnite-spinel-rock, as column I. NI 1608B. Lab. no. 2025. Anal. G. A. Sergeant; spect. work by C. Park (Sabine 1968).

IV, Larnite, corresponding to analysis, column II, on basis of 16 oxygens.

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(v) *Spinel*

The spinel, present principally in larnite-spinel-rocks, has already been described (Sabine 1968). It is finely divided, deep brown ferrian chlorospinel with $n = 1.86_2$, relative density (calculated from the analysis and cell size) 3.97, and unit cell dimension $a = 8.1905 \pm 0.0003$ Å. The analysis is shown in table 4, column III, yielding the composition

$$(Mg_{6.82}Fe^{+2}_{0.76}Mn_{0.12}Ni_{0.01}Zn_{0.01})\ (Al_{11.2}Fe^{+3}_{4.94}Ti_{0.03})O_{32}.$$

In two comparable rocks the cell size was 8.171 ± 0.001 Å reflecting a slightly different composition.

(vi) *Spurrite and merwinite*

These minerals occur in examples of the more highly metamorphosed rocks, and their properties appear to be unexceptional. Both occur in complex calc-silicate associations, representing metasomatized chalk, but spurrite is more commonly present in larnite-(spinel/magnetite-)rocks, poikiloblastically cementing the larnite prisms, as well as in a similar mineral association in metasomatized dolerite (NI 1593). Merwinite is also present as the principal mineral with melilite veining dolerite (I 1313).

(vii) *Calcium magnesiosilicate*

X-ray powder patterns matching those of the artificial compound $(2CaO.SiO_2)_{5.6}$ $(3CaO.MgO.2SiO_2)_{4.4}$ described by Gutt (1961) were obtained from a larnite-magnetite-rock (NI 1596). The optical properties of the artificial compound and of larnite are too close to allow ready determination of the possible natural magnesiosilicate occurring in a larnite matrix. In the light of this and the controversy about the components of the CaO–MgO–SiO_2 stability field, further work is in hand to establish whether the compound is present at Carneal as a natural mineral.

(*b*) *Hydrated and alteration products*

Just as the main mineral suites of the endomorphic and exomorphic zones are of rare types, so also are many of the minerals of the amygdales, and alteration products of the rock-forming minerals, notably tobermorite, tacharanite, and xonotlite, which are generally absent from basaltic amygdales, as well as allophane, bicchulite, gismondine, gyrolite, hydrogrossular, plombierite, portlandite, and thomsonite.

Their presence together must be more than coincidence and probably is due to the relatively long-continued high temperatures under which they were emplaced, as compared with those that exist during the zeolitization phase of basaltic magmatism, and also to the highly calcic nvironment.

(i) *Allophane*

This mineral has been found with tacharanite in amygdales up to 5 mm across in hybrid pyroxenite (NI 1591). It is a pale fawn apparently structureless mineral, isotropic, with refringence 1.476–1.480. Oval blebs less than 0.1 mm across of a pale brown mineral showing a tendency to original crystal outline are probably also allophane and may represent original hibschite. Allophane probably also occurs with gyrolite in amygdales in a complex wollastonite-feldspar-rock (NI 1595). The refringence of brown isotropic mineral interstitial to wollastonite

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is partly 1.492, partly 1.530. McConnell (1955*a*), accepted by Heller & Taylor (1956, p. 40), found a similar range of refringences (1.494–1.530), for unidentified (calcium) gel hydrates which may suggest a stable system of this composition range.

(ii) *Bicchulite*

A new mineral, the natural analogue of gehlenite hydrate synthesized by Carlson (1964), was discovered at Carneal by the present authors in 1972 and submitted to the Commission of New Minerals and Mineral Names. By extraordinary coincidence it had however also been discovered in Japan and submission to the Commission had been made a few days earlier by Henmi, Kusachi and Henmi. It was accordingly named by them, but through the good offices of the Chairman of the Commission, M. Fleischer, a joint description of the two occurrences has been published by the Japanese and the present authors (Henmi *et al.* 1973).

The mineral is body-centred cubic, colourless, with refractive index $n = 1.625$. Its ideal formula is $2CaO.Al_2O_3.SiO_2.H_2O$. At Carneal it occurs mixed with hibschite forming a buff-coloured coating to a wollastonite-rock (NI 1597).

(iii) *Gismondine*

The white rims of amygdales up to 1 cm across in olivine-basalt (NI 1615) are composed of gismondine. This mineral, which is biaxial, negative, with refractive index $\alpha = 1.522$ or slightly above, could formerly be confused with garronite but its X-ray pattern (X4545, A) and the optical properties (Walker 1962) are distinctive.

(iv) *Gyrolite*

This hydrated calcium silicate is present in amygdales up to 1 cm across in wollastonite-feldspar-rock (NI 1595) and it also occurs in the groundmass of the rock. It seems likely that it is an alteration product of the wollastonite. The mean refringence of the gyrolite is 1.515 but it is variable or in a mixture with some about 1.525. The birefringence is low. These properties are reasonably close to some published figures for the mineral. The refringence given by Tilley & Harwood (1931) for gyrolite from the closely comparable paragenesis of Scawt Hill, $\omega = 1.59$, is however markedly higher than that normally found for the mineral.

(v) *Hydrogrossular (hibschite)*

A fine-grained grey rock (NI 1305) is composed of hibschite ranging from idiomorphic equidimensional crystals to irregular aggregates of variable grain size (but some about 0.5 mm across) in a groundmass of isotropic to very weakly birefringent brown tobermorite, calcite, spinel, and other minerals. The hibschite (X4157) has $n = 1.672$ with some variation 1.667–1.680. The name is used here following Beliankin & Petrov (1939) and later authors for a member of the hydrogrossular series with refringence about 1.68, representing low Si and high (OH).

The surface of the hydrous wollastonite specimen (NI 1597) has a buff crust, perhaps a vein, 1–5 mm thick, of mixed calcium silicates, some hydrated. The base of this crust contains hibschite and melilite (X6447), the outer part melilite, hibschite and bicchulite (X6448). The hibschite has somewhat variable refringence mainly 1.675–1.679 but ranging from 1.671 to 1.682. Hydrogrossular is also associated with tobermorite as irregular turbid brown areas in a xonotlite-scawtite-rock (NI 1588). Cristobalite, with $\beta = 1.489$, was suspected in association

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with the hydrogrossular. This association was found by Mason (1957) in New Zealand contact-metamorphosed limestone. Hydrogrossular is also developed in association with other minerals in larnite-spinel-rock (NI 1606, 1609).

A vein of turbid brown nearly isotropic patchily variable material in pyroxene-rich dolerite (NI 1587) is hibschite showing many laminae, perhaps relict twinning (McConnell 1955*a*), in small areas commonly 0.1 mm across with very low birefringence and resembling kaolin books in some sediments. The refractive index is mainly $n = 1.64$–1.65, and the cell size is approximately 12.23 Å (X4214), but the absence of high angle reflexions prevented accurate determination. In a metasomatic merwinite vein in dolerite (I 1313), hibschite, with $n = 1.672$ cements melilite and merwinite.

(vi) *Plombierite*

Associated with the xonotlite in xonotlite-scawtite-rock (NI 1300) is the 14.6 Å calcium silicate hydrate that is identified as plombierite (McConnell 1954). The line at 14.6 Å is not very sharp. The mineral has not been distinguished optically with certainty among the various alteration products of the rock but is probably a colourless, apparently isotropic mineral in which scawtite is set. A mineral similar to and associated with tobermorite in an amygdale in basalt (NI 1299) and having the refractive index 1.552 may also be plombierite. This mineral was also detected from its X-ray powder pattern among the calc-silicates of metasomatized flint (NI 1678).

(vii) *Portlandite*

A somewhat bluish coating on the surface of larnite-spinel-rock (NI 1606) proves to be portlandite (X4795), the naturally occurring calcium hydroxide. It is uniaxial negative, with $\omega = 1.575$, strong birefringence, and yields a pronouncedly alkaline reaction with universal indicator paper (pH = 10.5 or more). In the rock it forms irregular patches with hazy extinction and there is a similar occurrence in another specimen (NI 1609) in which it encloses colourless isotropic nodules of roughly spherical shape, possibly lime. As in the original description of the mineral from Scawt Hill by Tilley (1933) the Carneal mineral arises by the hydration of larnite, since it is only on larnite-rock that it has been found.

(viii) *Tacharanite*

In hybrid pyroxenite (NI 1591) and in coarse olivine-basalt (NI 1615) angular amygdales up to 1 cm across contain an apparently structureless pale pinkish fawn mineral that proves to be tacharanite, margined by gismondine, and it also occurs in narrow irregular veinlets. In wollastonite-rock it is interstitial to the blades of wollastonite (NI 1595, 1678). It has the largest spacing at 12.85 Å, and does not expand with glycerol. The remaining pattern matches that of the Skye tacharanite (Sweet, Bothwell & Williams 1961) although the Carneal values are slightly lower. The mineral is isotropic and the refringence of the Carneal tacharanite is partly 1.530 but mainly 1.500–1.510, considerably lower than the type specimens, and presumably indicating its poorer crystallinity. Cliff, Gard, Lorimer & Taylor (1975) have examined the Carneal specimen by electron diffraction techniques and found that a large proportion of the material yielded no diffraction pattern.

It was suggested by Sweet that tacharanite (named from the Gaelic for a changeling) readily alters on exposure to air. The Carneal specimen was collected in 1961, and the X-ray

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film from which the tacharanite was identified was taken in 1966. A film repeated from the original powder still recorded the mineral $5\frac{1}{2}$ years later, $9\frac{1}{2}$ years after it had been collected. Specimen and powder had been kept in a heated, dry building, so the Carneal material is considerably more stable than the Skye mineral, perhaps owing to the presence of impurities giving a stabilizing effect in the lattice.

The mineral has been recorded by Koritnig (1972) as a metamorphic reaction product from quartzite inclusions in basalt associated with gyrolite and okenite from Germany, and has been briefly mentioned by Walker (1970) from Mull.

(ix) *Thomsonite*

This mineral is a very common alteration product in the endomorphic rocks: pyroxenite, pyroxene-rich dolerite and gabbro, melilite-titanaugite-rock and related types. The mineral, whose identity was confirmed by X-ray methods, may be recognized by its refringence, birefringence, and elongation $Y = c$. It is extensively developed replacing basic plagioclase, and also occurs as an amygdaloidal mineral in the endomorphic rocks and in the olivine-basalt. In an amygdale in an olivine-basalt (NI 1299) the thomsonite, of fibrous conical form, is central to tobermorite; it has the refringence $\alpha = 1.520$, $\gamma = 1.530$, which places it in the middle of the range of the composition of the mineral. A similar filling to amygdales is found in a pyroxene-rich metasomatized dolerite (NI 1304). Thomsonite fills some irregular amygdales around which the pyroxene is coarser (NI 1581) and in others spreads out as an irregular mesostasis in the rock (NI 1582).

In titanaugite-melilite-rock (NI 1302) and in hybrid gabbro (NI 1303) thomsonitized plagioclase (bytownite or labradorite) has the titanaugite moulded upon it; it commonly forms spherulitic growths. Melilite encloses the zeolite in hybrid pyroxenite (NI 1681).

There is considerable general chemical similarity between the composition of the members of the thomsonite series and plagioclase. The principal differences between the two series are the lower silica in thomsonite (about 16 %) and its considerably higher quantity of water (about 13 %); the silica difference is less if water differences are allowed for. The ratio of lime to soda is remarkably similar in labradorite and thomsonite ($CaO/Na_2O = 2.95$ in labradorite An_{62}, 3.02 in thomsonite, taking the mean of the four analyses in Deer, Howie & Zussman 1963), so that in agreement with the desilication and lime enrichment by which the rock suite is formed, thomsonite is a mineral to be expected on chemical grounds as the alteration product of the plagioclase. In the change, silica is released to take part in the general sequence of late contact metasomatic changes.

(x) *Tobermorite*

This mineral occurs in small (5 mm) round amygdales in basalt (NI 1299) surrounding thomsonite. It is white in hand specimen, very pale fawn or colourless in thin section, although sometimes turbid, apparently isotropic with variable refringence around 1.565 and gives the basal spacing 11.4 Å. An X-ray film taken in a normal powder camera gives a sharp, strong, 002 line but all the other lines are weak, in distinction from published values for the mineral. All the other tobermorite specimens tested yielded similar results with 11.4–11.5 Å as the 002 line. The refringence is lower than the normal value for the mineral (1.570–1.575) and there are large numbers of submicroscopic inclusions. In scattered amygdales that reach 10 mm across in melilite-titanaugite-rock (NI 1302) tobermorite is present with calcite, and in pyroxene-

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rich dolerite (NI 1304) it is marginal to thomsonite. The mineral was deposited centrally to drusy calcite in veins cutting hibschite-calcite-spinel-rock (NI 1305). As in the other thin sections, it is turbid brown by transmitted light, white by reflected light. It also occurs interstitially or veining pyroxenites. In a complex xonotlite-scawtite-(hibschite)-rock (NI 1588) tobermorite forms veins with xonotlite and also occurs in small brown patches associated with the hibschite, and with tacharanite and allophane in hybrid pyroxenite (NI 1591). Tobermorite forms veins and material interstitial to wollastonite in wollastonite-rock (NI 1678) in which plombierite also occurs.

(xi) *Xonotlite*

Xonotlite occurs forming the groundmass of complex calc-silicate assemblages, or veining them or other rocks. In xonotlite-scawtite-rock the xonotlite, white in hand specimen, is colourless or very pale buff in thin section. It appears to be almost isotropic, giving a speckly depolarization, with mean refractive index 1.575 (NI 1300). The mineral is cryptocrystalline and the apparently low birefringence may be due to this extremely fine grain size. In a wollastonite-xonotlite-quartz-rock, representing metasomatized flint (NI 1301) the xonotlite forms radiating, fibrous, lowly birefringent veinlets with some spherulitic crystallization that spreads out to form the host of the wollastonite in the rock. Much of the xonotlite forms coronas around wollastonite prisms. The modal composition of the rock and the nature of the xonotlite are discussed below.

The white part of a complex rock (with scawtite, hydrogrossular, tobermorite and other minerals; NI 1588A) is mainly xonotlite which is biaxial positive with small optic axial angle. Veinlets of xonotlite are also present traversing scawtite-rock (NI 1589) and in larnite-rock (NI 1592, 1608). Wherever xonotlite is found at Carneal the host-rock is one of the more extreme lime-rich types and it may be concluded that the veins are exudations that have their origin very locally and do not persist in other parageneses. In other occurrences some xonotlite is present however in different environments, as reported, for example, by Coombs & Lauder (1960).

3. Exomorphic assemblages

Scawtite-, spurrite-, merwinite-rocks (with or without hydration products), representing metamorphosed and metasomatized chalk are not well represented in the Carneal rock suite. There are however sporadic occurrences of larnite-spinel assemblages, well-developed examples of metasomatized and metamorphosed flint, and a few examples of the other assemblages also occur.

(*a*) *Xonotlite-scawtite-rock*

In a white rock adjacent to hybrid gabbro (NI 1300), nodules of spherulitic scawtite, commonly 0.1 mm across, are set in glassy-looking xonotlite and plombierite. The scawtite has $\alpha' = 1.592$, $\gamma' = 1.622$ with a fibrous structure which makes refractive index determination difficult. Identification was confirmed by X-ray methods. The nodules of scawtite occur in clusters that spread out from cracks (figure 6, plate 13) and the mineral is also present in discrete radiating crystals. Some abuts against very narrow veinlets and aggregates of calcite. There are clusters of tiny (1 μm) strongly refringent granules possibly spinel.

In other examples of similar assemblages (NI 1588, 1589), the effects of metasomatic zoning are more noticeable. Xonotlite, white in hand specimen, forms a patchy crystal aggregate of

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approximately 0.1 mm grain size, enclosing discrete crystals and veins and aggregates of scawtite. Veining by mixed tobermorite and xonotlite is present. Melilite is commonly present enclosing abundant opaque spinel and margined by fine-grained calcite. The scawtite, dark grey in the specimens, encloses abundant magnetite and is associated with hydrogrossular. The magnetite is also concentrated around melilite-rich parts of the rock. There are some pseudomorphs after merwinite: cristobalite, anorthoclase and heulandite are probably all present but this has not been confirmed.

(*b*) *Larnite-spinel-rock and related assemblages*

A rare rock type that is nevertheless one of the commoner examples of the extreme silica metasomatism of chalk is larnite-spinel-rock, in which the spinel is ferrian chlorospinel or magnetite (NI 1592, 1596, 1606, 1608–9). The rocks are pale to dark grey with a bluish tinge. The larnite forms a rather granular aggregate of prisms mainly 0.1 mm or less in length showing some preferred orientation. Idiomorphic ferrian chlorospinel may be abundant, and magnetite commonly up to 0.5 mm in cubes and aggregates is also present, some moulded upon larnite. Spurrite poikiloblastically encloses larnite prisms, which are less densely packed in this mineral, and there are minor amounts of melilite, hibschite, perovskite, and calcite. The rocks are traversed by narrow veinlets of xonotlite, and portlandite. The mineral that corresponds to the artificial compound $(2CaO.SiO_2)_{5.6}(3CaO.MgO.2SiO_2)_{4.4}$ prepared by Gutt (1961) was also detected.

The chemical analysis of the rock is shown in table 5, column III.

(*c*) *Other complex associations*

Some metasomatized chalk consists of highly complex associations, typically containing merwinite in well-developed prisms cemented by coarse spurrite which encloses calcite. Gehlenitic melilite is enclosed by merwinite and calcite; hydrogrossular forms equidimensional areas and there is a little scattered ore mineral. Larnite, spinel, perovskite and scawtite may occur and hydrous minerals include xonotlite (I 1296, 1299, 1303). The minerals are typically full of inclusions and the rocks may be metasomatically banded.

In one instance (NI 1593) the larnite-spurrite-spinel association with other minerals occurs in a complex rock that probably represents completely metasomatized dolerite.

(*d*) *Metasomatized equivalents of flint*

The metasomatic introduction of lime into flint, or its thermal metamorphism with included chalk, produces several related mineral assemblages. These consist principally of wollastonite, quartz, and hydrogrossular (hibschite) with common hydrated calcium silicates, especially xonotlite. There are lesser amounts of melilite, sphene, plagioclase, orthoclase, gyrolite, tobermorite, plombierite, bicchulite, and tacharanite, as well as small quantities of scawtite, merwinite, larnite, spurrite, calcite, apatite, pyrite, pyrrohotine and magnetite, probably epidote, and probably thomsonite. Okenite has been recorded in comparable rocks by Tilley & Alderman (1934) although they noted some discrepancies in the optical data.

(i) *Wollastonite-plagioclase-hydrogrossular-rock*

The rocks still macroscopically resemble flint (figure 3, plate 12) and consist of carious-weathering, compact fine-grained types. Angular areas, in shades of grey, are largely quartz,

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which is veined by paler wollastonite and white material, mainly xonotlite, and there are fawn and greenish areas that contain hydrogrossular and are pigmented by small quantities of sphene. The angular areas are generally 0.5–5 cm across. In *wollastonite-plagioclase-hydrogrossular-rock* (NI 1595, 1678) the grey part of the rock contains the coarsest part, composed of wollastonite, partly fine-grained, but partly in prisms up to several millimetres long with interstitial uniformly brown mineral, which is white in reflected light and which is mainly tobermorite. In other sections the interstitial mineral is tacharanite, with mean refringence 1.530, and barely detectable microspherulitic texture; and allophane, with $n = 1.492$ is probably also present (NI 1595). Some of the wollastonite is cemented by xonotlite.

In the fawn part of the rock the colour is due to sphene (confirmed by electron-probe analysis by Mrs A. Tresham) which occurs in abundant granules and euhedra that are commonly 0.02 mm long. In the green part of the rock, plagioclase was identified by X-ray methods.

TABLE 5. CHEMICAL ANALYSES OF EXOMORPHIC ROCKS

	I	II	III
SiO_2	64.29	43.64	28.47
Al_2O_3	1.56	12.42	7.87
Fe_2O_3	0.01	0.11	3.63
FeO	0.25	0.42	0.60
MgO	0.48	0.43	2.31
CaO	27.50	32.51	53.31
Na_2O	0.32	1.83	0.15
K_2O	0.11	0.11	0.06
$H_2O > 105\,°C$	3.03	5.26	1.22
$H_2O < 105\,°C$	1.55	1.00	0.55
TiO_2	0.11	0.99	0.11
P_2O_5	0.22	0.36	0.15
MnO	0.07	0.08	0.10
CO_2	0.48	0.40	1.36
FeS_2*	0.13	0.30	0.09
allow for minor constituents	0.05	0.10	0.10
totals	100.16	99.96	100.08
	parts per million		
Ba(s)	< 10	10	< 10
Co(s)	< 10	< 10	< 10
Cr(s)	< 10	< 10	< 10
Cu(s)	60	22	< 10
Ga(s)	< 10	18	10
Li	n.d.	n.d.	10
Ni(s)	< 10	< 10	60
Sr(s)	90	400	600
V(s)	54	50	60
Zr(s)	< 10	80	< 20
B	36	51	3
F	140	150	70

* total S calculated as FeS_2; (s), spectrographic determination; tr, trace; n.d., not detected.

I, Wollastonite-quartz-rock. West face of Carneal Plug, 55 m from wall, 1740 m N 56 ° E of Ballyvallagh Bridge, Co. Antrim. 1-in 29, 6-in 46NE. NI 1679 (Lab. no. 1968. Anal. W. H. Evans; spect. work by C. Park).

II, Wollastonite-plagioclase-hydrogrossular-rock. West face of Carneal Plug, 73 m from wall, 1735 m N 57 ° E of Ballyvallagh Bridge, Co. Antrim. 1-in 29, 6-in 46NE. NI 1678 (Lab. no. 1967. Anal. W. H. Evans; spect. work by C. Park).

III, Larnite-spinel-rock. East face of Carneal Plug, quarry beside laneway, 1840 m N 56 ° E of Ballyvallagh Bridge, Co. Antrim. 1-in 29, 6-in 46NE. NI 1608 (Lab. no. 2021. Anal. G. A. Sergeant; spect. work by C. Park).

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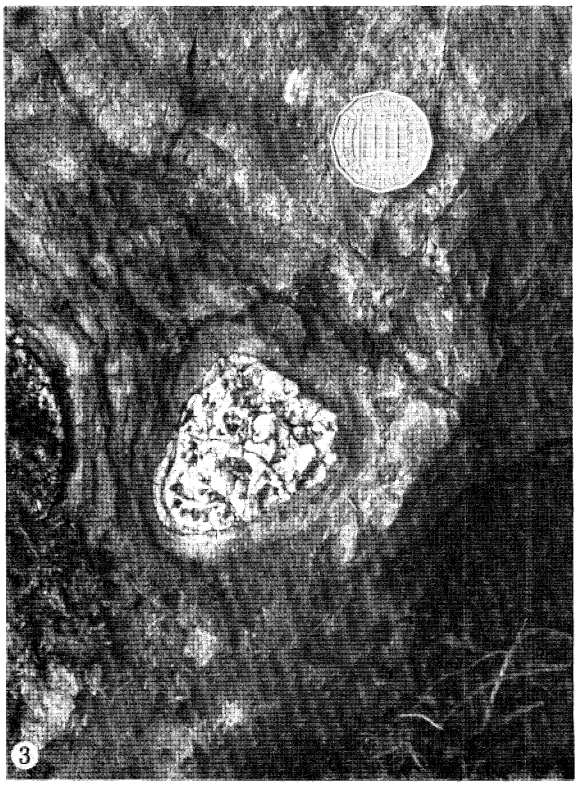

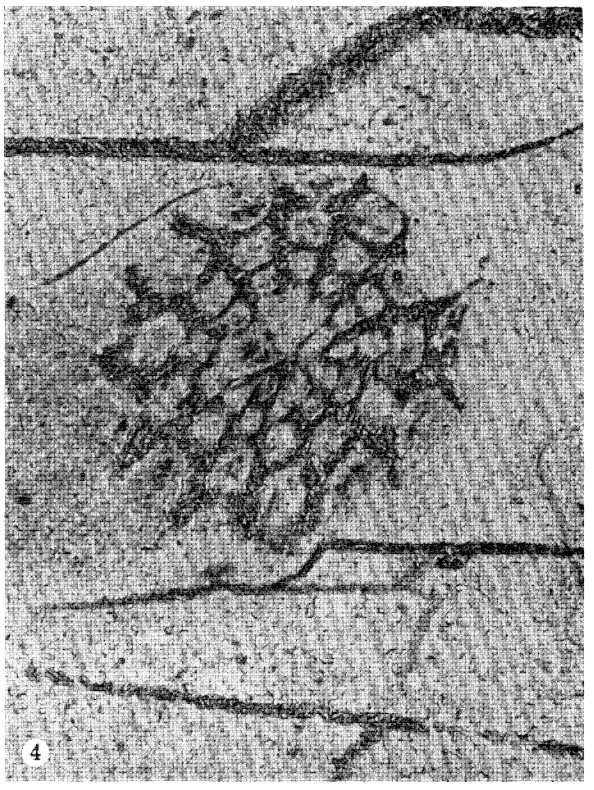

FIGURE 3. Reaction rims around metamorphosed flint nodules. N. face of Carneal plug. The coin measures 20 mm in diameter. Negative NI 349.

FIGURE 4. Cyclostome bryozoan now composed of wollastonite, in metamorphosed flint. NI 1301A. Magn. × 48. Negative M2824.

FIGURE 5. The Carneal Plug seen from the north across an alluvial flat. The west face of the plug is visible among the trees on the right. Negative NI 347.

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It is granulitized and is present in intimate fine-grained intergrowth with the other minerals. Irregular patchy hydrogrossular is present with inclusions of the other minerals. There are rare trains of magnetite. Gyrolite is present in coarse white patches or amygdales, and some gyrolite is mixed with feldspar. Gyrolite is one of the few calcium silicate hydrates in which silica is in molecular excess over lime, and this accounts for the presence of the mineral in a rock in which silica was initially predominant. Among other hydrates a little plombierite (15 Å) was detected. Okenite was not found. There is some scawtite present, and a small area of mixed exomorphic minerals inclusive of spurrite is presumably a small chalk enclave.

The rock has been analysed with the results shown in table 5, column II.

(ii) *Wollastonite-quartz-rock and wollastonite-xonotlite-rock*

In wollastonite-quartz-rock, (NI 1679) chalcedony is now represented by very fine-grained quartz that forms a mesostasis to abundant, mainly fine-grained, wollastonite. The quartz forms a dark to medium grey mosaic of crystals from about 0.05 mm down to about 0.005 mm that show some preferred orientation. This aggregate is veined by calc-silicates, principally pale grey wollastonite, mainly fine-grained but partly as a coarse bladed aggregate together with larnite and merwinite, and also interstitial hydrated calcium silicates, largely xonotlite.

Leucoxenized sphene forms very fine granules or euhedra and there is some fine lath-like twinned plagioclase apparently labradorite zoned to oligoclase. Small quantities of larnite, merwinite, and melilite can occur in enclaves.

The rock has been analysed with the results shown in table 5, column I.

A rock which has the general appearance of flint (NI 1301), is partly composed of white wollastonite-xonotlite-rock, and partly of a grey mosaic of fine-grained quartz aggregate traversed by an angular mesh of wollastonite and wollastonite-xonotlite veins filling fractures in two main planes at about 70° to each other, and running into the wollastonite-xonotlite-rock.

(iii) *Cyclostome bryozoan preserved in wollastonite*

In the quartzitic part of the metasomatized flint, an organism is now preserved in wollastonite. The organism (figure 4, plate 12), set in a fine aggregate of quartz representing recrystallized chalcedony, is approximately circular (in cross section) measuring about 1 mm diameter, and preserves the fine detail of the original morphology but not the ultrastructure of the organism. Mr C. J. Wood and Dr A. W. Medd suggest that the organism was a cyclostome bryozoan, so the thin section represents an oblique slice of a cylindrical object. It is composed of a dispersed to dense array of wollastonite prisms forming radial and concentric structures which roughly define 'cells' (zooecia) that are 0.05–0.1 mm across. There is a suggestion of bilateral symmetry to the organism. The prisms of wollastonite, originally in the place of zooecial walls, are smaller towards the centre (where they are about 0.003–0.01 mm across) than at the margins of the organism (zooarium) where they are mainly 0.015–0.06 mm long. They show some preferred orientation parallel, or perpendicular, to the zooecial walls which reach 0.05 mm at the margins but only 0.01 mm or less towards the centre. The quartz aggregate is generally finer grained within this central area. All the original structure has been replaced by the wollastonite. In Devonian rugose corals replaced by wollastonite described by Morgan, Perrott & Petherbridge (1971) the corals consist of massive wollastonite and all detailed structure has been obliterated. The authors concluded that the metamorphism, although revealing a high

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temperature (at least as high as in the hornblende–hornfels facies) had not operated by metasomatic activity. In contrast, the Carneal bryozoan preserves the coarser zooecial morphology of the organism and shows a true metasomatic origin, albeit perhaps on a very small scale. The silica by which the original calcite was converted to wollastonite came from the abundant surrounding flint. That the microscopic structure has been but little disrupted by the metamorphism shows that the metasomatism was essentially a tranquil process.

With the Quantimet analyser, Dr R. Dearnley found that it was not possible to measure the exact percentage of wollastonite in the wollastonite-xonotlite-rock (despite the use of filters) because of detection problems against the background, but he estimated the figure to lie between 30 and 35%. From three published analyses of xonotlite, in which H_2O+ was determined separately, the mean, 2.96%, agrees well with the 3.0% of the theoretical composition $5(CaSiO_3).H_2O$. It is also commensurate with modern analyses in which total water has been determined and accords with dehydration studies by Buckner, Roy & Roy (1960). From structural considerations, Mamedov & Belov (1956) conclude that the formula is $6(CaSiO_3).H_2O$. The figure quoted by Dana (5th edition, 1875), 3.73% H_2O agrees with $4(CaSiO_3).H_2O$, and is not apparently borne out by modern analyses. Taking therefore the H_2O+ figure at 3.0%, and a normal wollastonite content in the wollastonite-xonotlite-rock of 33%, the rock is equivalent to a monomineralic wollastonite-rock containing 2.0% H_2O+. This is in reasonable agreement with the 1.8% calculated for the hydrous wollastonite. It thus seems likely that from the same initial composition, either wollastonite-xonotlite-rock or monomineralic hydrous wollastonite could result.

(iv) *Other assemblages*

The wollastonite-rock (NI 1597) described above, in which wollastonite has abnormally low refringence, and on which bicchulite occurs, may also represent metasomatized flint.

Flint converted to quartz mainly in crystals 0.05 mm or less across, enclosed by an association of hibschite, tobermorite (11.5 Å), calcite, a little plagioclase and probably perovskite (NI 1305) has also been referred to above.

(*e*) *Marble*

One example of metamorphosed chalk (not converted to silicate) is a pale grey foliated wollastonite-marble (I 1301; 101929 in the Cambridge collection). It is composed of an aggregate of calcite up to 3 mm across enclosing idiomorphic magnetite and wollastonite. The latter was originally in prisms up to 1 mm long but is now a mosaic of angular grains commonly 0.01 mm across, some showing spherulitic crystallization. There is a trace of perovskite, very small granules that are probably hypersthene, and some more highly refringent carbonate. Some afwillite may be present. The rock is veined by calcite. Some spurrite-bearing rocks could also be considered as marble.

4. Endomorphic rocks

Aside from the olivine-dolerite itself, endomorphic rocks consist mainly of pyroxene-rich dolerite and pyroxenite. As the reaction becomes more extreme, the striking titanaugite-melilite-rock, and limited occurrences of melilite-rock, and aegirine- and nepheline-bearing types appear.

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(*a*) *Olivine-dolerite*

The normal igneous rock, described more fully in an account in preparation (in Cobbing *et al.*), is a typical Antrim olivine-dolerite, a non-porphyritic type with grain size that ranges well down into basalt. The chemical analysis and norm are shown in table 6, column I.

(*b*) *Pyroxenite and pyroxene-rich dolerite*

The hybrid pyroxenites are black amygdaloidal rocks in which sahlite is associated with wollastonite, melilite, coarse opaque ore, thomsonite and calcite, as well as hydrated alteration products including tobermorite, allophane, and tacharanite that occur in the amygdales or in the groundmass.

The pyroxene is of variable grain size, generally idiomorphic or hypidiomorphic (figure 9, plate 13). The coarser crystals reach 7 mm but are commonly much smaller. There is some zoning (figure 7, plate 13), including partial hour-glass structure. In one example (NI 1582) the finer prisms are arranged to form a radiating texture. Olivine is generally absent in the pyroxenites. In some rocks the coarser pyroxenes poikiloblastically enclose the other minerals, especially thomsonite and ore; and some pyroxene has a granulated appearance. Scarce labradorite prisms are mainly 0.7 mm or less in length: they occur between the pyroxene prisms that abut against them. The interstitial material, present generally in small quantity, is commonly thomsonite which occurs also in the amygdales, around which the pyroxene may be coarser with well-developed crystal faces projecting into them. Sphene is present in scarce idiomorphic crystals. There are some patches containing aegirine and some highly altered mineral probably represents nepheline. A little alkali-feldspar is present. In some rocks, fine granules of higher refringence than the sahlite may be hypersthene. Patches of titanaugite-melilite-rock occur. Opaque ore is not common, in distinction from the Scawt Hill suite. It includes magnetite, pyrite and pyrrhotine. In a rock showing local metasomatism, bredigite is present.

Amygdales are typically present but generally only in small numbers. They are commonly round, equidimensional, 1–5 mm across, but many are of angular shape and some of irregular shape merge into the groundmass. The largest reach 1.5 cm across. Some calcite occurs interstitially to the sahlite, melilite and wollastonite of the rock: the final filling to the amygdales is also sometimes calcite but other amygdales have thomsonite as a central filling to tobermorite containing strings of small crystals probably calcite, and in some veins calcite occurs marginally to (probable) tobermorite.

The pyroxenites are common immediately adjacent to calc-silicate rocks; in places the pyroxenite composition varies to pyroxene-rich dolerite or, occasionally, pyroxene-rich gabbro (figure 8, plate 13; table 6, column III). These rocks have the same general characters as the pyroxenites but abundant prisms of sahlite, commonly up to about 2 mm long have plentiful plagioclase (labradorite-bytownite, An 63–78 but with some zoning to andesine) moulded upon them. A little olivine occurs with the refractive index $\alpha = 1.745$ (NI 1587) indicating a fayalitic mineral of composition about $Fa_{55}Fo_{45}$ (hortonolite). The X-ray powder evidence is consonant with this composition which arises from the increasing tenor of ferrous iron. Similar secondary minerals to those of the pyroxenite are present. In one rock a vein is composed of hydrogrossular.

A pyroxenite was analysed with the results shown in table 6, column IV. Owing partly

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TABLE 6. CHEMICAL ANALYSES OF OLIVINE-DOLERITE AND ENDOMORPHIC ROCKS

	I	II	III	IV
SiO_2	47.62	40.70	42.12	43.85
Al_2O_3	17.39	14.12	11.13	11.82
Fe_2O_3	2.90	1.68	4.78	5.26
FeO	7.12	8.86	5.63	6.30
MgO	8.58	2.21	5.88	6.67
CaO	11.64	23.22	23.65	19.99
Na_2O	2.03	2.54	0.79	1.20
K_2O	0.20	0.49	0.11	0.29
$H_2O > 105\,°C$	0.62	2.58	2.63	1.83
$H_2O < 105\,°C$	0.88	0.62	0.87	0.70
TiO_2	1.00	1.85	1.60	1.62
P_2O_5	0.15	0.33	0.16	0.17
MnO	0.17	0.21	0.15	0.19
CO_2	n.d.	0.29	0.54	0.12
FeS_2*	—	0.20	0.10	0.13
allow for minor constituents	0.14	0.17	0.21	0.14
totals	100.44	100.07	100.35	100.28
		parts per million		
Ba(s)	60	180	< 10	< 10
Co(s)	35	< 10	25	25
Cr(s)	< 10	< 10	< 10	30
Cu(s)	140	180	780	173
Ga(s)	25	25	25	22
Li	n.d.	20	10	25
Ni(s)	200	30	230	85
Sr(s)	100	310	90	190
V(s)	250	320	300	300
Zr(s)	10	90	60	100
B	5	7	8	7
F	140	375	245	220
S	tr	—	—	—
norms				
or	1.18	—	—	1.71
ab	17.18	—	—	3.41
an	37.75	25.68	26.50	26.01
lc	—	2.27	0.51	—
ne	—	11.64	3.62	3.67
di	15.34	34.84	39.16	45.04
wo	—	3.40	13.92	6.29
hy	14.62	—	—	—
ol	6.28	—	—	—
cs	—	11.30	1.22	—
mt	4.20	2.44	6.93	7.63
il	1.90	3.51	3.04	3.08
ap	0.35	0.76	0.37	0.39
cc	—	0.66	1.23	0.27
pr	—	0.20	0.10	0.13

* total S calculated as FeS_2; (s) spectrographic determination; tr, trace; n.d., not detected.

I, Olivine-dolerite. Top of plug in centre of intrusion, Carneal, 1800 m N 54° E of Ballyvallagh Bridge, Co. Antrim. NI 1605 (Lab. no. 1966, Anal. G. A. Sergeant, spect. work by C. Park).

II, Titanaugite-melilite-rock. Quarry on east side of Carneal Plug. 1840 m N 56° E of Ballyvallagh Bridge, Co. Antrim, NI 1302 (Lab. no. 1963).

III, Wollastonite-bearing hybrid gabbro. Locality as I, NI 1303 (Lab. no. 1964).

IV, Hybrid pyroxenite. West face of plug, 9 m south of fence at top of bank. 1740 m N 54° E of Ballvallagh Bridge, Co. Antrim, NI 1582 (Lab. no. 1965).

II–IV, Anal. P. R. Kiff and G. A. Sergeant; spect. work by C. Park.

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to the presence of plentiful thomsonite the norm and mode are not very similar, especially in the paucity of fresh modal plagioclase. Some of the general characters of the rock are however discernible in the norm, for example the abundant diopside, the alkaline character, and the presence of wollastonite. The differences are due partly to the presence of plentiful modal thomsonite, and some of the alumina that forms normative anorthite is present in the sahlite.

(*c*) *Titanaugite-melilite-rock*

This very rare rock type is medium to dark grey in hand specimen, coarse grained, with scattered amygdales. It is composed (NI 1302) of a hypidiomorphic to idiomorphic aggregate of tablets of melilite, reaching 3 cm, and prisms of titanaugite 5–10 mm long. Tablets of bytownite (An_{85}) are common, some intergrown with melilite. There is a plentiful interstitial alkaline residuum, including somewhat skeletal green sodic and purple pyroxene and small amounts of opaque ore and apatite as well as feldspathic or feldspathoidal material. The titanaugite is mainly crystallized peripherally to and moulded upon the melilite tablets, although both minerals enclose each other and also enclose thomsonitized plagioclase: the melilite has mainly been the first mineral to crystallize. A very small amount of wollastonite is probably present. Nepheline is represented by hydronepheline. The plagioclase is partly altered to thomsonite to which chemically the bytownite shows considerable resemblance. Iron ore, present largely in the alkaline residuum, is mainly ilmenite. It occurs in homogeneous hypidiomorphic crystals and aggregates, but much is skeletal. Rare corroded round twinned yellowish grains that are strongly anisotropic prove to be of pyrrhotine, which on electron-probe analysis by Mrs A. Tresham yielded as the mean of five determinations:

Fe	60.5 ± 0.7
Ni	0.32 ± 0.03
Cu	0.06 ± 0.02
S	38.9 ± 0.2
total	99.78

This gives the formula $FeS_{1.12}$ (almost exactly Fe_8S_9) with $Ni_{0.005}$, $Cu_{0.0009}$.

Some pyrrhotine is moulded on ilmenite but some has cracks filled by ilmenite.

The scattered amygdales occur in planar zones and reach 10 mm across but are mainly much smaller, and many are indistinguishable from alteration products of the mesostasis. The largest tend to be of rectangular outline and are mainly filled by tobermorite; many of the smaller are rounded.

The melilite (humboldtilite) and titanaugite have been analysed and are described above. The rock has also been analysed with results shown in table 6, column II.

(*d*) *Melilite-rock*

Only one occurrence of this rock type has been found, as an enclosure in pyroxenite. It is a flinty-looking rock (NI 1590) composed of fine-grained tablets of gehlenitic melilite mainly less than 0.05 mm across but showing by mosaic polarization a finer constituent grain size, set in a groundmass of similar material rich in magnetite. There are drops of highly refringent material probably larnite, a little wollastonite and veinlets of calcite. Merwinite and cebollite were suspected but not proved.

This rock contrasts markedly in its fine grain size with rocks of related composition at Scawt Hill, and is perhaps best considered as a melilite-buchite.

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(e) *Aegirine- and nepheline-bearing residua*

There are restricted amounts of alkaline residua mainly of very fine grain size in which aegirine or aegirine-augite and nepheline occur. Green soda-pyroxene, present as skeletal crystals with ore, apatite and perhaps feldspathoid is typical of such occurrences forming an interstitial residuum in melilite-titanaugite-rock (NI 1302). In thomsonitized wollastonite-bearing hybrid gabbro (NI 1303) a mesostasis has the same minerals, with coarser aegirine. Altered nepheline is present in other pyroxenites or pyroxene-rich dolerites, commonly in examples that are thomsonitized.

5. The reaction process

(a) *Introduction*

The major variable affecting the metamorphic reactions are pressure and temperature, the chemical composition of the silicate phases and the composition of the fluid phases, especially where the reactions involve CO_2. These reactions are accordingly not univariant, which restricts their suitability as temperature indicators. Establishment of the relatively low approximate pressure of emplacement does however allow some indication of the conditions of formation in the light of the probable magmatic temperature.

The trends of the reaction processes are similar to those of Scawt Hill (figure 12) but allow the relationships in the intermediate area between the olivine-dolerite and the extreme hybrids to be defined more clearly.

The nature of the trends was discussed by Tilley (1952). The Camas Mòr, Carneal and, to a less extent, Scawt Hill suites show related curved paths and there is also a relation to the vergency of the main magma suites depicted by Tilley.

(b) *Pressure at time of emplacement*

The thickest sequence of Tertiary basalt lavas in Northern Ireland was found in the Langford Lodge borehole on the eastern side of Lough Neagh where 232 m of Upper Basalts overlay the Interbasaltic bed and 531 m of Lower Basalts. In the Carneal area, however, the Lower Basalts appear to be only some 200 m thick and at the Carneal plug probably less than 100 m are present.

The original thickness of Upper Basalts is unknown. At Trostan, in north Antrim, 240 m are still present and at Agnews Hill, 8 km to the northwest of Carneal, 140 m remain. The 50 million years of post-eruptive erosion in the Tertiary must have removed some hundreds of metres and it may be conjectured that the original thickness of the Upper Basalts was perhaps 500 m and that at the end of the volcanic period there were up to 600 m of basalt lava in the area above the present land surface. The total thickness at Carneal was thus probably 700 m.

Plugs of the Carneal type in Antrim appear to penetrate the whole thickness of basalt, although it cannot of course be shown that they may have terminated at some level above the present land surface but below the original top of the now eroded lava pile. Preston (1963) has shown that the Slemish plug was formed by multiple intrusions and it is probable that most if not all of the 30 plugs acted as feeders.

The probable total thickness is equivalent to a load pressure of only about 200×10^5 Pa

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or slightly less at the height of the present land surface. There are various factors of unknown effect. These include on one side, gas overpressure due to the metamorphism, although this cannot have been high in the proximity of the conduit through which the magma was flowing. The amount of magmatic pressure from beneath is also unknown. Since fragmental rocks are uncommon in the Basalt succession, and only one rock from the plug has the appearance of a tuff, it is likely that pressure from this cause is not very high. Lowering of the pressure could have occurred to an unknown extent along fractures although minor dykes are not found.

The source rocks of the Carneal metmorphic suite occur immediately beneath the basalts, so the pressure of 200×10^5 Pa could not have been greatly exceeded below the volcanic pipe. Even if the stratigraphic assumptions now made are not very accurate, this pressure cannot be greatly in error.

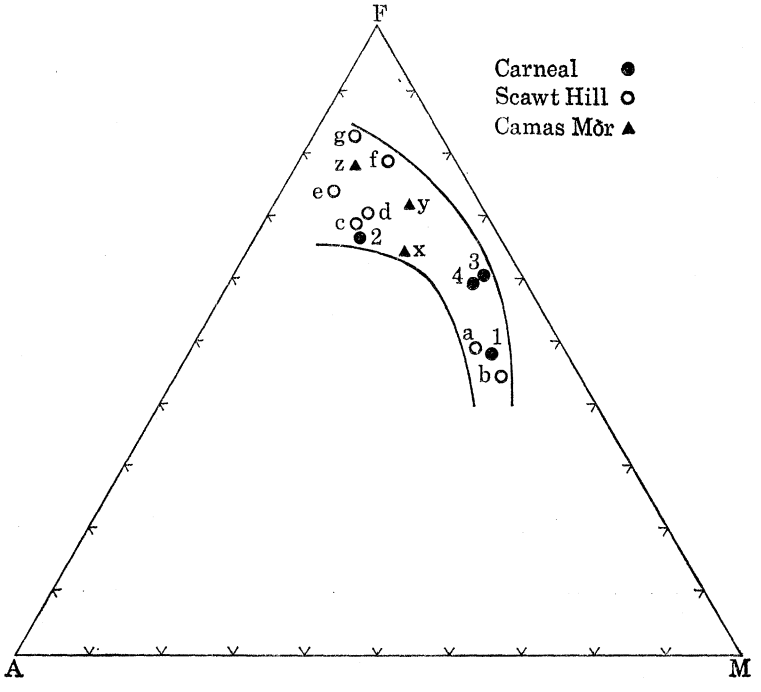

FIGURE 12. Plot of the oxides $FeO+Fe_2O_3$ (F), MgO (M) and Na_2O+K_2O (A) for the endomorphic rocks of Carneal, and Scawt Hill, Co. Antrim, and Camas Mòr, Muck. Carneal: 1, olivine-dolerite; 2–4, hybrid rocks, as table 6. Scawt Hill: a, olivine-dolerite; b, pyroxene-rich dolerite; c, anorthoclase-fayalite-dolerite; d, titanaugite-dolerite; e, nepheline-dolerite; f, melilite-bearing nepheline dolerite; g, melilite-rock (Tilley 1952). Camas Mòr: x, olivine-gabbro; y, pyroxenite; z, wollastonite-nepheline-gabbro (Tilley 1952).

(*c*) *Temperature of reaction*

The pressure of CO_2 could have been high in restricted environments, although in view of the small quantity of rocks present that were formed originally from limestone or by reaction with limestone, the total CO_2 component of the gas phase is unlikely to have been high. The reaction probably varied considerably, with lower temperatures corresponding to lower partial pressures of CO_2. As might be expected, as the gaseous mixture changed from CO_2- to H_2O-rich, the temperatures of formation of wollastonite from calcite and quartz were much lower, as shown by Greenwood (1962).

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(i) *Temperature of the basaltic magma*

The numerous determinations of the temperatures of erupting magmas, especially basalt, that have been made provide a guide to the temperature of emplacement of the Carneal plug. Five recent sets of determinations of temperatures in or near vents (as opposed to lava fountains, or flows far from vents, both of which depart from magma temperatures), range from 1040 to 1150 °C. From a depth of 11.5–13 m in Halemaumau lava lake at Kilauea, Jagger (1917) measured a temperature of 1170 °C which has been taken by Macdonald (1972) as close to that of the magma as it rose from depth. Tanguy & Biquand (1967) provide lower measurements for Etna, which near the vent were 1000–1050 °C. It seems reasonable to take a temperature of about 1050–1100 ° as that of Carneal, and since the measurements quoted are from active volcanoes, they probably relate to a geothermal gradient reasonable for Carneal. There seems no reason to think that the prograde metamorphic and metasomatic reactions in the small quantities of country rock involved, departed far from these temperatures.

(ii) *Temperature of the main metamorphism*

Many of the systems of equilibria within which the reaction mechanisms occur have been investigated and give an indication of the temperature prevailing during metamorphism, although the considerably greater complexity of the natural systems does not allow much precision. Although maximum temperatures reached yielded larnite and bredigite, individual specimens containing diopside, wollastonite, etc., indicate the lower steps in the decarbonization series. The conditions of formation of these species have been provided for diopside and forsterite by Weeks (1956); for wollastonite, by Harker & Tuttle (1956*a*) and under hydrothermal conditions, by Kalinen (1967); for artificial melilite by Nurse & Midgley (1953); åkermanite, Harker & Tuttle (1956*b*); spurrite, Tuttle & Harker (1957), Wyllie & Haas (1966) and Zharikov & Shmulovich (1969); merwinite and calcite, Walter (1965), and for the composition and formation of bredigite, Biggar (1971) and Midgley (1974), and in the hydrous system, Roy (1958).

Dicalcium silicate, present principally as larnite, has been extensively investigated (Nurse 1952; Midgley 1974) and is stable at pressures of 200×10^5 Pa CO_2 at over 1000 °C (Zharikov & Shmulovich 1969). That the minerals are mainly more complicated than the simpler systems considered in these studies reduces the validity of the temperatures found. This arises especially from the presence of iron, aluminium and titanium in the pyroxenes, iron and sodium in the melilite and magnesium and iron in the wollastonite, as well as the complex associations of minerals. There are also differing physical conditions including time of crystallization. The probable approximate temperatures at which the reactions occurred are however indicated, and within themselves are reasonably consistent in their increasing magnitude and hence the sequence of minerals.

For related assemblages from next to a dolerite sheet in the Anakit district of Siberia, where conditions of heat transfer were somewhat different, Reverdatto (1970) has suggested a temperature of 1000 °C for the exomorphic rocks.

(iii) *Temperature conditions of the retrograde phase*

In the waning conditions of metamorphism the anhydrous suite was succeeded by low-temperature carbonate-bearing and hydrous minerals in their respective stability fields as

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hydrothermal fluids became re-established. As detailed below, the highest temperature minerals in this phase are hydrogrossular (520–620 °C or less), bicchulite (below 650 °C) and the lowly-refringent wollastonite probably also belongs here. Xonotlite, scawtite, tobermorite, analcime, thomsonite, plombierite and the other minerals formed at successively lower temperatures (figure 14). Portlandite forms below 680 °C but probably at much lower temperatures.

(*d*) *Metasomatic effects*

Apart from the chemical reactions revealed generally by the whole suite of rocks, fine-grained textural replacements can be distinguished and are to be measured in the range from millimetres to hundredths of a millimetre. That the process of metsomatism is, in part at least, one of precision and tranquillity, is shown by the retention of the structure of the cyclostome bryozoan now preserved in wollastonite (figure 4, plate 12). The change in the organism from calcite to wollastonite (assuming the content of lime remains constant) results in about 8% volume increase, so the very fine structure is disrupted to this extent at least, but otherwise it is preserved in fine detail.

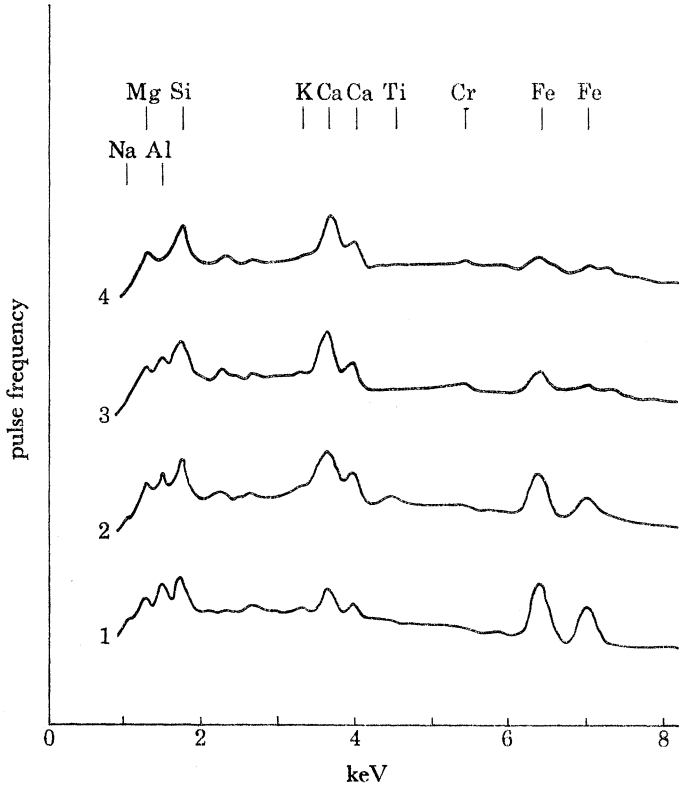

FIGURE 13. Spectra of successive zones from olivine-basalt to merwinite vein. I 1313. 1, Olivine-basalt; 2, melilite-magnetite zone; 3, melilite-hydrogrossular-merwinite zone; 4, merwinite zone.

Other rocks display the details of metasomatic changes and fronts. They are well shown by a vein of merwinite traversing dolerite, which gives rise to several metasomatic zones (I 1313; figure 10, plate 13). The igneous rock is here a basalt on the basis of its grain size; zonation within a few millimetres from the igneous rock to the vein is as follows:

Olivine-basalt – melilite (+magnetite) – melilite-(hydrogrossular) – (merwinite) – melilite-hydrogrossular-merwinite – merwinite. Although the metasomatic zones are very obvious in

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thin section, they show more restricted variation chemically than the appearance suggests, the turbidity and grain-size of the minerals, or dusting with small quantities of opaque ore, exercising a marked effect. The overall chemical similarity is shown by the appearance of spectra (figure 13) taken with a non-dispersive (Ortec) multichannel analyser on a scanning electron microscope (Stereoscan).

Electron linear-probe analyses of successive zones from the igneous rock to the merwinite-vein are the basis of analyses shown in table 7.

TABLE 7. CHEMICAL COMPOSITION ACROSS A METASOMATIC MERWINITE VEIN

	I	II	III	IV	IVA	V
SiO_2	47.62	47.3	47.7	35.0	36.8	40.4
Al_2O_3	17.39	16.1	15.5	11.2	11.3	0.3
Fe_2O_3	2.90	2.1	1.9	1.5	1.6	—
FeO	7.12	5.1	4.9	3.7	3.8	2.4
MgO	8.58	7.6	7.6	3.5	4.6	9.7
CaO	11.64	13.8	16.0	39.1	35.6	46.0
Na_2O	2.03	3.0	4.2	1.3	2.1	—
K_2O	0.20	0.3	0.2	—	—	—
TiO_2	1.00	0.7	0.1	0.7	0.2	—
MnO	0.17	0.1	0.1	0.1	0.1	0.1
SO_3	tr.	1.1	—	—	—	—
H_2O, rest	1.79	[1.8]	[1.8]	[3.9]	3.9	[1.1]
totals	100.44	99.0	100.0	100.0	100.0	100.0

Analyses II–V are not individually accurate.

I, Olivine-dolerite. Centre of Carneal plug; full analysis, table 6, column I.
II, Olivine-basalt *ca.* 8 mm from its edge against merwinite vein.
III, Olivine-basalt. Edge next to melilite zone around merwinite vein.
IV, Melilite-hydrogrossular zone.
IVA, Calculated analysis, $\frac{2}{3}$ melilite + $\frac{1}{3}$ hydrogrossular.
V, Central merwinite zone (main part of vein).
II–V, Based on electron-microprobe (Geoscan) analyses by Mrs A. Tresham. H_2O, minor constituents, and valency of iron estimated from known analyses of comparable rocks.

The analyses show that the basalt resembles that analysed from the centre of the plug (table 7, columns I and II), although lime is present in greater quantity, rising nearer the margin (column III). In this instrumental analysis, and the others in the table, water, minor constituents, and the valency of iron have been allowed for by reference to full analyses of similar rocks. They are not very accurate in themselves, but indicate the nature of the changes.

The olivine-basalt contains normal olivine and has a decidedly foliated texture, with plagioclase laths, An_{55}, mainly 0.1–0.2 mm in length. The foliation, which is cut by the edge of the basalt, continues into the adjacent metasomatized zones, but the plagioclase laths have been replaced by melilite (figure 11, plate 13). There are other examples of metasomatic effects. In the basalt, a thin (0.4 mm) band rich in olivine can be traced for about 2 mm up to the boundary which cuts a microphenocryst that in the melilite zone continues as a coarse mosaic of melilite with a dense dusting of magnetite at its margin. The variation of texture and continuation of the foliation die out towards the merwinite vein.

The metasomatic zone closest to the basalt is composed of melilite dusted for about 0.2 mm with opaque mineral, probably magnetite. Away from the basalt, merwinite is developed in very fine granules and there is hydrogrossular and also thomsonite. The melilite laths show granulation to a much finer grain size. The hydrogrossular (hibschite), seen best when the

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rock is crushed to a very fine grain size, cements the melilite. It increases towards the merwinite vein. The merwinite is in prisms mainly 1.5 mm long oriented parallel to the vein margin and showing typical twinning. It is intergrown with melilite, and hydrogrossular is also developed. There is some pyrrhotine in this zone. Both larnite and monticellite are just detectable on X-ray powder films, and are indicated optically, although the results of Reverdatto (1964), and others, show that these minerals must be mutually out of equilibrium.

The analysis of the melilite-hydrogrossular zone, adjusted for water content, minor constituents and valency of iron (column IV) may be compared with a calculated analysis for $\frac{2}{3}$ melilite plus $\frac{1}{3}$ hydrogrossular (column IVA).

Other electron-probe analyses confirm the composition of the hydrogrossular, merwinite, and the mixed melilite–merwinite–hydrogrossular zones. An analysis based on that of the central merwinite zone, shown in column V, is essentially merwinite with minor amounts of melilite and probably other minerals.

The metasomatic production of the melilite–merwinite–hydrogrossular suite is dominantly caused by lime-rich solutions. The plagioclase is converted to melilite. With conventional molecules

$$\underset{\text{anorthite}}{2(CaO.Al_2O_3.2SiO_2)} + 3CaO \longrightarrow \underset{\text{gehlenite}}{2CaO.Al_2O_3.SiO_2} + \underset{\text{pseudo-sarcolite}}{3CaO.Al_2O_3.3SiO_2}.$$

Soda from the albitic component enters the melilite molecule as soda-melilite, or, with alumina, soda-sarcolite $3Na_2O.Al_2O_3.3SiO_2$, or pseudo-sarcolite. Augite and olivine react similarly to the metasomatism, magnesia entering the åkermanite molecule, $2CaO.MgO.2SiO_2$. With excess lime the reaction goes to merwinite $3CaO.MgO.2SiO_2$. Alumina enters the melilite molecule and, with water, hydrogrossular, $3CaO.Al_2O_3.2SiO_2.2H_2O$.

The products and reaction they indicate are schematically as follows (mass percent of oxides):

		CaO	Al_2O_3	MgO	SiO_2	H_2O
6 gehlenite		12	6	0	6	0
9 merwinite		27	0	9	18	0
12 hydrogrossular		36	12	0	24	24
	sum	75	18	9	48	24
oxides in olivine-basalt		12	18	9	48	1
	difference	63	—	—	—	23

The olivine-basalt thus requires 63 CaO plus 23 H_2O (parts by mass) to be converted to the melilite–hydrogrossular–merwinite association. Iron, sodium, and other minor constituents to some extent enter the lattices of the calcium-bearing silicates, but are partly fixed in separate minerals (for example, magnetite in melilite). The lime and water required is only slightly more hydrous than calcium hydroxide (portlandite).

(*e*) *Paradox of the chemical processes of hybridism*

The conditions governing the additions necessary to change olivine-dolerite magma into hybrid rocks are of some complexity. The chemical analyses of the olivine-dolerite and of the endomorphic rocks are shown in table 6. In table 8 are shown the analyses of the dolerite, hybrid gabbro and hybrid pyroxenite recalculated free of CO_2 and H_2O to 100% together

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with calculated compositions of similar rocks derived from the olivine-dolerite principally by the addition of lime and silica. On a plot of the oxides against silica, for the olivine-dolerite to hybrid gabbro, alumina falls to zero at about 35.3 % SiO_2. Magnesia falls close by. It is reasonable to take the addition (or subtraction) of the oxides to the olivine-dolerite in their proportions at this silica percentage. At this position the proportion of the alumina contents in the two rocks governs the amounts of the other oxides. The figures may readily be obtained by nomography or more precisely by calculation.

TABLE 8. EFFECT OF LIMESTONE ASSIMILATION ON OLIVINE-DOLERITE

	1	2	3	4	5
SiO_2	48.13	45.52	44.91	44.06	43.74
Al_2O_3	17.58	12.27	12.11	11.64	11.56
Fe_2O_3	2.93	5.46	5.39	4.99	4.96
FeO	7.20	6.54	6.45	5.88	5.84
MgO	8.67	6.05	6.83	5.74	6.11
CaO	11.76	20.76	20.48	24.73	24.56
Na_2O	2.05	1.24	1.23	0.83	0.82
K_2O	0.20	0.14	0.30	0.13	0.11
TiO_2	1.01	1.68	1.66	1.67	1.66
rest	0.47	0.34	0.64	0.33	0.64
	100.00	100.00	100.00	100.00	100.00

1, Olivine-dolerite (table 6, column I) recalculated to 100 % after removal of H_2O.

2, Calculated composition of 100 parts olivine-dolerite plus 17.07 SiO_2, 4.89 Fe_2O_3, 2.16 FeO, 17.97 CaO and 1.40 TiO_2, minus 0.27 Na_2O, corrected to 100 %.

3, Hybrid pyroxenite (table 6, column IV), recalculated to 100 % after removal of CO_2 and H_2O.

4, Calculated composition of 100 parts of olivine-dolerite plus 18.39 SiO_2, 4.61 Fe_2O_3, 1.68 FeO, 25.58 CaO, and 1.51 TiO_2, minus 0.80 Na_2O, corrected to 100 %.

5, Wollastonite-bearing hybrid gabbro (table 6, column III), recalculated to 100 % after removal of CO_2 and H_2O.

Since there is a considerable total addition of material, largely lime, to the parent magmatic rock the desilication required for the final (hybrid) rock paradoxically also requires the addition of silica or the proportion of silica in the resultant rock is too low. The principal additions are lime (from the chalk) and silica (from flint) with minor amounts of other oxides, and minor subtraction of alkalis. Silica could also come as available from the more highly silicic minerals at different stages of alteration elsewhere in the plug. At Scawt Hill Tilley was able to derive the hybrid rock with the algebraic addition of only four oxides, but the net amount added was only 7.68 % and was not sufficient to distort the figures for the other oxides. This is not the case at Carneal where precise correspondence can only be achieved by minor addition to each oxide. The necessary lime and silica to convert the olivine-dolerite to hybrid pyroxenite are CaO 17.97 %, SiO_2 17.07 %; and to wollastonite-bearing hybrid gabbro CaO 25.58 %, SiO_2 18.39 % are required, in each case with minor amounts of iron and titania and removal of alkalis. In contrast, the amounts of lime and silica to produce the Scawt Hill pyroxene-rich dolerite was CaO 7.40 %, SiO_2 3.06 % only. The total net additions for the two Carneal rocks are 45.16 % and 52.06 % respectively. If the metasomatic additions and subtractions are restricted to calcium, titanium, iron and alkali oxides, as shown in the table, there is necessarily some departure as indicated from precise correspondence between final calculated analysis and actual rock. The close correspondence of the actual and calculated compositions is seen in table 8 by comparison of columns 2 and 3, and 4 and 5.

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6. The petrogenetic sequence

(*a*) *Prograde phase*

The more advanced stages of the pure thermal metamorphism (decarbonation) of siliceous limestone and dolomite were charted in theoretical and practical detail by Bowen (1940) and later authors including Tilley (for example, 1951*a*). Most of the successive steps are found at Carneal, where the concomitant or controlling metasomatic reactions are displayed, and they are followed by the successive but less precise phases of retrograde metamorphism. Each of Bowen's reaction steps is based on the (negative) evidence of the disappearance of the mineral phase, but the order of the initial appearance of the various minerals is with rising temperature. It is suggested that at Carneal this is not simply a temperature phenomenon but the reactivity of the minerals under the constraints of temperature, pressure (including build-up and release), and chemical potential of fixed and volatile components, especially changing partial pressure of CO_2 and H_2O. This might loosely be referred to as metasomatic pressure.

In the sequence of minerals in the contaminated igneous rocks (in which forsterite is already a principal component of the olivine), the earliest mineral formed is sahlite, pyroxene that contains 71 % diopside and 15 % hedenbergite. In the metamorphism of dolomitic siliceous limestones this mineral was fourth in the steps of Bowen as modified by Tilley and others, next above forsterite. Although in the limestones Bowen suggested the formation of diopside by the disappearance of tremolite (treated as anhydrous), in the igneous rock it forms at the expense of olivine.

The direct equation is

$$\underset{\text{forsterite}}{Mg_2SiO_4} + 2CaCO_3 + 3SiO_2 \longrightarrow \underset{\text{diopside}}{2CaMg(SiO_3)_2} + 2CO_2. \qquad (1)$$

The fayalitic component of the olivine (which is chrysolite, about Fo_{81}) yields the hedenbergitic component of the diopside

$$\underset{\text{fayalite}}{Fe_2SiO_4} + 2CaCO_3 + 3SiO_2 \longrightarrow \underset{\text{hedenbergite}}{2CaFe(SiO_3)_2} + 2CO_2. \qquad (2)$$

In the olivine-dolerite there is a substantial hypersthene component in the pyroxene, amounting to 14.62 % in the norm (nearly half of the total normative pyroxene), which also contributes to the sahlite

$$\underset{\text{hypersthene}}{(Mg,Fe)SiO_3} + CaCO_3 + SiO_2 \longrightarrow \underset{\text{sahlite}}{Ca(Mg,Fe)(SiO_3)_2} + CO_2. \qquad (3)$$

As pointed out by Tilley for Scawt Hill, silica comes from plagioclase, the albite molecule yielding nepheline in the liquid

$$\underset{\text{albite}}{3NaAlSi_3O_8} + 4CaCO_3 + \underset{\text{forsterite}}{2Mg_2SiO_4} \longrightarrow \underset{\text{diopside}}{4CaMg(SiO_3)_2} + \underset{\text{nepheline}}{3NaAlSiO_4} + 4CO_2. \qquad (4)$$

Labradorite may be desilicated to thomsonite in which the Na_2O/CaO ratio is very similar, and SiO_2 is about 16 % lower.

For the production of wollastonite in both the contaminated igneous rocks and in the exomorphic suite, the direct reaction of silica and limestone occurs

$$CaCO_3 + SiO_2 \longrightarrow \underset{\text{wollastonite}}{CaSiO_3} + CO_2. \qquad (5)$$

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Melilite is extensively developed, principally as humboldtilite containing 65% åkermanite, 6% gehlenite and 29% pseudo-sarcolite. It is formed by the reaction of the pyroxene and plagioclase with chalk

$$\underset{\text{diopside}}{CaMgSi_2O_6} + CaCO_3 \longrightarrow \underset{\text{åkermanite}}{Ca_2MgSi_2O_7} + CO_2, \tag{6}$$

$$\left.\begin{matrix}\underset{\text{anorthite}}{CaAl_2Si_2O_8}\\ \underset{\text{albite}}{NaAlSi_3O_8}\end{matrix}\right\}_{\text{plagioclase}} + 2CaCO_3 \longrightarrow \underset{\text{gehlenite}}{Ca_2Al_2SiO_7} + \underset{\text{wollastonite}}{CaSiO_3} + 2CO_2, \tag{7}$$

$$\underset{\text{albite}}{NaAlSi_3O_8} + 2CaCO_3 \longrightarrow \underset{\text{pseudo-sarcolite}}{CaNaAlSi_2O_7} + \underset{\text{wollastonite}}{CaSiO_3} + 2CO_2. \tag{8}$$

Excess silica combined with lime to form wollastonite, which is plentiful. The anorthite reaction was found experimentally by Yoder (1950). A closely related reaction was postulated by Tilley & Harwood (1931) for Scawt Hill

$$\underset{\text{anorthite}}{2CaAl_2Si_2O_8} + 3CaO \longrightarrow \underset{\text{gehlenite}}{Ca_2Al_2SiO_7} + \underset{\text{pseudo-sarcolite}}{Ca_3Al_2Si_3O_{12}}. \tag{9}$$

Following Hey (1950) the theoretical pseudo-sarcolite composition of equation (8) is taken as containing sodium, in agreement both with the original description and the occurrence at Carneal.

Removal of olivine, principally forming diopside as in equation (1), may also yield melilite

$$\underset{\text{forsterite}}{Mg_2SiO_4} + 4CaCO_3 + 3SiO_2 \longrightarrow 2\underset{\text{åkermanite}}{Ca_2MgSi_2O_7} + 4CO_2. \tag{10}$$

Iron from olivine may be precipitated as magnetite.

In the metasomatic reaction, melilite (and merwinite) may be associated with hydrogrossular which develops at the expense of calcic plagioclase

$$\underset{\text{anorthite}}{CaAl_2Si_2O_8} + 2CaO + 2H_2O \longrightarrow \underset{\text{hydrogrossular}}{Ca_3Al_2Si_3O_{10}.2H_2O}. \tag{11}$$

Direct metasomatism of the olivine-dolerite yields the melilite, merwinite and hydrogrossular by the addition of lime and water (p. 257).

Monticellite is present in small quantity in a metasomatic vein where it forms (Bowen 1940) at the expense of diopside and olivine in the dolerite

$$\underset{\text{diopside}}{CaMgSi_2O_6} + \underset{\text{forsterite}}{Mg_2SiO_4} + 2CaCO_3 \longrightarrow 3\underset{\text{monticellite}}{CaMgSiO_4} + 2CO_2. \tag{12}$$

Spurrite occurs in small quantity in association with larnite, melilite and some merwinite in a rock that is probably completely metasomatized dolerite.

Various mechanisms have been postulated for the formation of merwinite, which is of relatively limited occurrence in metasomatic veins

$$\underset{\text{diopside}}{CaMgSi_2O_6} + 2CaCO_3 \longrightarrow \underset{\text{merwinite}}{Ca_3Mg(SiO_4)_2} + 2CO_2, \tag{13}$$

$$\underset{\text{åkermanite}}{Ca_2MgSi_2O_7} + CaCO_3 \longrightarrow \underset{\text{merwinite}}{Ca_3Mg(SiO_4)_2} + CO_2. \tag{14}$$

Dicalcium silicate is present to a very minor extent as larnite and as bredigite, the α'_L higher temperature form of Ca_2SiO_4 at atmospheric pressures, although the exact nature of the phase

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has been the subject of enquiry (Midgley 1974). The bredigite occurs in a hybrid pyroxenite showing local metasomatism and its precise relations are uncertain.

In the various equations developed theoretically or found experimentally for the production of the successive mineral phases of thermal metamorphism, the limestone, with or without silica, reacts with the igneous or endomorphic constituents, or the products do. The metamorphic reactions in the igneous rocks and in the calcium carbonate are thus nearly all complementary and the reactions appropriate to the dolerite or chalk and flint may be the same. The principal differences may lie in the essentially solid state reactions in the chalk and flint and the liquid ones in the dolerite magma, save where metasomatism is demonstrable.

The pure chalk that was assimilated by the dolerite of the plug cannot be readily sampled but from Scawt Hill, Co. Antrim a specimen of marble yielded only 0.24 % acid-soluble MgO (NI 1682; Lab. no. M 328, analyst G. A. Sergeant); MgO is very low in the exomorphic rocks (table 5).

There has been considerable discussion of the theoretical appearance of forsterite before diopside and it is pointed out by Weeks (1956) that its absence in metamorphosed siliceous dolomite precludes the reaction with calcite or with calcite and diopside, to yield monticellite and monticellite and periclase respectively. This is directly relevant at Carneal where monticellite has not been recognized in metamorphosed limestone. In the metasomatic fronts the forsterite may be provided from the igneous rock.

For the production of wollastonite the basic reaction of the metamorphism of all siliceous limestones (5) occurs. Wollastonite is especially well developed, replacing flint by tranquil metasomatism.

The production of melilite in metamorphosed chalk can only have resulted from the introduction of silicate solutions, which yielded gehlenitic or some åkermanitic species depending upon the magnesium content. These are effectively the metasomatic equivalents of equations (6), (7), (8), and (9).

Tilleyite, discovered by Larsen & Dunham (1933) at Crestmore, was shown by Nockolds (1947) to be an intermediate step to spurrite. It has not been found in the present study although small amounts are to be expected.

Spurrite is produced by the reaction of wollastonite and limestone

$$\underset{\text{wollastonite}}{2CaSiO_3} + 3CaCO_3 \longrightarrow \underset{\text{spurrite}}{Ca_5(SiO_4)_2CO_3} + 2CO_2. \qquad (15)$$

The reaction of diopside and åkermanite with limestone (equations (13) and (14)) produces merwinite and the reaction of spurrite, monticellite and åkermanite has been postulated by Walter (1965) following Burnham

$$\underset{\text{spurrite}}{Ca_5(SiO_4)_2CO_3} + 2\underset{\text{monticellite}}{CaMgSiO_4} + \underset{\text{åkermanite}}{Ca_2MgSi_2O_7} \longrightarrow 3\underset{\text{merwinite}}{Ca_3Mg(SiO_4)_2} + CO_2. \qquad (16)$$

The paucity of monticellite suggests that this reaction, if it occurred, was uncommon.

Larnite may be formed by the reaction

$$\underset{\text{wollastonite}}{CaSiO_3} + \underset{\text{spurrite}}{Ca_5(SiO_4)_2CO_3} \longrightarrow 3\underset{\text{larnite}}{Ca_2SiO_4} + CO_2, \qquad (17)$$

ignoring a possible intermediate step through rankinite, the tricalcium silicate which has not been found at Carneal.

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Larnite and merwinite occur to a limited extent in the same rocks at Carneal and may form from the reaction

$$\underset{\text{spurrite}}{Ca_5(SiO_4)_2CO_3} + \underset{\text{åkermanite}}{Ca_2MgSi_2O_7} \longrightarrow \underset{\text{larnite}}{2Ca_2SiO_4} + \underset{\text{merwinite}}{Ca_3Mg(SiO_4)_2} + CO_2. \qquad (18)$$

In the presence of alumina concentrations, gehlenite is stable (Reverdatto 1964).

(*b*) *Retrograde phase*

The general conditions of formation of the scarce to rare minerals that characterize the Carneal rock suite are less well established than for the prograde phase, since it is not always possible to distinguish between different stages of crystallization, and the minerals continue to exist in stable or metastable state when cooled. The stability states of several of the hydrated minerals are in overlapping ranges so that individual works generally deal with more than one mineral, and the minerals are conveniently reviewed together.

(i) *Hydrogrossular*

As pointed out by Yoder (1950), many of the minerals described as grossular are in fact hydrogrossular, so their parageneses may be relevant to that of the Carneal mineral. Under hydrothermal conditions Yoder synthesized a hydrogrossular with refringence $n = 1.67$, similar to the Carneal hibschite, at 520 °C under 180×10^5 Pa in 100 h. The refringence for two of the minerals found at Carneal, with a range $n = 1.667$–1.682, is close to the minimum energy configuration for the series, a concept suggested by Buerger (Yoder 1950). At 200×10^5 Pa the refractive index range indicates a temperature from about 520 to 620 °C. Later, Roy & Roy (1957) suggested that these figures were too high because of the shortness of experimental runs and failure to quench. They also reported (1962) that hydrogrossular had a maximum thermal stability for a composition of 1.5 mol SiO_2 (perhaps 2.0) which was only stable below 360 °C (at 1000–2000×10^5 Pa). In the hydrothermal breakdown of gehlenite, Christie (1964) found hydrogarnet (hydrogrossular) was formed (with boehmite) at 135×10^5 Pa at 355 °C. The equation Christie gives is

$$\underset{\text{gehlenite}}{3Ca_2O_2Al_2SiO_5} + 7H_2O \longrightarrow \underset{\text{hydrogrossular}}{2Ca_3Al_2(Si_{1.5}H_6)O_{12}} + \underset{\text{boehmite}}{2AlO(OH)}.$$

Mixed åkermanite-gehlenite crystals studied at 600 MPa pressure yielded hydrogarnet together with boehmite at various temperatures from 369 °C (0–75 % åkermanite) to 615 °C (for gehlenite). Christie concluded that melilite broke down to anhydrous products that were transformed to 'vesuvianite type minerals' that were in turn converted to assemblages with hydrogarnet. This does not appear to be the paragenesis at Carneal where vesuvianite has not been an intermediate phase.

(ii) *Portlandite*

The presence of portlandite suggests that SiO_2 and Al_2O_3 were not available when it crystallized, or wollastonite or melilite would have formed at high temperatures. Apatite would have formed if P_2O_5 had been available. Calcium hydroxide is normally readily carbonated and must have crystallized under the extreme conditions of absence of CO_2. It is stable (in the system CaO–CO_2–H_2O) up to 680 °C at 250×10^5 Pa, falling to 640 °C at 27×10^5 Pa (Wyllie & Tuttle 1960) but at Carneal it formed at low temperatures when CO_2 had been removed.

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(iii) *Bicchulite*

The synthetic compound to which this mineral corresponds is formed only in the temperature range 260–650 °C, under pressures from 45 to over 2000×10^5 Pa (Carlson 1964). At lower temperatures the low-temperature ($8H_2O$) hydrate is stable. The natural monohydrate (Henmi *et al.* 1973) is stable on heating for periods of 2–6 days up to about 595 °C at 3000×10^5 Pa ranging up to about 630 °C at 1000×10^5 Pa; at higher temperatures gehlenite is stable.

(iv) *Scawtite*

Scawtite was sometimes formed in the hydrothermal CaO–Al_2O_3–SiO_2–H_2O systems under various pressures ($780–1900 \times 10^5$ Pa) from 255 to 320 °C when CO_2 was present (Buckner *et al.* 1960). Depending upon its composition, its stability lies between 140 and 300 °C, irrespective of pressure (Harker 1965). Its stability field lies between the tobermorite-calcite and xonotlite-calcite fields.

(v) *Xonotlite, tobermorite*

The sequence of equilibrium states on dehydration was presented by Roy & Harker (1962) to be tobermorite and gyrolite (stable at about 160 °C), xonotlite (*ca.* 200 °C), with gyrolite disappearing above 200 °C and tobermorite by 300 °C, and xonotlite persisting to 360–410 °C. Comparable results have been reported elsewhere. Gyrolite has been reported by Harker (1964) to be stable (on dehydration) from 150 to 300 °C, but the mineral breakdown is not sharp. It can, but need not, form with xonotlite from the expulsion of water from tobermorite (Harker 1964). Increase of pressure favours stability at higher temperatures of tobermorite, although Speakman (1968) did not find that pressure affected the reaction temperature much. The 11.3 Å tobermorite is unaffected on heating to 125 °C, partially converted to the 9.35 Å form at 180–240 °C and completely converted to 9.35 Å at 300 °C (Taylor 1959*a*). Both tobermorite and xonotlite can transform into β-$CaSiO_3$ on heating (400 °C hydrothermally or 800 °C and atmospheric humidity), and Taylor (1959*b*), reporting this, has shown that xonotlite is produced under hydrothermal conditions between about 150 and 400 °C. It forms at 380 °C under 250×10^5 Pa pressure from tobermorite, most readily from the 11.3 Å hydrate, which is the one found commonly at Carneal. With change of vapour phase from $CO_2 > H_2O$, the transformation temperature (to wollastonite) changes from 300 °C to about 430 °C at 100 % H_2O (Agrell 1965).

Xonotlite and tobermorite thus exist together over a considerable range of temperatures, and it has also been noted that xonotlite forms from compositions richer in lime, and tobermorite from those slightly richer in silica, although this is disputed by Speakman (1968).

(vi) *Tacharanite*

The temperature range of formation of tacharanite under hydrothermal conditions was suggested by Koritnig (1972) to be 160–250 °C. The mineral was found by Sweet *et al.* (1961) to alter to tobermorite, gyrolite and xonotlite. In one amygdale at Carneal, gismondine occurred surrounding tacharanite.

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(vii) *Zeolites and related minerals*

There are considerable complications to the stability fields of zeolites and related minerals. Analcime, present in a brecciated olivine-basalt, is produced (from albite) between 220 and 300 °C (Fyfe & Turner 1958) but under 200×10^5 Pa pressure it is stable up to about 500 °C (Yoder 1954). Thomsonite may be stable up to 300 °C in silica-deficient environments, and gismondine appears to be somewhat more stable than thomsonite (Coombs, Ellis, Fyfe & Taylor 1959); it shows a marked dehydration between 125 and 160 °C (Hoss & Roy 1960).

(viii) *Plombierite*

As at Ballycraigy, Co. Antrim, the 14.6 Å mineral probably results from hydration of the 11.3 Å tobermorite. This is consonant with water contents of the two minerals, which are $H_2O:SiO_2$ 1.0 for tobermorite and 2.0 for plombierite (McConnell 1954). The Carneal plombierite and tobermorite were detected by X-rays after the specimens had been stored in a heated, dry building so it is probable that as with tacharanite their lattices are stabilized by impurities. The turbidity of the thin sections may be due to variability of hydration state.

The stability range is accepted by Roy & Harker (1962) to be below 100 °C, and the 14.0 Å hydrate was not detected on films of material heated to 125 °C (Taylor 1959*a*).

(ix) *Textural relations*

The textural relations of the hydrated minerals give some indication of the sequence of crystallization although complete reliance cannot be placed on this in view of the difficulties of discerning some relations and the variation of physical conditions, especially temperature and pressure, and chemical compositions.

Bicchulite occurs in association with hydrogrossular (hibschite) as the outer part of the crust on wollastonite, its relatively high temperature of formation fitting in with that of the hydrogrossular. The stability ranges of thomsonite and tobermorite appear to be similar but in several amygdales tobermorite generally crystallized first, around thomsonite. Gyrolite, present in wollastonite-rock, can form in the same temperature range as tobermorite, which may be associated with the wollastonite. Gismondine is marginal to tacharanite which probably has the lower temperature of formation.

Calcite is present in various associations. In a vein in pyroxenite it is marginal to tacharanite and cements a brecciated olivine-basalt with which analcime is associated. In xonotlite-scawtite-rock the xonotlite cooled and cracked and was then invaded by CO_2-bearing solution which crystallized as scawtite and calcite. This confirms the suggestion of McConnell (1955*b*) that scawtite (containing H_2O and CO_2) can be considered as a carbonated xonotlite. Veins of drusy calcite in metasomatized flint (largely hibschite) contain tobermorite. In a different association, calcite is central to a coarse plagioclase-aegirine-perthite-wollastonite-sphene vein in wollastonite-quartz-rock.

There is considerable correlation between the textural relations shown at Carneal, the successively lower temperature-stability ranges and the order of increasing hydration (figure 14).

(*c*) *Metamorphic facies*

The concept of metamorphic facies is applied with some difficulties at Carneal where in the highest prograde phase the contact-metamorphosed limestone falls within the so-called

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sanidinite facies (larnite-merwinite-spurrite subfacies) representing the highest metamorphic temperatures and low pressures.

The name 'sanidinite' facies has been criticized by various authors; it is not very appropriate at Carneal, although the concept if not the name is valid. No single mineral could perhaps be selected as eponymous, but at the highest temperatures larnite (and spinel) are well developed, and melilite and merwinite are of more restricted occurrence. Of the minerals of the retrograde phase, xonotlite, scawtite, tobermorite, thomsonite, and hydrogrossular all occur extensively. In assemblages of these and the other minerals described above the rocks record mixtures of facies within a single specimen, both in the prograde and retrograde phases.

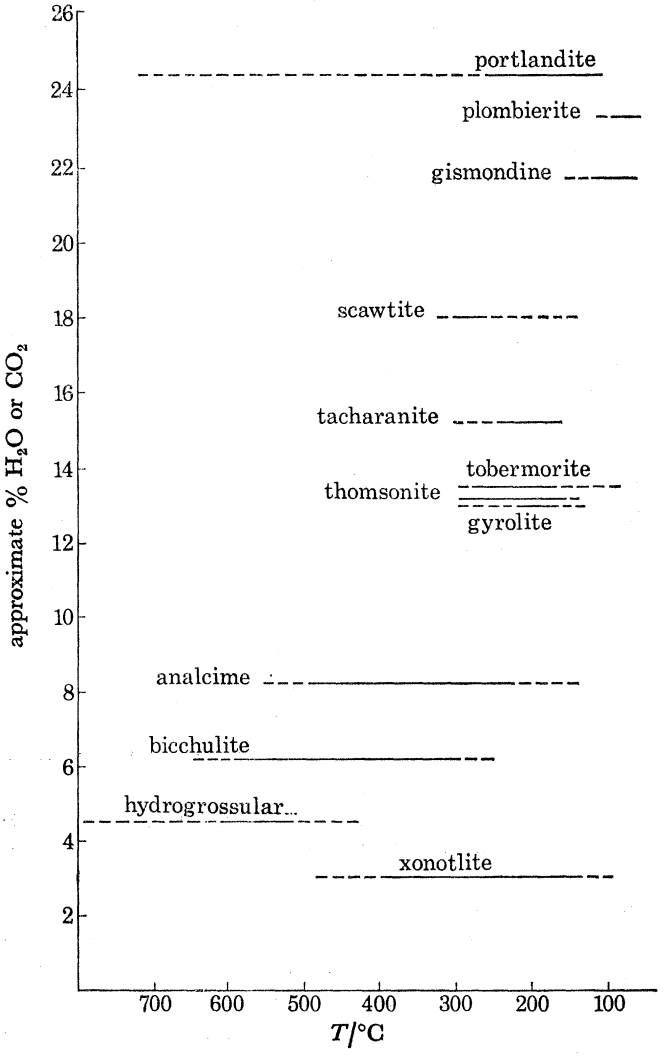

FIGURE 14. Approximate temperature-stability ranges of successive minerals in the retrograde phase. The H_2O and CO_2 contents are taken from standard analyses of the mineral species.

7. CONCLUSIONS

Of the various conditions that result in the production of the contaminated igneous rocks under the prevailing pressure (about 200×10^5 Pa), the presence of the medium-temperature lime silicates shows that chemical composition (or reaction) exercises the controlling influence.

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The addition of lime, and as available, silica from the flint (and the more highly silicic minerals), to the olivine-dolerite magma leads to the development of wollastonite-bearing hybrids rich in diopsidic pyroxene, at the expense of olivine and plagioclase. In the norm of the olivine-dolerite the pyroxene consists of 14.6 % hypersthene, 15.3 % diopside, which in wollastonite-bearing hybrid gabbro becomes 39.2 % diopside, 13.9 % wollastonite, and in the hybrid pyroxenite 45.0 % diopside, 6.3 % wollastonite. The analysis of sahlite from hybrid pyroxenite shows it to contain 70.9 % diopside. Six per cent olivine in the dolerite norm disappears, and plagioclase is markedly reduced, normative anorthite from 37.7 % to 26 %, albite from 17.2 % to 0–3.4 %.

In the olivine-dolerite the Fe/Mg ratio is 1.46, rising in the wollastonite-bearing hybrid pyroxenite and gabbro to 2.13–2.17. In agreement with this, where there is olivine in the pyroxenitic hybrids it is fayalitic. Desilication of the sodic plagioclase molecule leads to the development of late nepheline. Titanaugite-melilite-rock is the eventual product of further lime assimilation by the magma, with Fe/Mg ratio reaching 6.05. Titania rises in the titanaugite to 5.2 % (with lime 22.7 %). In the norm of this rock, nepheline is present to the extent of 11.6 %, and larnite 11.3 %. The rocks are very coarse-grained, as a result of the lowering of viscosity by the carbon dioxide from the chalk. Melilite is derived from diopsidic pyroxene and from plagioclase (albitic and anorthitic components) by further lime assimilation. In the melilite, CaO reaches 34.15 %, and the mineral retains a high soda content (3.20 % Na_2O). The final product is the alkaline residuum of nepheline-aegirine-dolerite, and thomsonite forms a late zeolitic alteration product.

One of the fundamental questions of metamorphism – how much water is involved – is answered for the special case of metasomatic replacement by the evidence of the small veins of merwinite-melilite[-hydrogrossular]. These require the addition of lime and water only, in the proportions of 63 % and 23 % respectively of the olivine-dolerite, with a sufficient mobility to form narrow metasomatically-zoned veins. This metasomatic process involves a

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Description of plate 13

Figure 6. Xonotlite-scawtite-rock. Spherules of scawtite adjacent to cracks in xonotlite. NI 1300A. Crossed polarizers. (Magn. × 80.)

Figure 7. Pyroxenite. Strongly zoned sahlite with (clear) labradorite altered along cracks to thomsonite. Interstitial material includes aegirine, iron ore minerals and nepheline. NI 1616. Partly crossed polarizers. (Magn. × 10).

Figure 8. Wollastonite-bearing hybrid gabbro (analysed rock). Slender prisms of wollastonite, on which coarse pyroxene is moulded, with thomsonitized plagioclase. The pyroxene is centrally pale green to pale brown, zoned marginally to deep brown; some prisms are titanaugite. There is a mesostasis of aegirine, opaque ore and nepheline. NI 1303. Plane polarized light. (Magn. × 10.)

Figure 9. Pyroxenite (analysed rock). Zoned idiomorphic sahlite having deeply coloured borders, with some plagioclase prisms and abundant interstitial thomsonite. NI 1582. Plane polarized light. (Magn. × 18.)

Figure 10. Metasomatically zoned merwinite vein cutting olivine-basalt. The central part of the merwinite vein (right centre) has coarse merwinite prisms. Olivine-basalt (left and extreme top right) has small (clear) olivine phenocrysts. Clear melilite prisms are abundant in melilite-hydrogrossular-merwinite zones. I 1313. Plane polarized light. (Magn. × 5½.)

Figure 11. Margin of metasomatic vein against olivine-basalt. The olivine-basalt (left) has a parallel orientation of plagioclase laths which is cut by the sharp metasomatic front. In the vein the laths continue in the same orientation but are composed of melilite, although this better seen under the microscope than in the photomicrograph. I 1313. Plane polarized light. (Magn. × 27.)

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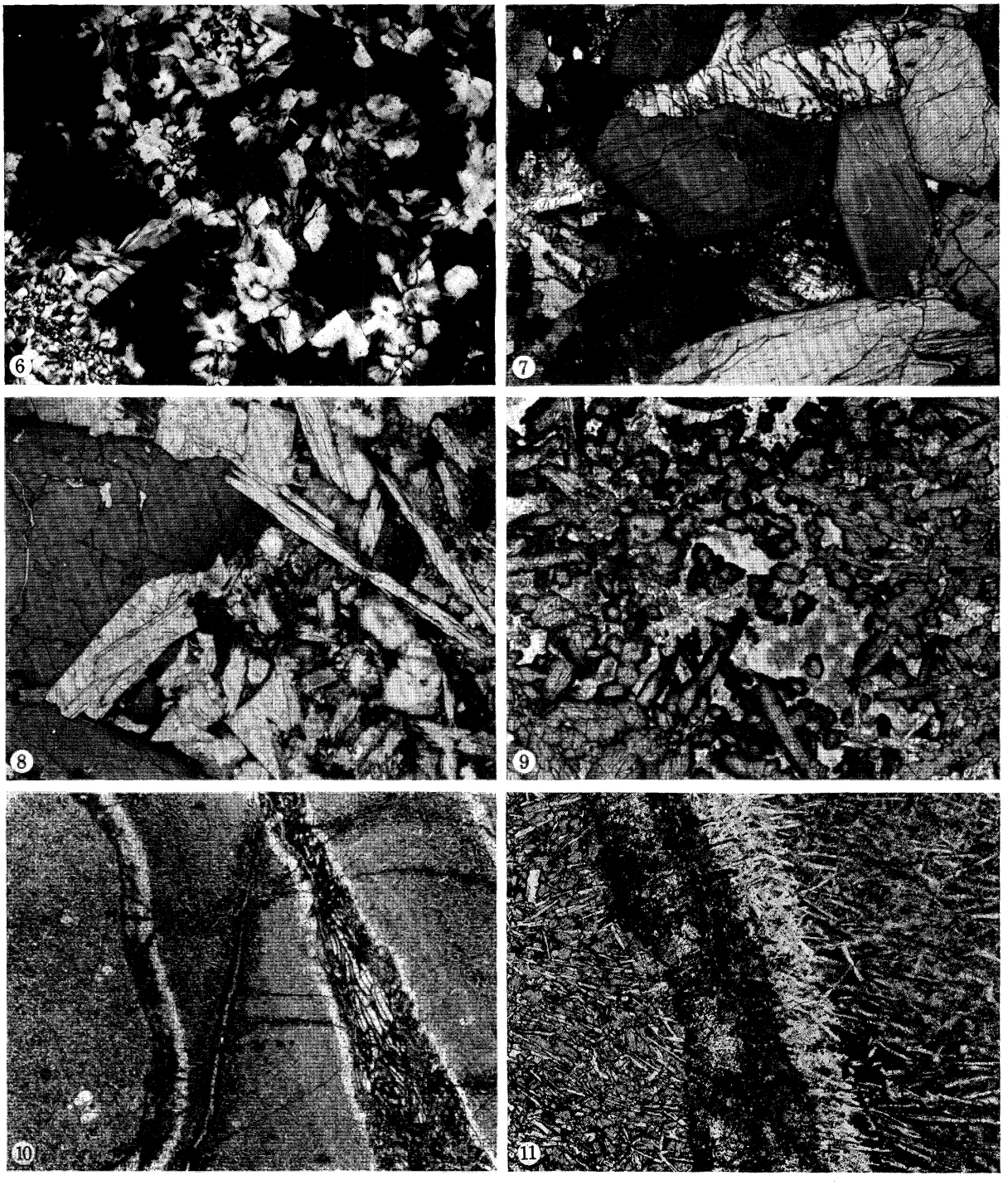

PHOTOMICROGRAPHS OF CARNEAL ROCKS

FIGURES 6–11. For description see opposite.

(*Facing p.* 266)

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considerably greater quantity of lime than in the production of the hybrid pyroxenite and gabbro by assimilation (CaO 18–26 %, with SiO_2 17–18 %) and a limited amount of water. In the exomorphic rocks wollastonite is the typical product of reaction of silica from flint with limestone, and the other complex associations are also formed at high temperatures. In the retrograde phase there are a number of hydrous calcium and related silicates formed, partly rare species.

The assimilation of limestone has been held to be the source of alkaline or peralkaline rocks, notably by Shand & Daly. However, the occurrence at Carneal, and others of comparable size, while showing that the reaction of limestone with basic magma is a mechanism for the production of alkaline-rocks, provide a convincing argument, as Tilley has pointed out, that the scale on which this mechanism operates is only very small. The direct evidence in the form of inclusions or chemical heterogeneity at the present levels of exposure is against this mechanism operating, at least to moderate pressures and depths for the production of alkali-rocks, especially of the huge masses of only slightly alkaline types. Since there is no evidence that the mechanism is of widespread importance under conditions of moderate pressure and depth, there is no reason to conclude that it was operable at much deeper levels and pressures where direct evidence would have been destroyed. The mechanisms revealed at Carneal are on a small scale, and rocks of the type found there remain extemely rare.

The authors are indebted to Dr S. O. Agrell for generously presenting specimens he had collected and lending thin sections, and to Dr R. Dearnley, Dr R. R. Harding, Mr R. K. Harrison, Dr J. R. Hawkes, Mr N. J. Fortey, Dr B. G. Jamieson (and the referee) for constructive criticism. Mr J. M. Pulsford took the photographs and photomicrographs. Mr H. E. Wilson who, with Dr E. J. Cobbing, discovered the plug, provided much advice. The paper is published by permission of the Director, Institute of Geological Sciences.

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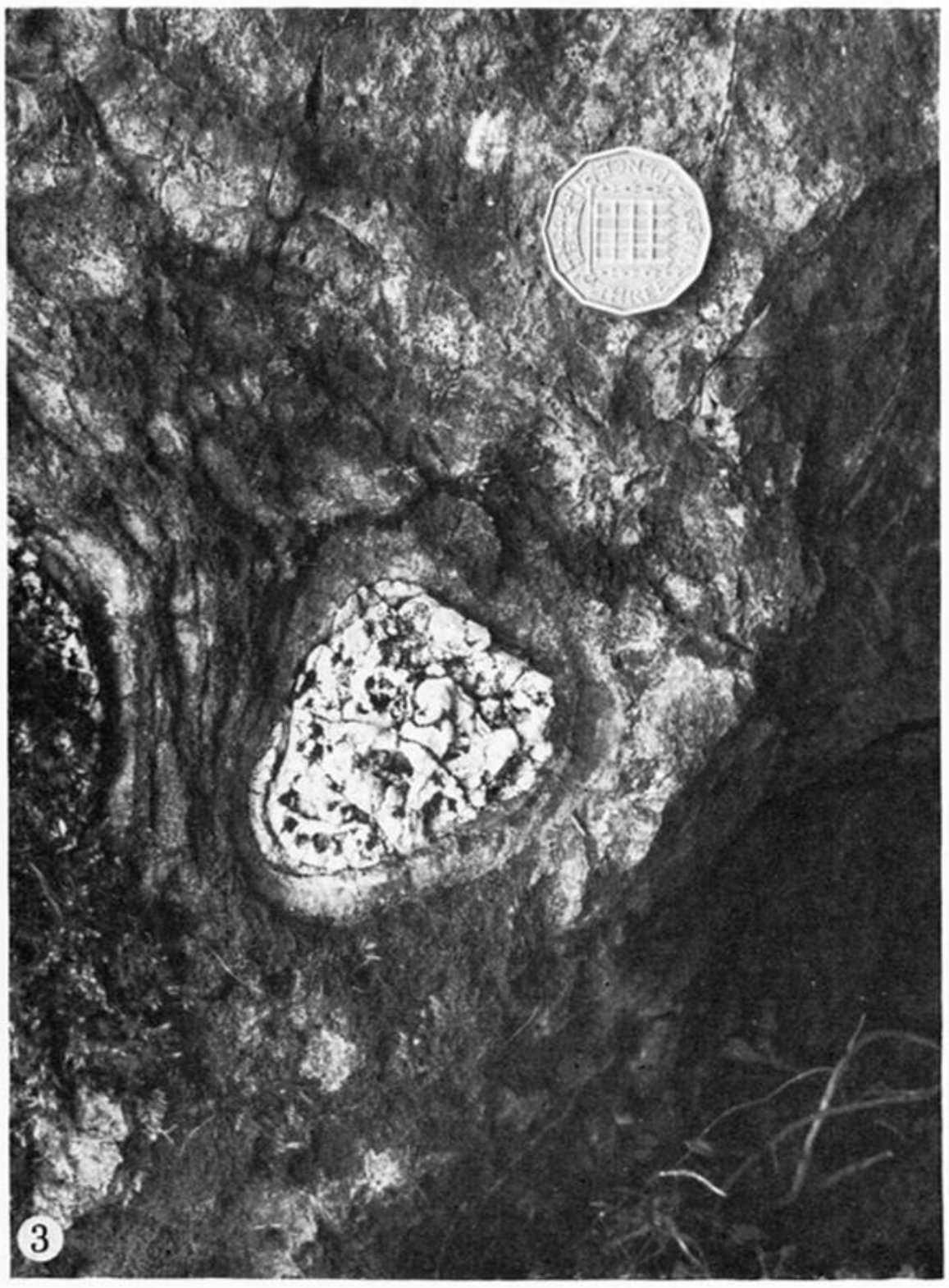

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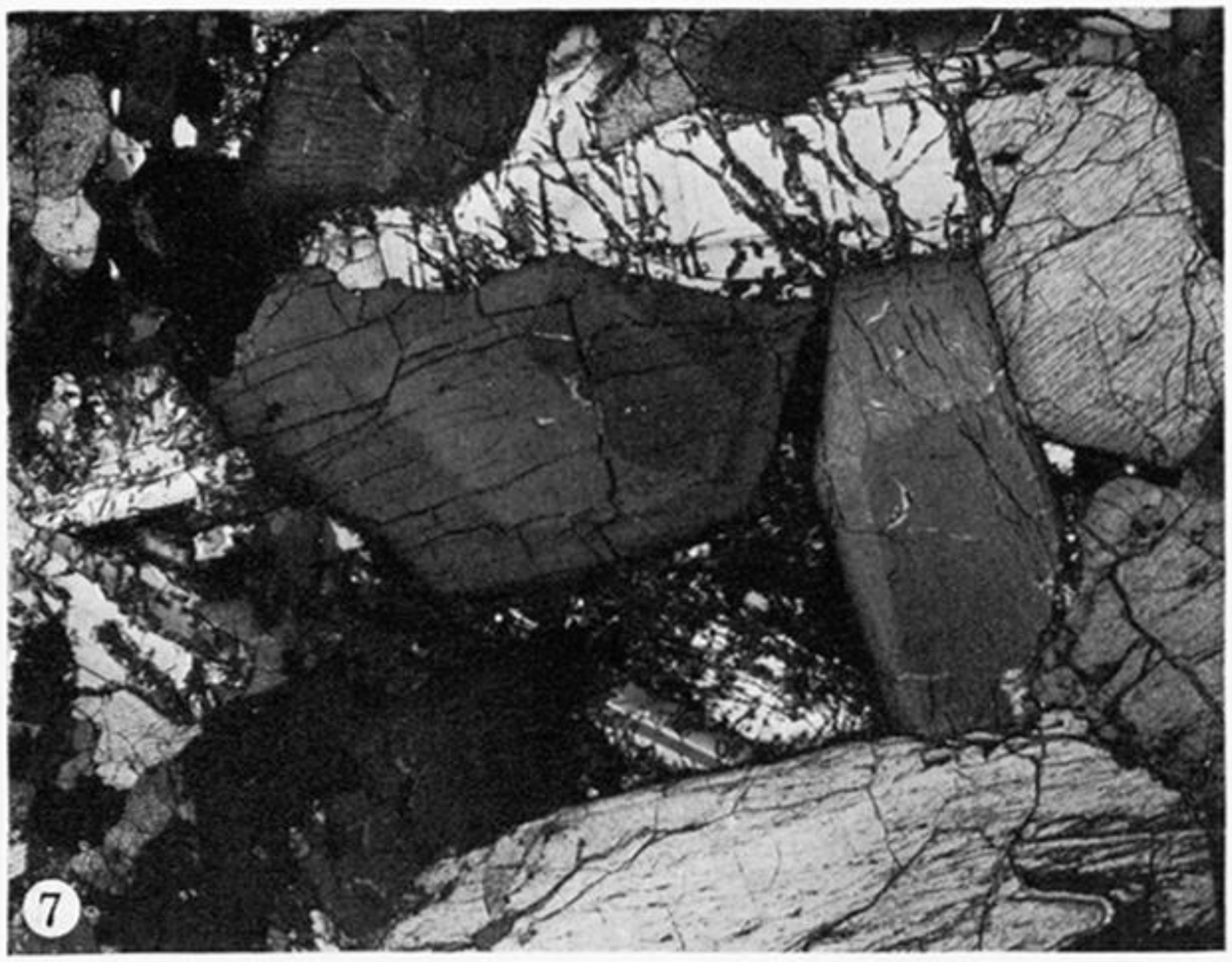
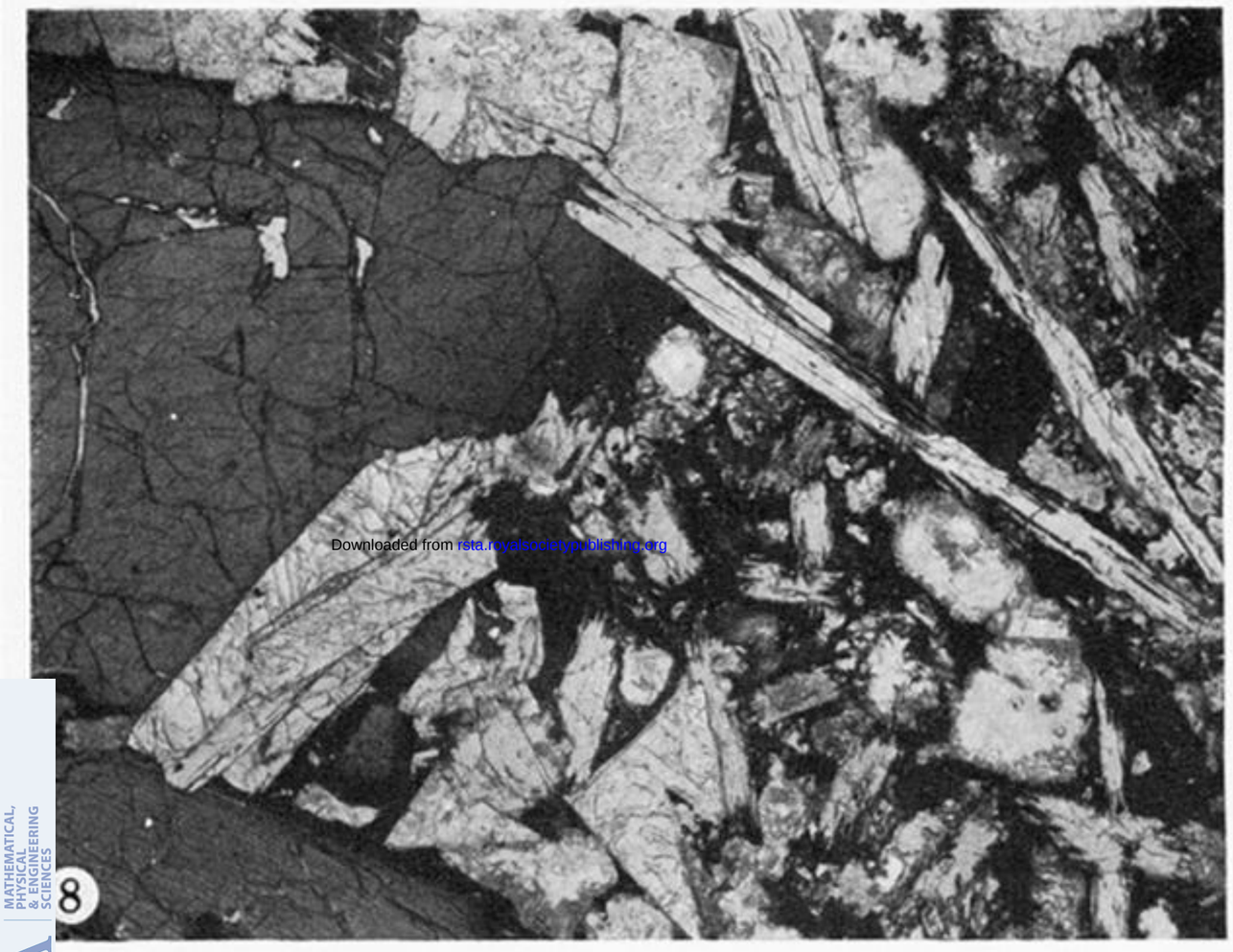
FIGURE 3. Reaction rims around metamorphosed flint nodules. N. face of Carneal plug. The coin measures 20 mm in diameter. Negative NI 349.

FIGURE 4. Cyclostome bryozoan now composed of wollastonite, in metamorphosed flint. NI 1301A. Magn. ×48. Negative M2824.

FIGURE 5. The Carneal Plug seen from the north across an alluvial flat. The west face of the plug is visible among the trees on the right. Negative NI 347.





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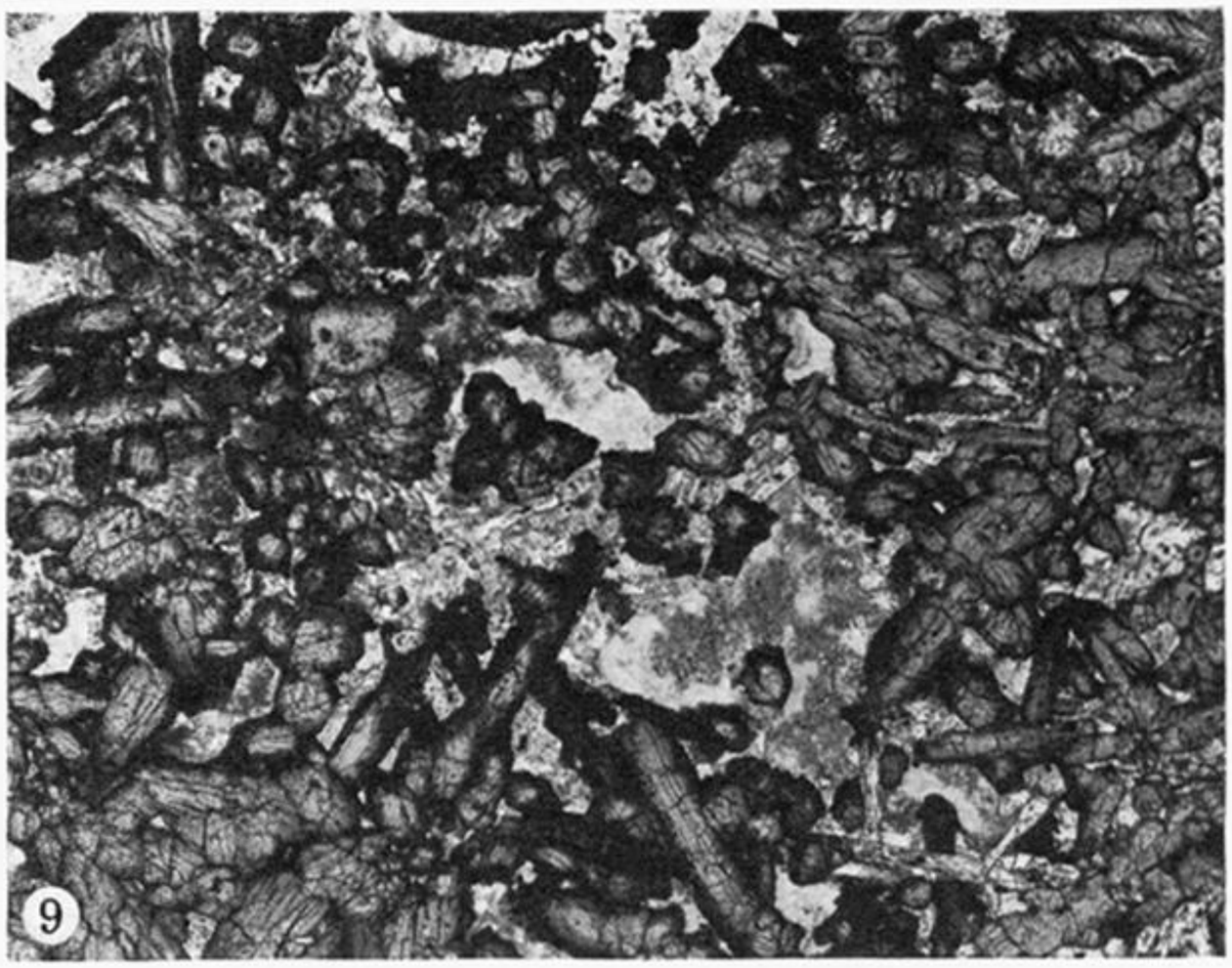

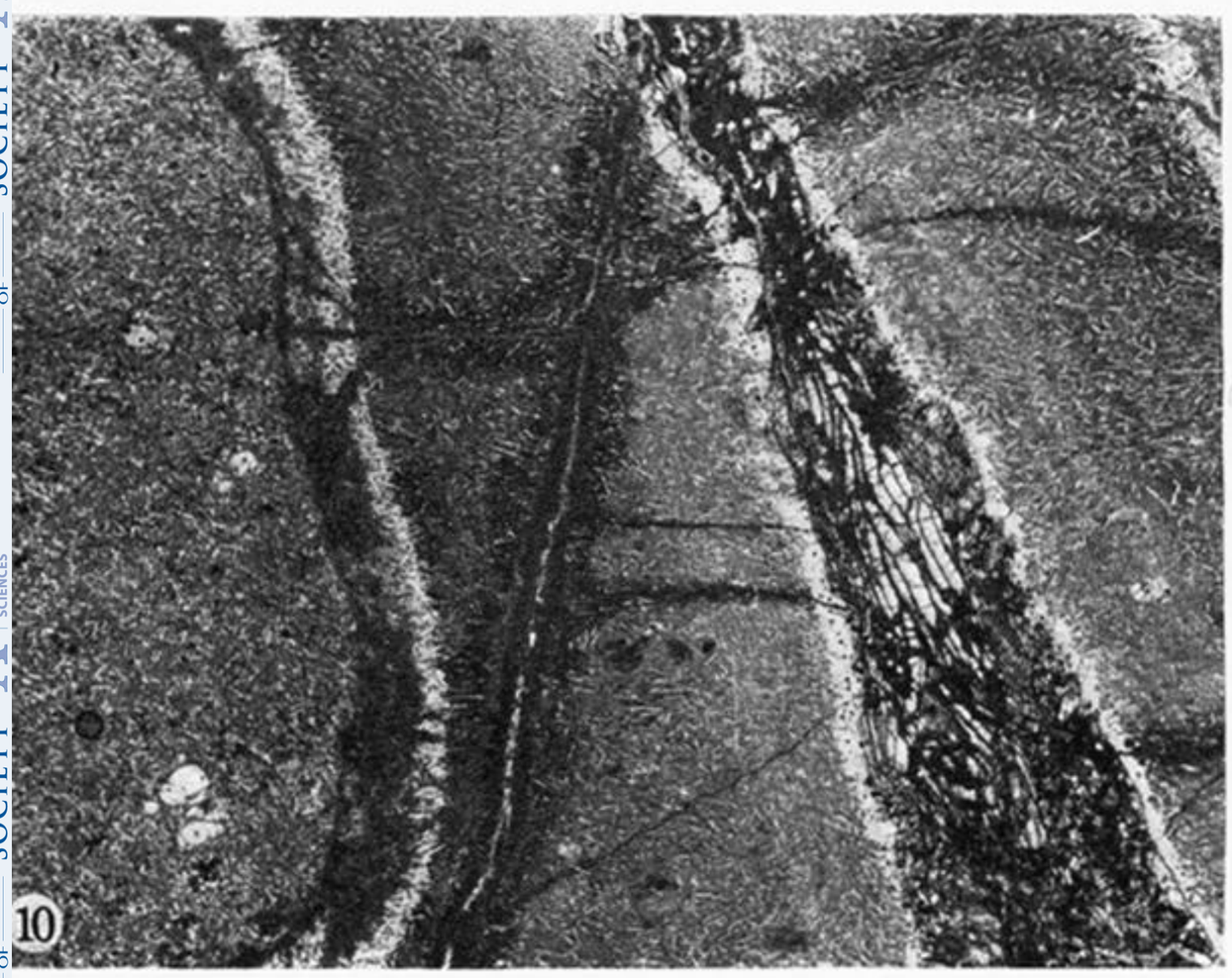

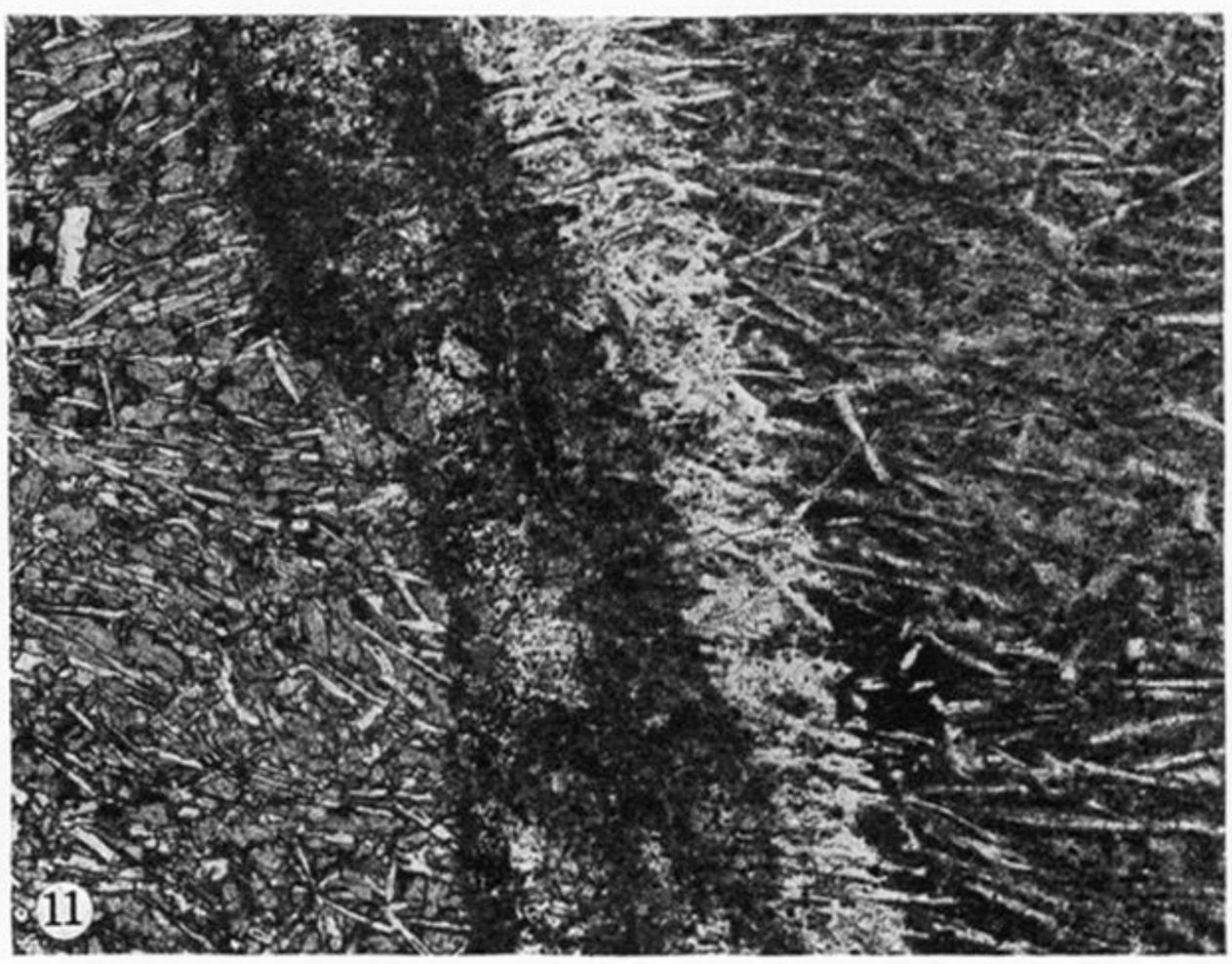

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PHOTOMICROGRAPHS OF CARNEAL ROCKS

FIGURES 6–11. For description see opposite.