

Iron–titanium oxide minerals in the Upper Layered Series, Kap Edvard Holm, East Greenland

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SUMMARY. Electron-probe data are presented for coexisting magnetite and ilmenite from seven gabbro cumulates and a gabbro pegmatite, and bulk FeO–Fe₂O₃–TiO₂ analyses for ten magnetite–ilmenite assemblages. The oxides equilibrated at 580 to 720 °C and oxygen fugacities of 10^{-15.0} to 10^{-20.5} bar. Late-stage alteration, associated with growth of hydrothermal silicates, resulted in the breakdown of ilmenite to rutile and the dissolution of magnetite. Higher levels in the intrusion equilibrated at lower temperatures than deeper levels. All features described are consistent with the retention of the initial water content of the magma within the walls of the intrusion.

THE Upper Layered Series of the Kap Edvard Holm complex consists of plagioclase–augite–magnetite and plagioclase–augite–olivine–magnetite orthocumulates. The presence, throughout the exposed succession, of cumulus magnetite indicates a high Fe³⁺:Fe²⁺ ratio in the crystallizing magma and this is considered to be the result of a high water content.

Other features associated with the high water content are, firstly, the ubiquitous occurrence of pargasitic amphibole as a primary crystallizing phase; secondly, extensive hydrothermal alteration of the primary minerals over a wide range of temperature during cooling, including metasomatic formation of quartz–albite–epidote–ilmenite rock (Elsdon, in preparation); thirdly, variations in the sites of initial primocryst nucleation (Elsdon, 1970, 1971a).

The petrographic and chemical features shown by the Fe–Ti oxides of the Upper Layered Series afford information on the physico-chemical conditions prevailing during crystallization of the magma and on the cooling history of the rocks down to temperatures at which plagioclase was replaced by albite+epidote and albite+prehnite. The clinopyroxenes similarly yield information regarding the cooling history of the rocks over an extensive temperature range (Elsdon, 1971b).

General petrography. All the Upper Layered Series cumulates contain cumulus magnetite, which forms subhedral to euhedral crystals up to 1 mm in size, with cumulus plagioclase, augite, and occasional olivine. Typical textures are shown in fig. 1. With increasing height in the stratigraphic succession the modal proportion of Fe–Ti oxides increases, partly because of more extensive cumulus crystallization but mainly through an increasing quantity of intercumulus material, which often forms extensive clots up to 5 mm in size, enclosing silicate phases (fig. 1A). Near the top of the succession the cryptic layering is obscured by irregular variations in the iron ratios

$((\text{FeO} + \text{Fe}_2\text{O}_3)/(\text{FeO} + \text{Fe}_2\text{O}_3 + \text{MgO}))$ of the bulk rock analyses. These may, however, be correlated directly with the ratios of the modal Fe-Ti oxides to the modal Fe-Mg silicates (fig. 2).

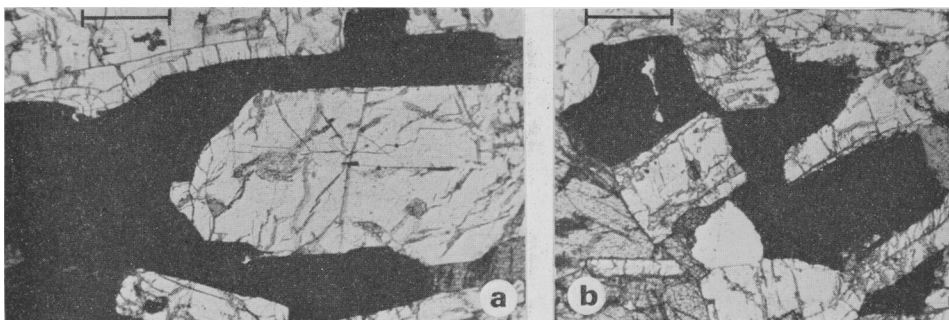


FIG. 1. Transmitted-light photomicrographs illustrating Fe-Ti oxide textures. Horizontal bar corresponds to 0.5 mm. Specimen EG7127, southern end of Alex' Mountain, 6750 metres. *a*: intercumulate Fe-Ti oxides along plagioclase-plagioclase grain boundaries. *b*: cumulate Fe-Ti oxides showing secondary enlargement.

Reflected-light studies. Various terms have been used to describe the compositional variations in cubic and rhombohedral Fe-Ti oxides (Buddington, Fahey, and Vlisidis, 1963; Anderson, 1968a). In the Upper Layered Series, however, the range of solid solution in each is limited, and the terms magnetite and ilmenite are adequate for the scope of this report.

The cumulus Fe-Ti oxide in the Upper Layered Series is magnetite, containing lamellae of ilmenite (fig. 3a). The intercumulus generation is mostly magnetite, accompanied by ilmenite, which has three modes of occurrence—as lamellae in magnetite, as granular intergrowths with magnetite (fig. 3b), and as occasional discrete grains, which may contain lamellae of magnetite.

As with the silicates (Elsdon, in preparation), the oxide minerals show appreciable late-stage alteration in some cases. There are two different modes of alteration, which are not often found in the same specimen. The first is resorption of magnetite; in transmitted light

it is seen that the alteration products are chlorite intergrown with an extremely fine-grained dark brown material (fig. 4). The second is alteration of ilmenite to rutile; this is seen within discrete ilmenite grains, at the edges of silicate inclusions within

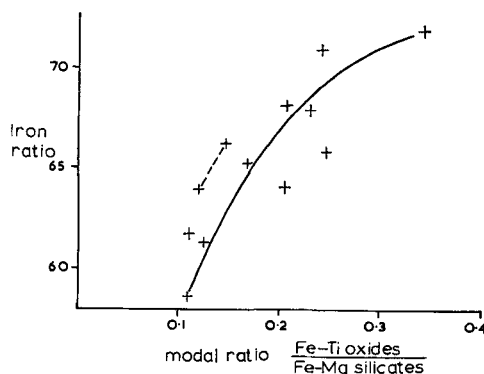


FIG. 2. Relationship between iron ratio of Upper Layered Series cumulates and the modal ratio Fe-Ti oxides : Fe-Mg silicates. Dashed line connects crosses representing adjacent layers rich in felsic and mafic constituents respectively. (From Elsdon, unpublished Ph.D. thesis.)

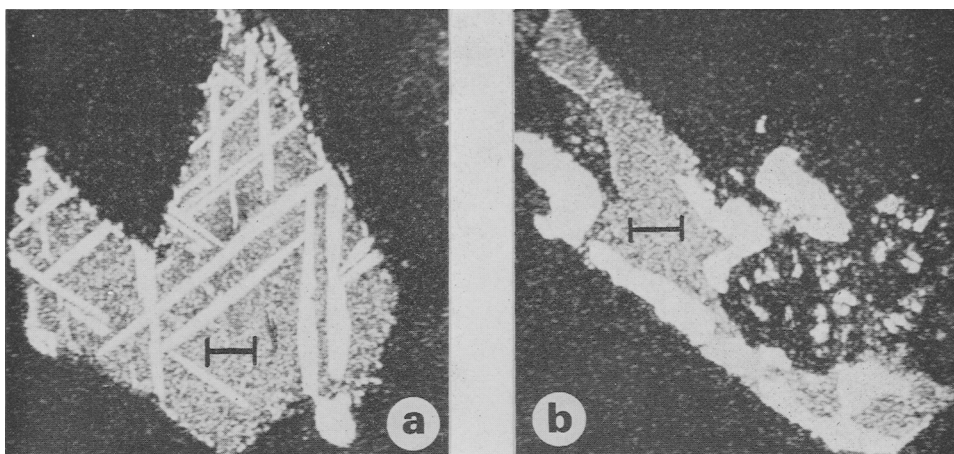


FIG. 3. Electron-probe scanning pictures of Fe-Ti oxides from EG7127, using Ti-K radiation. White areas are ilmenite, grey areas are magnetite, dark areas are silicates. *a*: magnetite containing ilmenite lamellae. *b*: granular intergrowth. Length of horizontal bar corresponds to 0.1 mm.

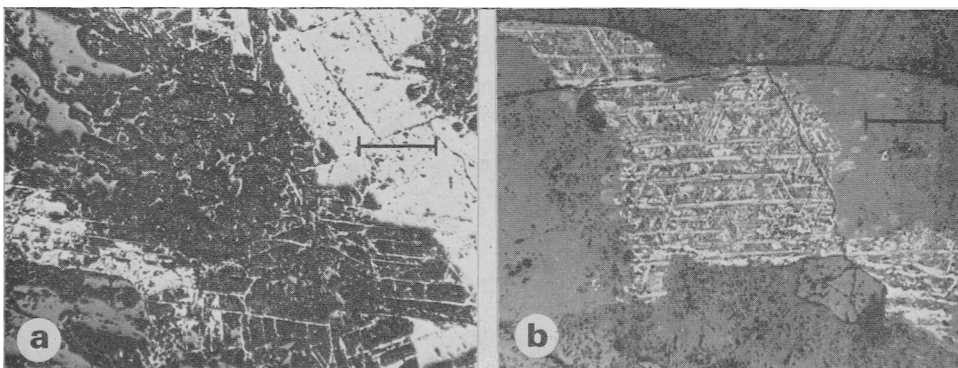


FIG. 4. Resorption of magnetite in lamellar intergrowths with ilmenite. Length of horizontal bar corresponds to 0.2 mm. Specimen EG7031, bottom of Junction Nunatak, 2950 m. *a*: transmitted light; *b*: reflected light.

ilmenite, and at magnetite-ilmenite boundaries. The significance of alteration will be considered in a later section.

Chemistry. The standard methods of analysing Fe-Ti oxide assemblages involve either separation followed by wet-chemical analysis (Vincent and Phillips, 1954), or direct electron micro-probe analysis (Carmichael, 1967). Both techniques were used in this study.

The oxides were separated from the silicates by centrifuging rock powder (-80 mesh) in Clerici solution. In most cases only two treatments were necessary to effect an almost pure yield of oxides, which were then washed with hot water and dried at

105 °C. A known quantity of oxides was fused with potassium pyrosulphate and this was followed by digestion in dilute sulphuric acid. Part of the solution was reduced and titrated against ceric sulphate to determine total Fe as Fe_2O_3 ; another part was treated with dilute sulphuric acid and hydrogen peroxide, and the TiO_2 -content determined by colorimetry. A second known weight of oxides was treated with concentrated sulphuric acid and hydrofluoric acid, and titrated against potassium permanganate solution to determine FeO. Fe_2O_3 was then calculated by subtraction.

The magnetite and ilmenite in intergrowths were also analysed separately using a Geoscan electron-probe microanalyser. Pure metal standards were used for Fe, V, Ti, Mn, Cr, and Ni; wollastonite was used for Ca, periclase for Mg, quartz for Si and corundum for Al. Operating conditions were: voltage 20 kV, specimen current 5×10^{-8} amps, beam diameter 1 micron. Corrections to the raw data for instrument dead-time, absorption, and secondary fluorescence were applied using a computer programme devised by Dr. J. V. P. Long. Total iron was then recalculated as Fe^{2+} and Fe^{3+} using the procedure outlined by Anderson (1968b).

Some of the arguments that follow, concerning chemical changes associated with dissolution of magnetite, assume that the composition of discrete ilmenite is the same as that in the magnetite-ilmenite intergrowths. There are no precise data available, but rough electron-probe measurements show them to be the same to within 2 % relative. Investigations by Anderson (1968b) show that, in the La Blache Lake titaniferous magnetite deposit, the two generations of ilmenite have the same composition and the same oxygen isotope ratio, indicating that equilibrium was maintained during cooling from magmatic temperatures. It is possible that, as a general rule, two generations of ilmenite in a plutonic rock will have compositions that are not greatly different.

Electron-probe analyses of magnetite and ilmenite from seven cumulates and one gabbro pegmatite are presented in Table I. Measurements were obtained in most specimens from a granular intergrowth since the lamellar intergrowths are more susceptible to late-stage alteration. Each pair of analyses in Table I represents one intergrowth rather than an average of several, because measurements of Fe and Ti on four grains from each rock revealed inter-grain homogeneity to within 2 % relative, with respect to Fe and Ti. Individual grains were similarly found to be homogeneous, except near magnetite-ilmenite boundaries. Ilmenite lamellae, when selected for analysis, were broad and free from inclusions and alteration; spots analysed were at the centres of lamellae. Lamellar intergrowths were analysed in those specimens where granular intergrowths were absent, EG7015 and 7083.

Equilibration conditions. The percentages of ulvöspinel in magnetite and hematite in ilmenite were calculated using the method of Anderson (1968b), and $T-f_{\text{O}_2}$ equilibration conditions calculated in each case according to Buddington and Lindsley (1964); data are presented graphically in fig. 5, along with data from the Skaergaard intrusion (Buddington and Lindsley, 1964).

In cases where magnetite-ilmenite intergrowths are not accompanied by discrete ilmenite, equilibration conditions deduced give the *maximum* f_{O_2} at any given temperature (see Nicholls, 1971, p. 103). The Upper Layered Series oxides equilibrated under

TABLE I. *Electron-probe analyses of coexisting magnetites and ilmenites*

	1		2		3		4		5		6		7		8	
	mt	il	mt	il	mt	il	mt	il	mt	il	mt	il	mt	il	mt	il
FeO	31.7	44.4	33.7	36.9	35.0	35.3	34.9	41.0	30.8	37.9	36.0	42.5	32.4	42.2	35.2	37.8
Fe ₂ O ₃	59.6	6.7	51.1	11.5	53.8	14.7	55.6	8.3	61.7	13.1	56.8	8.1	59.1	8.8	53.3	13.3
TiO ₂	2.7	47.8	8.6	46.0	6.4	44.2	5.6	47.7	3.5	43.8	5.8	49.0	4.6	48.4	6.4	44.4
MnO	0.1	1.4	0.4	1.1	0.3	2.5	0.1	1.8	0.2	1.4	0.1	1.5	0.1	1.4	0.3	1.9
V ₂ O ₅	1.3	n.f.	0.5	0.9	1.0	0.1	0.7	n.f.	1.1	2.3	0.9	0.1	1.5	0.4	1.4	2.9
Cr ₂ O ₃	0.4	0.1	n.f.	n.f.	0.3	0.2	n.f.	n.f.	0.4	0.1	0.8	0.1	0.5	0.1	1.0	0.1
NiO	0.1	n.f.	0.1	0.1	n.f.	0.1	0.1	n.f.	n.f.	0.1	n.f.	n.f.	n.f.	n.f.	n.f.	n.f.
CaO	0.3	0.1	0.2	0.4	0.1	0.1	0.2	0.5	0.4	0.1	0.2	0.1	0.3	0.4	0.1	0.1
Al ₂ O ₃	1.1	0.1	0.8	0.6	1.1	1.7	1.3	n.f.	1.3	0.9	n.f.	n.f.	0.1	n.f.	3.0	0.3
SiO ₂	0.8	0.1	0.8	0.1	0.7	0.5	0.3	1.3	0.6	0.5	0.2	n.f.	n.f.	n.f.	0.1	0.1
MgO	n.f.	n.f.	2.5	2.0	0.5	1.1	n.f.	0.1	n.f.	0.1	n.f.	n.f.	n.f.	n.f.	n.f.	n.f.
Total	98.1	100.7	98.7	99.6	99.2	100.5	98.8	100.7	100.0	100.3	100.8	101.4	98.6	101.7	100.8	100.9
% ulv	8.4	—	18.8	—	18.1	—	10.0	—	10.0	—	17.1	—	13.4	—	18.0	—
% hm	—	6.8	—	12.3	—	15.8	—	8.3	—	13.5	—	8.0	—	8.6	—	13.5
T °C	580	700	700	720	720	600	600	610	610	660	660	620	620	680	680	680
log f _{O₂}	-20.5	—	-16.5	—	-15.0	—	-19.2	—	-17.8	—	-18.0	—	-19.0	—	-16.0	—

1. EG7015 plagioclase-augite-olivine-magnetite orthocumulate, bottom of Birthday Nunatak, 5100 m.
2. EG7030 plagioclase-augite-olivine-magnetite orthocumulate, bottom of Junction Nunatak, 2950 m.
3. EG7031 gabbro pegmatite, same locality.
4. EG7068 plagioclase-augite-magnetite orthocumulate, northern end of Alex' Mountain, 6950 m.
5. EG7083 plagioclase-augite-magnetite orthocumulate, southern end of Alex' Mountain, 6650 m.
6. EG7119 plagioclase-augite-magnetite orthocumulate, southern end of Alex' Mountain, 6700 m.
7. EG7127 plagioclase-augite-magnetite orthocumulate, southern end of Alex' Mountain, 6750 m.
8. EG7062 plagioclase-augite-magnetite orthocumulate, top of Junction Nunatak, 3150 m.

oxygen fugacities an order of magnitude or so higher and temperatures 200 to 300 °C lower than the Skaergaard oxides. These results are compatible with the hypothesis of a high water content in the Upper Layered Series magma, capable of exerting a high f_{O_2} and promoting equilibration at lower temperatures than in a relatively anhydrous environment.

Higher levels in the intrusion equilibrated at lower temperatures than deeper levels (fig. 6) although, for reasons that are not understood, the lowest equilibration temperature is obtained from EG7015, which is only halfway up the exposed succession. Ignoring the presence of water in the cooling body, the general trend is difficult to explain; it is, however, envisaged that water diffused upwards in response to a vertical temperature gradient. The effect of water on equilibration temperatures has already been proposed in comparing T - f_{O_2} conditions for the Upper Layered Series and the Skaergaard intrusion. Further, the apparently high equilibration temperature for the oxides of the gabbro pegmatite EG7031 (Table I) appears reasonable bearing in mind that it comes from the lowest exposed stratigraphic level and that this temperature is only slightly higher than that of its host cumulate EG7030.

Minor elements. Manganese distribution may be a function of equilibration temperature (Buddington and Lindsley, 1964). Plotted on a graph of % MnO in ilmenite against % MnO in magnetite, the Upper Layered Series Fe-Ti oxides lie within the field of acid plutonics (fig. 7; compare Buddington and Lindsley, 1964, p. 353). This is because the temperatures of equilibration (Table I) are near to liquidus temperatures for water-saturated granites.

The MnO-distribution may alternatively be related to fractional oxidation since, by analogy with the systems Fe-O (Hahn and Muan, 1960) and Mn-O (Darken and Gurry, 1946), Fe_2TiO_4 is oxidized at lower oxygen fugacities than Mn_2TiO_4 . Thus residual magnetite should be concentrated in Mn_2TiO_4 relative to Fe_2TiO_4 with increasing oxygen fugacity or increasing temperature. Fig. 8 shows that there may be slight fractionation with respect to temperature, but the trend is not clear.

Examination of Table I shows little evidence of regular variation of minor elements (V, Cr, Ni, Ca, Al, Si, Mg) with respect to stratigraphic height. MgO is present in appreciable quantities in both phases at 2950 m. but thereafter falls off rapidly to 0.1 % and less. The MgO-contents, where measurable, were taken into account when calculating the composition of magnetite and ilmenite from electron-probe

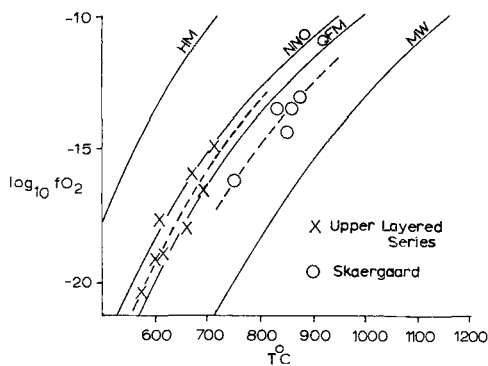
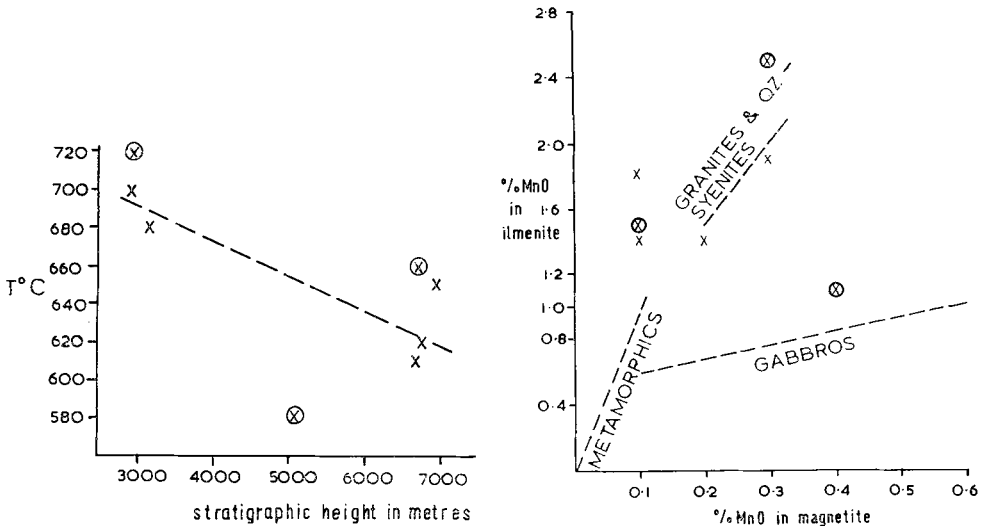


FIG. 5. T - f_{O_2} conditions of equilibration of Upper Layered Series Fe-Ti oxides, compared with data for the Skaergaard intrusion (Buddington and Lindsley, 1964). Full curves refer to buffer assemblages; NNO, nickel-nickel oxide; QFM, quartz-fayalite-magnetite; MW, magnetite-wüstite; HM, hematite-magnetite. Ordinates are $\log f_{O_2}$ bars.



FIGS. 6 and 7: FIG. 6 (left). The relationship between temperature of final equilibration of Fe-Ti oxides and stratigraphic height. Altered specimens are shown as crosses-within-circles. FIG. 7 (right). Plots of %MnO in magnetite. Altered specimens are shown as crosses-within-circles. Dashed curves indicate trends for metamorphic assemblages, mafic igneous assemblages, and salic igneous assemblages (Buddington and Lindsley, 1964).

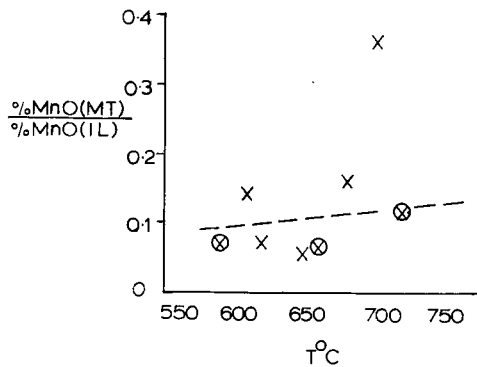


FIG. 8. The relationship between observed partition coefficients of MnO in coexisting Fe-Ti oxides and their temperature of final equilibration. Altered specimens are shown by crosses-within-circles.

data (Anderson, 1968b; Speidel, 1970). In some cases the minor element concentrations were below detection limits for short counting times (Table I).

Alteration of magnetite. The analyses of coexisting magnetite and ilmenite have been plotted, with the bulk Fe-Ti oxide compositions (Table II), in terms of FeO, Fe₂O₃, and TiO₂ (fig. 9). Because electron-probe analyses were made on unaltered

TABLE II. Bulk chemical analyses of magnetite and ilmenite; electron-probe analyses recast to 100 %

Bulk analyses	Electron-probe analyses		
	FeO	Fe ₂ O ₃	TiO ₂
1.	31.4	41.4	27.2
2.	35.7	42.8	21.5
3.	28.8	30.9	40.3
4.	35.7	42.0	22.3
5.	33.7	38.8	27.5
6.	34.4	27.2	38.4
7.	36.0	40.3	23.7
8.	n.d.	n.d.	n.d.
9.	38.2	34.8	27.0
10.	41.3	30.5	28.2
11.	36.9	36.4	26.7

1-8 as in Table I.

9. EG7071 plagioclase-augite-magnetite orthocumulate, top of Birthday Nunatak, 5250 m.

10. EG7086 gabbro pegmatite

11. EG7131 plagioclase-augite-magnetite orthocumulate. Both 10 and 11 from southern end of Alex' Mountain, 6650 m.

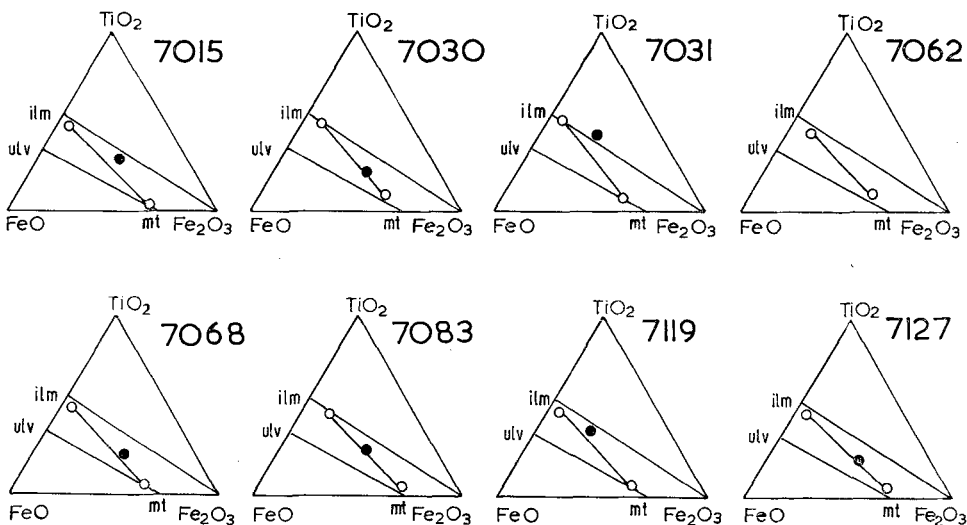


FIG. 9. FeO-Fe₂O₃-TiO₃ plots for seven Upper Layered Series cumulates and one pegmatite, showing compositions of magnetite and ilmenite as open circles and bulk Fe-Ti oxide compositions as closed circles. Magnetite(mt)-ulvöspinel(ulv) and ilmenite(il)-hematite(hm) tie lines are shown.

grains and because microscopic examination revealed substantial alteration in some cases, the bulk analyses of oxides must lie on the FeO-poor side of the magnetite-ilmenite tie-lines in the FeO-Fe₂O₃-TiO₂ system (fig. 9). This could be due either to oxidation of ferrous iron *in situ*, or to dissolution of magnetite. In fig. 10 the bulk analyses are plotted together. The three most altered specimens plot away from the cluster formed by the less altered ones, and two of them (EG7031, 7119) plot towards the TiO₂ apex. This is compatible with dissolution of magnetite; Davidson and Wyllie (1968) have also described this phenomenon, based on petrographic observations of diabase-granophyre associations from Pennsylvania.

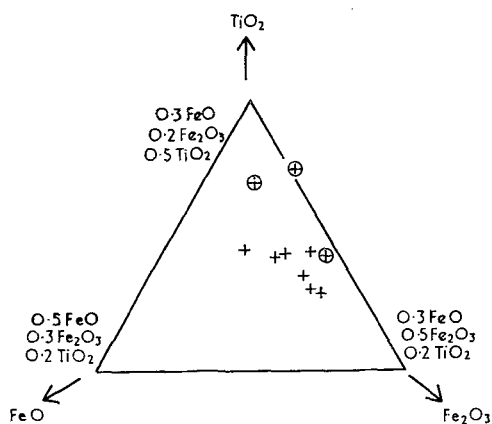


FIG. 10. Bulk compositions of Fe-Ti oxides from ten rocks plotted according to triangular co-ordinates within the system FeO-Fe₂O₃-TiO₂. The three altered specimens are shown as crosses-within-circles.

There is experimental evidence for the fractionation of iron into the vapour phase at subsolidus temperatures during cooling of magma (Martin and Piwinski, 1969). In the first quantitative study of magnetite solubility in aqueous solutions (Sweeton and Baes, 1970) it has been shown that the solubility varies little with temperature in the range 50 to 300 °C, but that it depends markedly on hydrogen-ion concentration. Under these conditions

the iron is removed as the soluble species Fe²⁺, FeOH⁺, Fe(OH)₂, and Fe(OH)₃⁻. No quantitative data are yet available on the solubility of ilmenite, or for magnetite above 300 °C.

Water content of the magma. The fresh nature of the contact rocks of the Upper Layered Series (Elsdon, 1969) indicates that the crystallization and cooling history was one in which the system was closed with respect to water. This means that water was not introduced from the country rocks and that the water content of the magma was retained. It is therefore possible to consider the water content of the magma in a quantitative manner.

Taylor (1964) showed that up to 0.5 % excess oxygen can be accommodated in titaniferous magnetite at 1300 °C; this value is 0.3 % at 1200 °C (Webster and Bright, 1961). Thus the amount of R₂O₃ that can be exsolved from titaniferous magnetite as ilmenite-hematite solid solution due to excess oxygen is up to 15 %; in view of experimentally determined basalt liquidus temperatures the maximum is probably closer to 10 %. On this basis Buddington and Lindsley (1964) argued that most of the ilmenite that is intergrown with magnetite in natural assemblages is formed by oxidation of ulvöspinel.

The amount of water necessary to produce an observed amount of ulvöspinel

oxidation is given by the equation (Anderson, 1968b) based on the reaction $3\text{Fe}_2\text{TiO}_4 + \text{H}_2\text{O} \rightarrow 3\text{FeTiO}_3 + \text{Fe}_3\text{O}_4 + \text{H}_2$:

$$B/A = (f_{\text{O}_2}^{\frac{1}{2}} + K_2\gamma_2)/3\{(1 - 1/X_1)f_{\text{O}_2}^{\frac{1}{2}} + K_2\gamma_2\},$$

where A = number of moles of ulvöspinel oxidized, B = number of moles of water initially present in the system, and X_1 = mole fraction of water present in the aqueous phase at the higher temperature T_1 . At the lower temperature T_2 , f_{O_2} = oxygen fugacity, K_2 = decomposition constant for water, and γ_2 = fugacity coefficient for water.

This calculation requires that the initial temperature T_1 at the beginning of oxidation and the final temperature of equilibration T_2 should be known. Using temperatures $T_1 = 1100^\circ\text{C}$ (see Yoder and Tilley, 1962, Tables 28–31) and $T_2 = 600^\circ\text{C}$ and appropriate data the initial water content of the magma is estimated to be approximately 7%. This is the maximum that can be dissolved in basaltic magma at a total pressure of 5 kb (Hamilton, Burnham, and Osborn, 1964).

Evidence from other sources is conflicting. The occurrence of small amounts of primary pargasite throughout suggests that the magma crystallized under conditions of $P_{\text{H}_2\text{O}} = P_{\text{total}} = 2\text{ kb}$ (Yoder and Tilley, 1962) and a water content of 4% (Hamilton Burnham, and Osborn, 1964). Application of observations to conditions where $P_{\text{H}_2\text{O}}$ is less than P_{total} is difficult because while Yoder and Tilley (1962) postulate diminishing stability of amphibole under these conditions, Nesbitt and Hamilton (1970) have presented experimental evidence to the contrary. Thus it is not yet possible to test whether the magma could have been intruded under conditions where *partial* pressure of water was 5 kb.

An alternative hypothesis is that, during cooling, hydrogen was able to diffuse outwards through the walls of the intrusion by virtue of its small atomic size and fast diffusion rate. In this case the oxygen fugacity is no longer related in a simple manner to the water content. Such a hypothesis has recently found favour with other workers (e.g. Gibb and Henderson, 1971).

Summary of cooling history. This study of the Upper Layered Series Fe–Ti oxides has yielded some information regarding the cooling history of the rocks:

Textural examination in transmitted light shows that magnetite began to crystallize as a cumulus phase at an early stage in the fractionation sequence. The magma therefore had a high $\text{Fe}^{3+}:\text{Fe}^{2+}$ ratio, consequent upon a high water content, as verified by the early crystallization of pargasite and the widespread hydrothermal alteration by water derived from the magma itself.

Concentration of Fe^{3+} and Ti in the residual magma resulted in the extensive crystallization of intercumulus magnetite and ilmenite.

Electron-probe determinations of coexisting phase compositions shows that equilibration occurred at temperatures 200 to 300 °C below those for the Skaergaard intrusion and at oxygen fugacities an order of magnitude or so higher at any given temperature.

The oxygen fugacity at lower temperatures was sufficient to cause breakdown of ilmenite to rutile on a small scale: $2\text{FeTiO}_3 + \text{O} \rightarrow \text{Fe}_2\text{O}_3 + 2\text{TiO}_2$. This reaction occurs

in the temperature range 200 to 650 °C (Karkhanavala and Momin, 1959), but there are no data available concerning the oxygen fugacities involved.

Comparisons of compositions of coexisting phases with the bulk Fe-Ti oxide compositions indicate removal of Fe₂O₃ from the system in some cases, especially where there is appreciable hydrothermal alteration (Elsdon, in preparation).

Higher levels of the intrusion equilibrated at lower temperatures than did lower levels, due to upward diffusion of water through the cooling body in response to a vertical temperature gradient.

Acknowledgements. I thank Professors W. A. Deer and G. M. Brown, and Drs. I. D. Muir, S. R. Nockolds, J. B. Wright, and R. W. D. Elwell for their kind criticisms of various drafts of this report. I am grateful to Dr. A. T. Anderson, Jr. for helpful discussion on the use of the formula quoted from his paper (1968b). Mr. C. McDonnell prepared the diagrams. The work was carried out at the Department of Mineralogy and Petrology, University of Cambridge, during the tenure of a N.E.R.C. Research Studentship.

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[Manuscript received 21 October 1971]