

Clinopyroxenes from the Upper Layered Series Kap Edvard Holm, East Greenland

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SUMMARY. Chemical compositions, unit-cell contents, and optical properties of six clinopyroxenes from the Upper Layered Series are presented. There is a gradual enrichment in iron upwards in the intrusion although there is no measurable effect on the optical properties, possibly because of Cr variation and exsolution of Fe and Ti as oxides. The chemistry of the clinopyroxenes is consistent with crystallization from a water-rich magma of transitional nature between tholeiite and alkali-olivine basalt. X-ray oscillation photographs of single crystals reveal the presence of sub-microscopic exsolution lamellae of pigeonite, a feature consistent with the transitional nature of the parent magma. Exsolution lamellae of titanomagnetite are ubiquitous and were formed at subsolidus temperatures in response to high buffered oxygen fugacity. Comparisons are made between the properties of clinopyroxenes from the Lower, Middle, and Upper Layered Series, and the conditions of crystallization of each series.

THE Kap Edvard Holm complex, consisting of layered gabbros and later quartz-bearing syenites, is situated on the west side of Kangerdlugssuaq Fjord, East Greenland. From an analysis of the structure of the gabbros, a Lower, Middle, and Upper Layered Series can be distinguished (Elsdon, 1969). The clinopyroxenes of the Lower and Middle Layered Series have already been described (Deer and Abbott, 1965). In this paper the clinopyroxenes of the Upper Layered Series are described, some general comparisons made, and their petrogenesis considered in the light of recent experimental data.

The clinopyroxenes occur in plagioclase-augite-olivine orthocumulates and plagioclase-augite orthocumulates (Wager, Brown, and Wadsworth, 1960). A detailed account of the petrography and chemistry of these rocks will be presented at a later date. Cumulate texture is usually well developed and the augite sometimes occurs as glomeroporphyritic aggregates. In a few rocks, however, the texture is more accurately described as sub-ophitic.

The augite is pale brown in colour, non-pleochroic, and invariably contains other minerals, notably Fe-Ti oxide inclusions and pargasite. The Fe-Ti oxide inclusions are almost ubiquitous, and are found in the Lower and to a lesser extent in the Middle Layered Series. They occur either as irregular trails of blebs or as sets of finely spaced lamellae parallel to the *c*-axis and at approximately 80° to it. The blebs are up to 0.1 mm in size, the lamellae may be 1 mm in length but are less than 0.1 mm in width.

Optical properties. Chemical analyses of six augites from the Upper Layered Series, together with β and $2V_p$, are in table I. The β index was measured by oil immersion, using sections showing a slightly off-centred acute bisectrix figure (Hess, 1949).

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Each value quoted is the mean of six determinations and is considered accurate to ± 0.001 . $2V_\gamma$ was determined using a 4-axis Universal Stage and is the mean of four readings; it is considered accurate to $\pm 2^\circ$.

The results show that values of β vary slightly but irregularly between 1.703 and 1.706, but are comparable with those of the Middle and Lower Layered Series (Deer and Abbott, 1965). The replacement of Fe by Mg is normally accompanied by a significant increase in β ; using the regression coefficients calculated by Winchell (1961), an augite of composition $Wo_{43}En_{44}Fs_{13}$ should have a value of β 0.009 higher than one of composition $Wo_{41}En_{39}Es_{20}$. No such increase is recorded in the Upper Layered Series augites, and this could be because the refractive indices in Mg-rich clinopyroxenes are very sensitive to variations in the Cr-content (Abbott 1962); an increase in Cr_2O_3 from 0.1 to 0.6% causes β to increase by 0.003. Thus, increasing Fe and decreasing Cr in Mg-rich clinopyroxenes might be expected to cancel each other out in their effects on the refractive indices. On the other hand as the chemical analyses include the Fe-Ti oxide inclusions that have been exsolved from the augite, the pyroxenes are more Mg-rich than the bulk analyses would suggest, suppressing the effect on optical properties of the substitution $Mg \rightleftharpoons Fe$, and so chemical analyses of the augite plus oxide inclusions gives the closest possible approximation to the initial augite composition.

Chemistry. The chemical analyses of the augites include the oxide exsolution and give the closest approximation to the initial augite composition. Comparison of the analyses with those of augites from the Middle and Lower Layered Series shows that the three series have closely similar chemical and optical properties. The results indicate that the higher Al_2O_3 and TiO_2 contents of the Kap Edvard Holm augites lead to higher refractive indices than those from, for example, the Skaergaard and Stillwater intrusions.

There are small differences in the chemistry of the augites from the three layered series: The Middle Layered Series augites have higher Al_2O_3 contents (LLS 3.4–4.1%, MLS 3.4–6.5%, ULS 2.7–4.0%); the Lower Layered Series augites are more calcic than those of the Middle Layered Series (20.5–20.95% and 19.45–20.5% CaO respectively); and the CaO-content of the Upper Layered Series augites is more variable than for the other two units, ranging from 19.3 to 21.1%.

Le Bas (1962) has suggested that the Al_2O_3 and SiO_2 contents of augites from basic igneous rocks can be correlated with the chemical characteristic of the parental magma. Basaltic magmas with alkaline affinities crystallize augites richer in alumina than those of tholeiitic parentage; exceptions to this general rule do exist, however, for example, the augites from tholeiitic basalts of Hawaii. Similarly, augites derived from alkali-basalt magmas tend to contain more TiO_2 than tholeiitic varieties, consequent upon the replacement of $Si^{[4]}$ by $Al^{[4]}$ (Hartman, 1969). The Kap Edvard Holm augites, plotted on the diagrams used by Le Bas (1962), straddle the boundary between the 'non-alkaline' and 'alkaline' fields, whereas augites of similar compositions from the Skaergaard intrusion lie well within the non-alkaline field (figs. 1 and 2). Thus the Kap Edvard Holm augites show transitional affinities; and the Middle Layered Series parental magma is considered to have been more alkaline in character than those of the Lower and Upper Layered Series.

TABLE I. *Upper Layered Series clinopyroxenes: chemical compositions*

	1	2	3	4	5	6
SiO ₂	50.73	50.48	49.96	50.47	49.44	50.09
Al ₂ O ₃	3.61	3.32	3.96	2.71	3.45	3.72
Fe ₂ O ₃	1.24	2.25	1.58	1.58	1.07	1.89
FeO	7.70	7.17	8.23	8.06	11.39	8.85
MnO	0.26	0.24	0.16	0.27	0.32	0.30
MgO	14.48	14.58	14.10	13.30	13.22	13.46
CaO	19.46	20.63	20.55	21.08	19.29	20.08
Na ₂ O	0.49	0.46	0.41	0.47	0.43	0.43
K ₂ O	0.10	0.02	0.03	0.03	0.06	0.04
H ₂ O ⁺	0.49	0.07	0.33	0.22	0.06	0.40
H ₂ O ⁻	0.10	0.03	0.20	—	0.18	0.04
TiO ₂	1.45	1.13	0.54	1.18	1.08	1.04
Total	100.14	100.34	100.16	99.37	99.99	100.38
<i>Atomic %</i>						
Ca	41.7	42.1	42.9	44.6	40.6	41.9
Mg	43.2	41.3	40.9	39.2	38.7	39.0
(Fe ³⁺ , Fe ²⁺ , Mn)	15.1	16.6	16.2	16.2	20.7	19.1
<i>Unit cell contents on the basis of 6(O,OH)</i>						
Si	1.873	1.875	1.859	1.899	1.870	1.876
Al ^[4]	0.127	0.125	0.141	0.101	0.130	0.124
Al ^[6]	0.030	0.020	0.033	0.019	0.024	0.041
Ti	0.040	0.062	0.015	0.033	0.031	0.058
Fe ³⁺	0.034	0.094	0.048	0.045	0.030	0.080
Mg	0.797	0.807	0.782	0.746	0.745	0.749
Fe ²⁺	0.238	0.223	0.256	0.254	0.360	0.277
Mn	0.008	0.008	0.005	0.009	0.010	0.009
Ca	0.770	0.821	0.820	0.850	0.782	0.806
Na	0.033	0.008	0.030	0.034	0.032	0.016
K	0.002	0.001	0.001	0.001	0.003	0.001
OH	0.113	0.009	0.082	0.025	0.015	0.054
O	5.887	5.991	5.918	5.975	5.985	5.946
% Al ^[4]	6.4	6.3	7.1	5.1	6.5	6.2
<i>Optical properties</i>						
β	1.704	1.705	1.705	1.703	1.703	1.706
2V _γ	50°	52°	47°	48°	52°	50°
<i>C.I.P.W. norms</i>						
Or	0.59	0.12	0.18	0.18	0.35	0.24
Ab	4.15	3.67	3.37	3.98	3.64	3.68
Ne	—	—	0.06	—	—	—
An	7.36	7.23	8.88	5.20	7.31	8.07
Di	71.36	75.92	74.79	79.81	71.85	73.67
Hy	7.38	3.07	—	1.87	2.83	4.96
Ol	4.13	4.99	8.85	3.59	10.17	4.70
Mt	1.80	3.24	2.49	2.29	1.55	2.59
Il	2.75	1.97	1.03	2.24	2.05	1.97

1. E.G. 7030, bottom of Junction Nunatak, 2950 m. Anal. R. Elsdon.

2. E.G. 6604, New Nunatak, 4500 m. Anal. M. Haslop.

3. E.G. 7071, top of Birthday Nunatak, 5250 m. Anal. R. Elsdon.

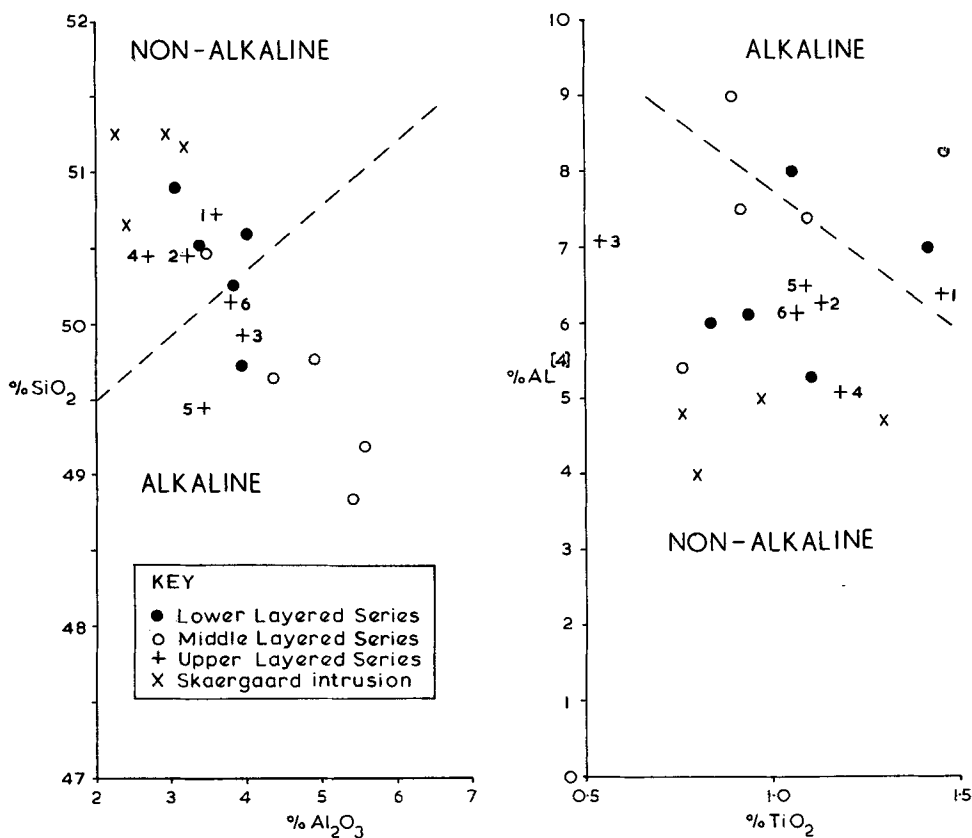
4. E.G. 7131, southern end of Alex' Mountain, 6600 m. XRF anal. by R. Elsdon.

5. E.G. 7109, southern end of Alex' Mountain, 6650 m. Anal. R. Elsdon.

6. E.G. 6559, Snow Dome, 7000 m. Anal. M. Haslop.

Optical properties of 2 and 6 determined by D. Abbott.

Further evidence of transitional characteristics of the Kap Edvard Holm parental magma is illustrated when the augite compositions are plotted on the 'pyroxene quadrilateral' (fig. 3). The trends lie between those of the Skaergaard augites (Ca-poor) and the alkaline trend parallel to the join diopside-hedenbergite (Wilkinson, 1956). The Na_2O and K_2O contents are, however, those characteristic of tholeiitic augites.



FIGS. 1 and 2. FIG. 1 (left). Plot of % SiO_2 against % Al_2O_3 for augites from the Kap Edvard Holm complex and the Skaergaard intrusion. Fields outlined from Le Bas (1962). FIG. 2 (right). Plot of % tetrahedral sites occupied by Al^{3+} against w % TiO_2 for augites from the Kap Edvard Holm Complex and the Skaergaard intrusion. Fields outlined from Le Bas (1962).

Finally, these transitional characteristics are also demonstrated by the 'norms' of the Kap Edvard Holm augites. In tholeiitic augites, hypersthene predominates over olivine: in those of alkaline rocks nepheline is present in the norm, and calcium orthosilicate and leucite are also occasionally present. Table I shows that the Upper Layered Series augites are either slightly nepheline-normative, or else olivine predominates over hypersthene. In this respect they are similar to the augite from the 'transitional' basalts of the Mid-Atlantic Ridge (Muir and Tilley, 1964).

Petrogenesis

The Kap Edvard Holm augites have three general characteristics: the restricted range of iron-enrichment compared with augites from the other basic intrusions such as Skaergaard, the absence of hypersthene as a discrete primary phase (it occasionally forms reaction rims to olivine crystals), and the presence in the Upper Layered Series augites of sub-microscopic exsolution lamellae of pigeonite.

In the Lower and Middle Zones of the Skaergaard intrusion the percentage of FeSiO_3 in the augite increases from 17 to 28 over a thickness of 1500 m: in the Upper Layered Series, however, the increase is only from 14 to 21 over 4000 m. It is not possible to relate this to differences in initial magma compositions since no chilled margins are present at Kap Edvard Holm (Elsdon, 1969). The water content of the Upper Layered Series magma was high, as can be judged from autometasomatic alteration of primary minerals over a wide temperature range and from calculations based on magnetite-ilmenite equilibration data (Andersen, 1968). Thus a high f_{O_2} restricted the enrichment in the magma of Fe^{2+} , shown by the relatively early appearance of cumulus magnetite.

Studies by Schairer and Yoder (1962) on the system $\text{CaMgSi}_2\text{O}_6$ – $\text{CaFeSi}_2\text{O}_6$ have shown that for Mg-rich augites crystallizing from a liquid the initial rate of iron enrichment is greater for the liquid than for the crystals nucleating from it. High f_{O_2} in a natural magma, however, will inhibit the increase of Fe^{2+} in the liquid to some extent, preventing a significant increase in the ferrosilite content of the crystallizing augite.

Crystallization of augite without accompanying hypersthene is usually indicative of alkali-basalt magmas, although this is not always so. Brown (1957) has postulated three ways in which a tholeiitic magma can give rise to assemblages containing only one pyroxene during the *later* stages of differentiation: reaction between initial Ca-poor pyroxene and liquid to give olivine; the binary eutectic between the joins Di–He and En–Fs ceases to exist, being replaced by a solid solution with a minimum; or the liquidus minimum in the pyroxene quadrilateral migrates towards one of the solvus limbs, when either a Ca-poor or a Ca-rich pyroxene crystallizes. The available chemical evidence indicates that even for augites as magnesian as $\text{Wo}_{43}\text{En}_{46}\text{Fs}_{11}$ there was no simultaneous crystallization of a primary calcium-poor pyroxene. Thus, unless one of the above mechanisms can be applied to the early magmatic stage, it is necessary to

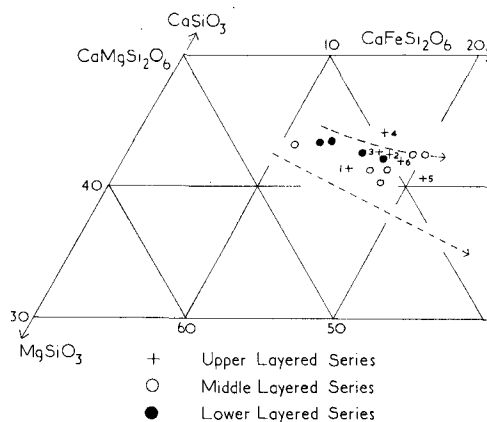


FIG. 3. Plot of compositions of augites from the Kap Edvard Holm complex on the 'pyroxene quadrilateral'. Lower broken line Skaergaard pyroxene trend; upper broken line trend of pyroxenes of alkali-olivine-basalt magmas (Wilkinson, 1956).

conclude that either the chemistry of the parent magma or the inherent physical conditions, or both, were the dominant factors in the control of the pyroxene crystallization.

Although the augites of the Upper Layered Series are optically homogenous, X-ray oscillation photographs of some augites, using the *c*-axis as rotation axis, reveal exsolution of pigeonite. The chemistry of the augites indicates that the percentage of pigeonite can be only small. It is impossible to state with certainty whether the pigeonite has inverted to hypersthene, but the obviously thin width of the lamellae (submicroscopic) would be a restraining influence on the tendency for inversion when the temperature was sufficiently low for pigeonite to become metastable. It is interesting to note that optically visible lamellae of uninverted pigeonite have been observed in augite from the pre-Cambrian diabase of Beaver Bay (Muir, 1954).

Boyd and Schairer (1962) and Kushiro and Schairer (1963) have demonstrated that augites that crystallize at lower temperatures are likely to contain less $(\text{Mg, Fe})\text{SiO}_3$ in solid solution than those that crystallize at higher temperatures. Based on the absence of optically visible exsolution lamellae of Ca-poor pyroxene in the Kap Edvard Holm augites, Deer and Abbott (1965) concluded that the crystallization temperatures were lower than at Skaergaard, due to depression of the liquidus and solidus consequent upon higher water pressure. The discovery of submicroscopic lamellae of pigeonite confirms this conclusion. Unless diffusion was in some way drastically inhibited, thus preventing the widespread exsolution of the Ca-poor pyroxene and leaving the augite supersaturated with it, then the quantity present in the augite as it crystallized was probably quite small. The bulk pyroxene composition must thus cut the solvus in the relevant $(\text{Mg, Fe})\text{SiO}_3$ - $\text{Ca}(\text{Mg, Fe})\text{Si}_2\text{O}_6$ composition plane at a temperature not far above the minimum temperature at which diffusion is effective at the cooling rate of the Upper Layered Series (fig. 4). At Skaergaard, however, exsolution lamellae of Ca-poor pyroxene in augite are common and the augites are less calcic than the Kap Edvard Holm augites, giving a larger temperature interval between the intersection of the bulk clinopyroxene composition with the solvus and the minimum temperature of effective diffusion, assuming this to have been the same as for the Upper Layered Series at Kap Edvard Holm.

It has already been noted that the Ca-content of the Middle Layered Series pyroxenes is slightly lower than that for the Lower Layered Series: comparison with the Upper Layered Series is difficult in view of the variable Ca-contents of these augites. From the above, however, it would appear that the Lower Layered Series magma had a higher water content than the Middle Layered Series magma, and this conclusion is supported by the relative paucity of Fe-Ti oxide needles in the Middle Layered Series augites, also by the less frequent hydrothermal alteration of the primary minerals. It might be expected, therefore, by comparison with the augites of the Upper Layered Series, that submicroscopic exsolution of Ca-poor pyroxene should also be a feature of the Lower Layered Series augites and possibly of those from the Middle Layered Series. There is as yet no X-ray evidence to support these speculations.

It would appear, from the previous discussion, that augites from alkali-olivine

basalts have lower crystallization temperatures than those from high alumina and tholeiitic basalts. This is not confirmed by melting experiments on tholeiitic and alkali basalts (Tilley, Yoder, and Schairer, 1965), which show that the clinopyroxene compositions are related to the magma composition itself as well as to the physical conditions under which crystallization took place. With the Kap Edvard Holm pyroxenes

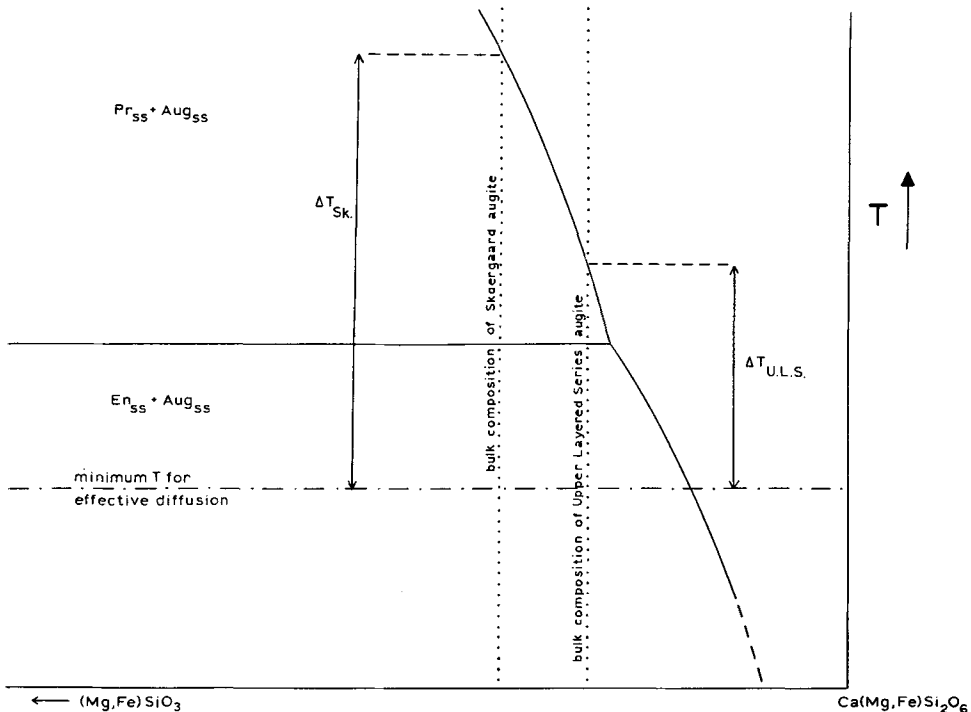


FIG. 4. Hypothetical diagram showing relationship between exsolution of Ca-poor pyroxenes from Skaergaard and Upper Layered Series augites.

the alkaline affinities of the magma resulted in the crystallization of a single pyroxene (almost invariably) and controlled the amount of (Mg,Fe)SiO₃ in solid solution in the augite, but comparisons between the augites of the three units in the Kap Edvard Holm complex show that water pressure, by controlling liquidus and solidus temperatures, exerted a further control on the bulk composition of this phase.

Exsolution of Fe-Ti oxide

Needles and irregular blebs of opaque minerals are a common feature of the Kap Edvard Holm augites (Abbott, 1962), and in the Upper Layered Series, very few of the augites are completely free of them. Electron-probe traverses on the needles (Abbott, 1962) show that they consist principally of iron with lesser amounts of titanium. X-ray *c*-axis oscillation photographs of these augites show extra diffraction maxima

corresponding to magnetite in the following orientations (Bown and Gay, 1959): (111) magnetite parallel to (100) augite and ($\bar{1}\bar{1}3$) magnetite parallel to (001) augite. Although it cannot be stated with certainty whether these maxima are due to the oxide needles, or blebs, or both, it is probable that the extra maxima are due to titanomagnetite exsolved in response to changing physical conditions.

Three possible mechanisms for this exsolution process will be considered: The replacement of Fe^{2+} by Mg^{2+} in the pyroxene could take place during the magmatic stage or as a subsolidus process. However, no mechanism is known whereby the equilibrium augite composition could become more magnesian during crystallization with falling temperature in a closed system. Influxes of more basic magma would lead to a break in the cryptic layering, which has not been observed. Objections also arise if this substitution is presumed to occur below the solidus, as volume considerations preclude the possibility of increasing the MgSiO_3 content if the expelled iron is retained within the crystals. Secondly, the substitution Fe^{3+} for Fe^{2+} would necessitate a second ionic substitution, e.g. Na^+ for Ca^{2+} , in order to maintain electrostatic neutrality. The alkali contents of all the analysed pyroxenes are fairly low and do not increase with increase of Fe^{3+} . The relatively high Al^{3+} and Ti^{4+} contents are considered to be due to the alkaline affinities of the parental magma. Thirdly, the oxidation of the ferrosilite component with $6\text{FeSiO}_3 + \text{O}_2 \rightarrow 2\text{Fe}_3\text{O}_4 + 6\text{SiO}_2$ could, at first consideration, have occurred during the magmatic or post-magmatic stage. However, the oxygen fugacity in the crystals on the floor of the magma chamber is not considered to have altered appreciably during crystallization; the cumulates exhibit, in their bulk chemistry, a slow and ill-defined increase upwards in the ratio $\text{Fe}^{3+}:\text{Fe}^{2+}$ reflecting the physio-chemical environment in which crystallization took place. The possibility of subsolidus oxidation of augite remains, and on a thermodynamic basis Mueller (1965) predicted that, at subsolidus temperatures, the equilibrium Mg:Fe ratios of silicates co-existing with magnesioferrite would rise with decreasing f_{O_2} . Subsequently, experimental work on the system $\text{MgO}-\text{Fe}-\text{O}-\text{SiO}_2$ (Speidel and Osborn, 1967; Speidel and Nafziger, 1968) has supported these predictions by showing that at subsolidus temperatures, the Mg:Fe ratio in Ca-free pyroxene can increase with appropriate f_{O_2} or df_{O_2}/dT values.

It is therefore suggested that exsolution of Fe-Ti oxide from pyroxene is due to an increase in the equilibrium ratio Mg:Fe as a function of df_{O_2}/dT , causing oxidation of the ferrosilite component and thus leading to an increase in the enstatite component. This assumes, however, that augite and Ca-poor pyroxene behave similarly under these conditions: further work on the system $\text{CaO}-\text{MgO}-\text{Fe}-\text{O}-\text{SiO}_2$ is needed to verify the assumption.

Presumably, the volatiles acted at subsolidus temperatures so that the oxygen fugacity did not fall with temperature as fast as it would if controlled by a solid buffering assemblage. A possible cause is that dissociation of water led to rapid loss of hydrogen from the system thus lowering df_{O_2}/dT , a feasible process in view of the greater diffusion rate of hydrogen than either oxygen or water. It is also possible that f_{O_2} was influenced by the high water content of the magma, calculated by two different methods to be approximately 5%. Another gabbroic intrusion where the clinopyroxene

contains exsolution lamellae of Fe-Ti oxides is at Kaerven, East Greenland, where the parental magma is again considered to have been relatively rich in water (Ojha, 1966).

If oxidation of the ferrosilite component is the correct mechanism, it remains to consider the fate of the silica released by the production of magnetite. One oscillation photograph of an augite crystal from EG7071 (table I) showed numerous extra diffraction maxima that could only be correlated with one or more of the polymorphs of silica. Work on the oxidation of olivine has revealed that in iron-rich varieties the silica released passes first through an amorphous phase, then with incipient crystallization to an embryonic silica structure and finally by recrystallization to quartz, tridymite, or cristobalite or a combination of these minerals (Gay, personal communication). Many oscillation photographs of the Upper Layered Series augites, however, show no features that can be correlated with crystalline polymorphs of silica. Either the silica is present in an amorphous form, or else it has escaped from the crystals by diffusion of Si and O atoms. Super-critical water is capable of dissolving appreciable quantities of SiO₂ (Morey, 1957). No further evidence is available at this stage on the fate of the silica.

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