Magnesian ilmenite in picritic basalts from the Karoo Province, South Africa

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Abstract

Some picritic lavas from near the base of the volcanic succession in the Lebombo region of the Karoo Province contain phenocrysts of olivine and skeletal microphenocrysts of ilmenite, the latter indicating rapid crystallization from the magma. These ilmenite grains contain up to 6% MgO. The near-liquidus crystallization and the high MgO content of the ilmenite are highly anomalous for ilmenite in tholeiitic rocks. These features reflect the atypical nature (high MgO and TiO_2) of the parental magma and/or highly reduced conditions of crystallization.

KEYWORDS: magnesian ilmenite, ilmenite, basalts, Karoo Province, South Africa.

Introduction

ILMENITE is a widely distributed but minor mineral in many igneous rocks. Its composition varies according to rock type and is often diagnostic of magma composition. In all alkaline rocks it tends to show variable solid solution towards $MgTiO_3$—the geikielite molecule, reaching a maximum of 20% MgO in groundmass ilmenite in kimberlite (Shee, 1984; Haggerty and Tompkins, 1984). In such rocks it also contains up to 1% Cr_2O_3. The Ti-rich lunar mare basalts also contain ilmenite with up to 9% MgO depending on the timing and conditions of its formation (Frondel, 1975). In acid rocks, solid solution is usually towards $MnTiO_3$—the pyrophanite molecule (Lipman, 1971). In tholeiitic rocks, ilmenite contains less than 3% MgO and little MnO or Cr_2O_3 (Haggerty, 1976), and textural evidence (Reynolds, 1983) and experimental studies (Thompson, 1976) show that it is normally one of the last minerals to crystallize.

An atypical occurrence of MgO-rich ilmenite has been reported by Cawthorn *et al.* (1985, 1988) and Groves *et al.* (1986) from the Mount Ayliff Intrusion in the Transkei of southern Africa. This is one of many tholeiitic intrusions of Karoo age in southern Africa, but is thicker than most and shows evidence of fractional crystallization. The lower olivine-rich portion of this body contains ilmenite grains with up to 11% MgO and 1.3% Cr_2O_3. Those authors attributed this to crystallization from an unusual magnesian magma, whereas Lightfoot *et al.* (1987) suggested subsolidus reequilibration with olivine and chromite as the cause.

Here we report on the occurrence of high-MgO ilmenite in picritic basalts from the Karoo Province which allows discussion of ilmenite formation under conditions where slow cooling and reequilibration can be discounted.

Karoo Igneous Province of southern Africa

The Karoo Igneous Province of southern Africa represents an extensive extrusive and intrusive suite of rocks ranging in age from 200 to 130 Ma (Bristow and Saggerson, 1983; Cox, 1983). Extrusive rocks are best preserved in the Lebombo, Nuanetsi, and Sabi areas of southeast Africa, in

Mineralogical Magazine, April 1989, Vol. 53, pp. 245–52

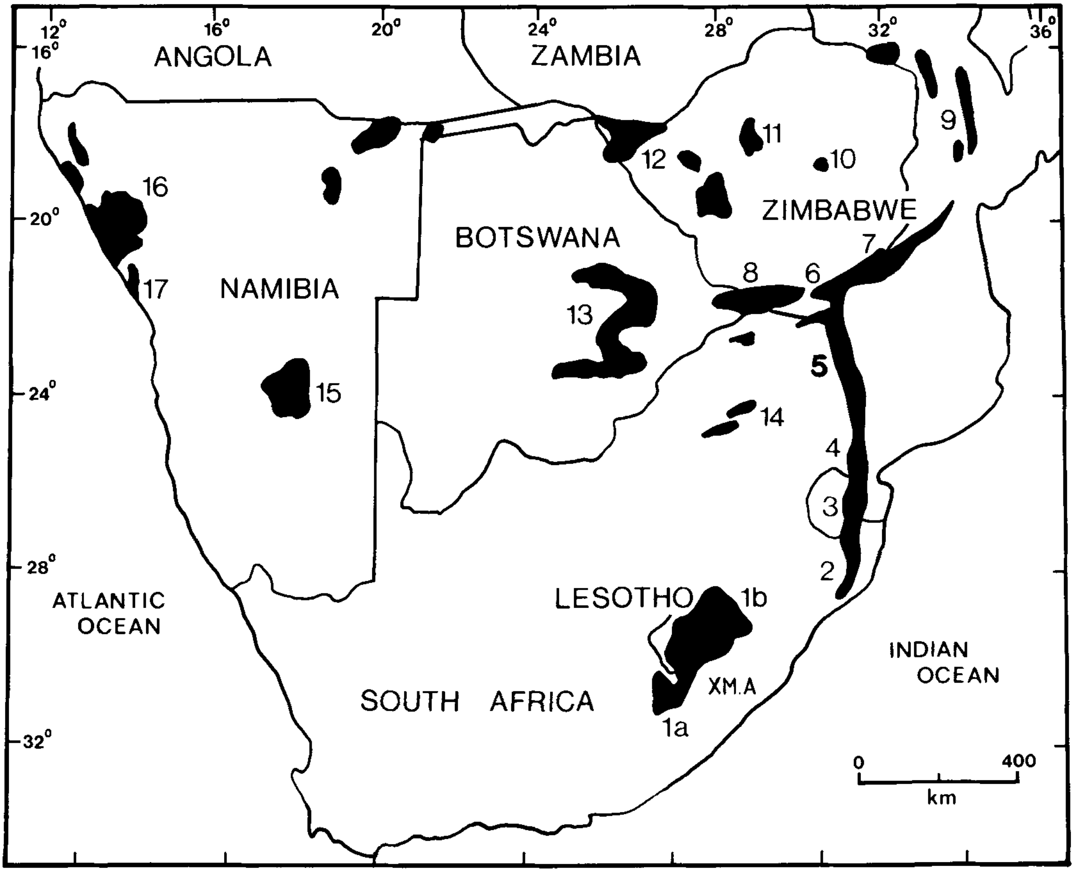


FIG. 1. Distribution of Karoo volcanics in southern Africa, after Eales *et al.* (1984). 1a. Stormberg; 1b. Lesotho; 2. Southern Lebombo; 3. Swaziland; 4. Central Lebombo; 5. Northern Lebombo (from where the magnesian ilmenite of this study came); 6. Nuanetsi; 7. Sabi; 8. Tuli; 9. Lupata; 10. Featherstone; 11. Nyamandhlovu; 12. Victoria Falls–Wankie; 13. Botswana; 14. Springbok Flats; 15. Mariental; 16. Kaokoveld; 17. Cape Cross. Locality of the Mount Ayliff Intrusion is indicated as M.A.

the central Karoo of South Africa and Lesotho, and in the Kaokoveld of South West Africa (Namibia), Eales *et al.*, 1984. The present outcrop of volcanic rocks is shown in Fig. 1. Considerable lateral and vertical variation occurs within these rocks. The complete succession in the Lebombo sector consists of nephelinite, voluminous picritic basalt, low-MgO tholeiite, rhyolite and alkali basalt (Bristow and Cleverly, 1983).

Cox (1983) and Duncan *et al.* (1984) have shown that there are major geochemical changes in magma composition from south to north, with a preponderance of basalt and high-MgO lavas characterized by high incompatible element abundances in the north which contrast markedly with the more typical tholeiitic rocks in the south.

Of interest in this study are the picritic basalts of the Lebombo region straddling the South Africa–Swaziland–Mozambique borders. The geochemistry of these lavas has been described by Bristow (1984), and only the occurrence and composition of the ilmenite are discussed in detail here.

For comparison, the occurrence of ilmenite in the dolerites and other intrusions in the Karoo Province are briefly reviewed. These rocks have been the subject of many studies including the classic monograph of Walker and Polldervaart (1949). Most geochemical models emphasize their similarity and relationship to typical low-MgO tholeiite magma (Eales and Marsh, 1979; Lightfoot and Naldrett, 1984), although anomalous types exist (Le Roex and Reid, 1978). Due to the presence of magmatic nickel–copper–

platinum sulphide deposits in the Insizwa lobe of the Mount Ayliff Intrusion this specific intrusion has been studied in detail (Scholz, 1936; Dowset and Reid, 1967; Tischler *et al.*, 1981; Lightfoot *et al.*, 1984, 1987; Cawthorn *et al.*, 1985, 1988; Groves *et al.*, 1986). Cawthorn *et al.* (1985) suggested, on the evidence of sulphide compositions and the presence of ilmenite (which contains up to 11% MgO and 1.3% Cr_2O_3), that the lower portion of this body crystallized from a magnesian magma, while Lightfoot *et al.* (1987) have argued that these rocks could have formed from a low-MgO magma. They suggested that the atypical ilmenite compositions owe their high MgO and Cr_2O_3 contents to reaction with olivine and chromite, even though the experimental data of Bishop (1980) indicate that the partitioning of Mg and Fe between ilmenite and olivine at low temperature is exactly the opposite from that required by their model.

Reynolds (1983) has presented a compilation of ilmenite compositions from the dolerites in southern Africa, and in none of these does the MgO content exceed 3%, and hence those from the Mount Ayliff Intrusion are highly anomalous in a regional context.

Petrography

The Lebombo picritic basalts described by Bristow (1984) are similar to those from the Nuanetsi province of Zimbabwe (Cox and Jamieson, 1974). They are characterized by an abundance of olivine phenocrysts, which sometimes display glomeroporphyritic textures. Rounded or resorbed and perfectly euhedral grains may occur together (Fig. 2*a*), suggestive of different generations, the former possibly being a high pressure macrocryst. Skeletal forms, with negative crystal faces or hopper forms (Fig. 2*b*) are similar to those described from rapidly cooled, MgO-rich liquids (Drever and Johnston, 1957). Olivine ranges from 6.5 to 46.1 modal per cent. The other phenocryst phases, plagioclase, clinopyroxene, orthopyroxene and ilmenite, are equally variable in abundances.

The groundmass textures of the picritic basalts vary from glassy through microcrystalline to examples which are fine to medium grained. The glassy lavas are characterized by the presence of numerous skeletal crystals of Fe–Ti oxides, abundant microlites or skeletal clinopyroxene, and apatite needles. In rocks with a microcrystalline groundmass, plagioclase appears as sheaves of quenched microlites accompanying acicular to equant euhedral grains of clinopyroxene. Coarser-grained lavas are characterized by well-formed laths of plagioclase, euhedral and rounded grains of clinopyroxene, spicular and lath-shaped Fe–Ti oxides, and apatite.

The ubiquitous Fe–Ti oxides appear as fine needles and fragile dendritic growths (Fig. 2*c*). In the microcrystalline rocks the oxides appear as skeletal needles or blades which may reach several millimetres long, and rectangular laths with ragged, swallow-tail terminations. Parallel growth of skeletal blades is common, and less commonly hopper textures are developed (Fig. 2*c* and *d*). These textures are very similar to those produced experimentally during controlled cooling and crystallization of lunar basaltic material (Usselman and Lofgren, 1976; their figures 3A and 3C).

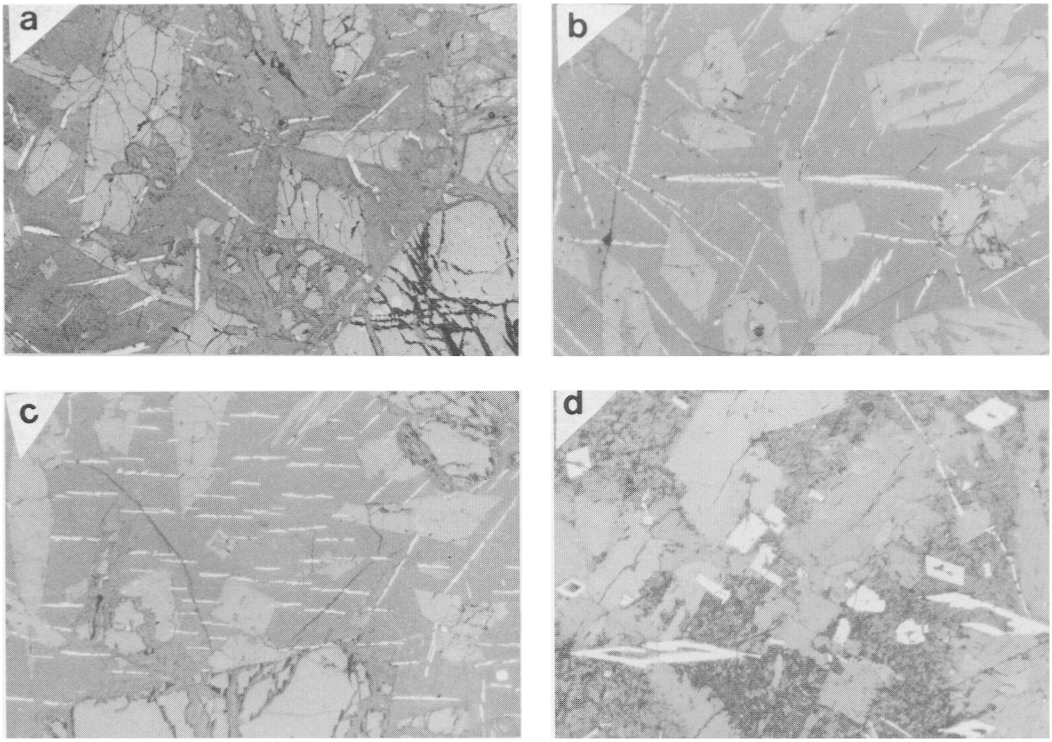
The olivine-poor basalts and dolerites of Swaziland and southern Lebombo contain very different oxide mineralogy and texture from the picritic basalt suite. The oxide minerals are generally confined to the groundmass, although infrequent phenocrysts have been observed. They always have a crude octahedral shape and are titano-magnetites. Ilmenite is only found in the andesitic intrusive bodies in the northern Lebombo, where it occurs as phenocrysts and microphenocrysts and as glomeroporphyritic aggregates with plagioclase and clinopyroxene.

The oxide minerals in the picritic basalt are generally opaque, though some of the blades and needles are dark orange-brown and translucent. Similar oxide minerals have been described in the Nuanetsi lavas, and were originally identified as kennedyite (Von Knorring and Cox, 1961), but are now defined as belonging to the pseudobrookite–armalcolite series (Bowles, in press). The orthorhombic outline of the crystals shown in Fig. 2*c* is typical of ilmenite forming a pseudomorph after armalcolite solid solution due to reaction with magma, as demonstrated by Haggerty (1973).

These textures in the picritic lavas suggest that ilmenite is an early-formed phase in the crystallization sequence. Olivine is indisputably the liquidus mineral, but the presence of ilmenite grains partially embedded in olivine and its presence as the only other mineral phase in a glassy matrix in some samples suggest that it is the second phase to crystallize.

Ilmenite compositions

The compositions of representative ilmenite grains analysed from the picritic basalts are shown in Table 1. These data are plotted in Fig. 3, showing the ratio of ilmenite–geikielite–pyrophanite, together with the generalized differentiation trend proposed by Reynolds (1983) for ilmenite in tho-



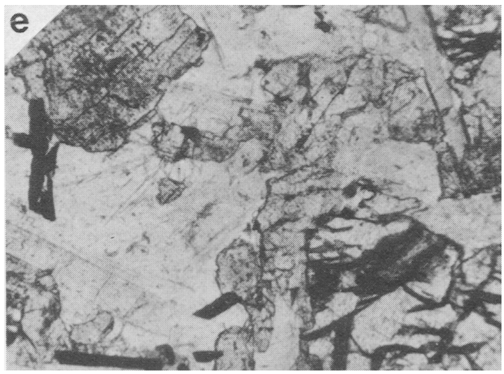


FIG. 2. Photomicrographs of ilmenite textures. (*a*) Large phenocrysts of olivine and blades of ilmenite (bright white) set in a glassy matrix. Note the rounded, fractured appearance of the olivine grain, lower right, as distinct from the angular terminations of the other crystals, suggesting the former may be a higher pressure macrocryst. Ilmenite blades in lower left display a swallow-tail termination. Note how two ilmenite grains penetrate the margin of the tabular olivine crystal in centre-right. Reflected light photograph; field of view, 3 × 2 mm, picritic basalt, Lebombo KP112. (*b*) Hopper olivine crystals (especially upper and lower right) and skeletal-to-chain ilmenite grains in a glassy groundmass. Reflected light; field of view, 3 × 2 mm, picritic basalt KP108. (*c*) Parallel growth of ilmenite blades. Note the different olivine morphologies, macrocysts (bottom centre), phenocrysts and hopper crystals (centre and top centre-right). Reflected light, field of view, 3 × 2 mm, picritic basalt KP108. (*d*) Hopper ilmenite crystals, olivine phenocrysts and microcrystalline matrix. Reflected light, field of view, 3 × 2 mm, picritic basalt KA29. (*e*) Resorbed anhedral olivine crystal (lower right) and granular intergrowth of orthopyroxene and plagioclase grains. Euhedral, prismatic crystals of ilmenite (black) are embedded in the pyroxene and plagioclase. Transmitted plane-polarized light, field of view, 7.5 × 5 mm, olivine gabbro, Mount Ayliff Intrusion.

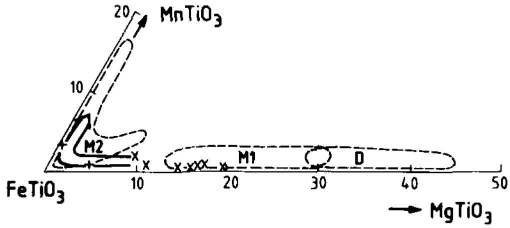


FIG. 3. Plot of weight proportions of components $FeTiO_3$–$MgTiO_3$–$MnTiO_3$ in ilmenite. Broad arrow shows general trend of differentiation of ilmenites from Karoo dolerites (Reynolds, 1983). Crosses represent Lebombo picrites and pluses low-MgO lavas. Fields M1 and M2 denote ranges in composition of the magnesian ilmenite found in picrite and the low-MgO ilmenite in felsic veins from Mount Ayliff (Cawthorn *et al.*, 1985; 1988; Groves *et al.*, 1986). Field D denotes ilmenite from kimberlite (Haggerty, 1976; Shee, 1984). Note how ilmenite from Mount Ayliff and Lebombo picrites are more MgO-rich than any found in other Karoo dolerites.

leiitic Karoo dolerites. The field occupied by the analyses from the Mount Ayliff Intrusion is included for comparison. Analyses from the picritic basalts and the Mount Ayliff Intrusion are far richer in MgO than any found in other Karoo intrusions.

The data are also plotted in Fig. 4, which shows the variation in ilmenite composition as a function of parent magma type as proposed by Haggerty (1976). Again, these analyses are atypical of ilmenite found in tholeiitic rocks, and have more similarities with kimberlitic rocks. However, a difference between these and kimberlitic ilmenite is apparent when the proportion of the hematite molecule is considered. This is shown in Fig. 5, and shows that the ilmenites from the picritic basalts and the Mount Ayliff Intrusion tend to have variable but lower hematite components than those from kimberlite. The Fe_2O_3 content is calculated indirectly on the assumption of perfect stoichiometry. As this joint project has involved the use of three different electron microprobes (Bristow, 1984; Cawthorn *et al.*, 1985), we believe that these calculated hematite values are real, and not due to systematic analytical error. Some of the ilmenite analyses from the picritic basalts also contain significantly higher TiO_2 contents than kimberlite ilmenites.

Discussion

High-MgO ilmenite is atypical of rocks crystallized from tholeiitic magmas (Haggerty, 1976). In the case of the ilmenite from Mount Ayliff, the debate about whether it results from subsolidus reequilibration or is a primary composition is difficult to resolve. In the case of the picritic basalts from the Lebombo, however, ilmenite is embedded in a glassy matrix. Thus, there could have been no appreciable length of time for reequilibration to occur. The MgO content of 6% in the ilmenite cannot therefore be attributed to such a process. Other possibilities are discussed below.

It has been suggested that ilmenite forming under high-pressure conditions may be MgO-rich (Mitchell, 1973). However, in the picritic basalts the ilmenite often appears as fragile, dendritic to skeletal crystals, which have formed by rapid, *in situ* crystallization. Such a delicate texture could not have been preserved during entrainment in a magma erupting from great depth. While this eliminates the possibility of their being high-pressure phases, it allows for an alternative explanation—that of disequilibrium crystallization, and hence that they are not true equilibrium compositions. However, the compositions of the ilmenite in the picritic basalts are similar to those in the Mount Ayliff Intrusion, which are larger subhedral to euhedral crystals (Fig. 2*e*) formed in a slowly cooled magma. Thus, rapid disequilibrium crystallization cannot be the general process producing high-MgO ilmenite.

Thompson (1976) showed that there was a positive correlation between the MgO content of ilmenite and the MgO content of the liquid from which it crystallized. The maximum MgO concentration in ilmenite which he produced experimentally was 5.3% even when it crystallized only 20 °C below the liquidus. This is comparable to that found in the Karoo picrites, but only half that of the maximum value reported from the Mount Ayliff Intrusion. However, the rock used in these experiments, while not being as MgO-rich as the compositions of the picritic basalts and that proposed for the Mount Ayliff magma, significantly contained 4.15% TiO_2 and so was extremely enriched in this element compared with normal basalt. As a result of the high-TiO_2 content, ilmenite will be a near-liquidus mineral. Experiments on tholeiitic samples with more typical MgO and TiO_2 contents show that ilmenite will not crystallize until close to the solidus (Walker *et al.*, 1976).

Geochemical evidence also shows that ilmenite is usually a near-solidus phase in tholeiitic rocks. For example, data from mid-ocean ridge basalt samples indicate that ilmenite does not begin to crystallize until the TiO_2 content of the liquid reaches 4% (Walker *et al.*, 1979). In a differentiating continental basaltic sequence, Eales *et al.* (1981) showed that ilmenite and titanomagnetite began to crystallize when the TiO_2 content reached 2%. Thus, the near-liquidus appearance

Table 1. Representative analyses of ilmenite grains from the Karoo Igneous Province.

	1	2	3	4	5	6	7
TiO_2	50.02	53.05	51.89	52.00	42.98	46.38	50.39
Al_2O_3	0.29	0.19	0.10	0.26	0.53	0.13	0.23
Fe_2O_3	7.54	3.40	4.70	3.06	21.28	15.65	5.46
Cr_2O_3	0.26	n.d	n.d	n.d	0.10	0.16	n.d.
FeO	36.49	36.48	37.36	40.27	30.79	32.90	41.77
MnO	0.38	0.28	0.38	0.94	0.29	0.36	0.32
MgO	4.46	6.14	5.03	3.11	4.29	4.74	1.79
CaO	0.12	n.d	n.d	n.d	n.d	n.d	0.02
Total	99.56	99.54	99.46	99.64	100.21	100.35	99.98
$FeTiO_3$	75.17	73.96	76.38	83.39	63.28	67.22	87.25
$MgTiO_3$	16.37	22.10	18.31	11.43	15.57	17.26	6.64
$MnTiO_3$	0.80	0.57	0.79	1.96	0.60	0.74	0.67
Fe_2O_3	6.99	3.09	4.37	2.84	19.68	14.38	5.11
Al_2O_3	0.42	0.27	0.14	0.38	0.76	0.23	0.34
Cr_2O_3	0.25	0.00	0.00	0.00	0.10	0.15	0.00

Analysis by Cambridge Microscan-5 Microanalyser (see Bristow, 1984). n.d - not detected. Analyses 1-6 are MgO-rich and come from picritic basalts; numbers 1-4 being of the high-TiO_2, low-Fe_2O_3 ilmenite type. Analysis 7 is a typical MgO-poor ilmenite, and comes from the MgO-poor lava suite. Ferric iron content is determined by calculation from the ideal ilmenite stoichiometry.

of ilmenite in the Karoo picritic basalts seems to imply a high TiO_2 content of the magma.

The calculated Fe_2O_3 contents of some of the ilmenite analyses from the picritic basalt, and those from the MgO-rich ilmenites from the Mount Ayliff Intrusion, are lower than found in other geological settings, except for lunar samples (Frondel, 1975). It is suggested that these low ferric contents may reflect crystallization under low oxygen fugacity. There are some experimental data which support this hypothesis. Woermann *et al.* (1969) determined the composition of ilmenite as a function of f_{O_2} and their results are included in Fig. 5. Their curves tend to converge with increasing MgO content of the ilmenite, and so are less diagnostic, but the data are consistent with the hypothesis that the ilmenite in the picritic basalt crystallized under relatively reducing conditions. While this may explain the low hematite component of the ilmenite, reducing conditions, by themselves, will not stabilize ilmenite, as shown by the experimental study of Usselman and Lofgren (1976). Hence the near-liquidus appearance of the ilmenite in a tholeiitic magma still requires some additional atypical parameter.

Bristow (1984) has argued that the picritic basalts are not simply olivine-enriched low-MgO basalts as suggested by Eales and Marsh (1979), even though there has undoubtedly been some accumulation of olivine in these rocks. This is based on the fact that the picritic basalts are enriched in the incompatible elements such as TiO_2, K_2O, Sr, Ba, and Zr compared to low-MgO tholeiite, which is the opposite from that expected if they contained abundant intratelluric olivine. This high TiO_2 content (which may range up to 3%) permits the near-liquidus crystallization of ilmenite and, as the liquid still has a high-MgO content at this stage, the ilmenite will be correspondingly enriched in MgO as shown by the experiments of Thompson (1976). Thus the formation of high MgO ilmenite is consistent with crystallization from a magma rich in both MgO and TiO_2.

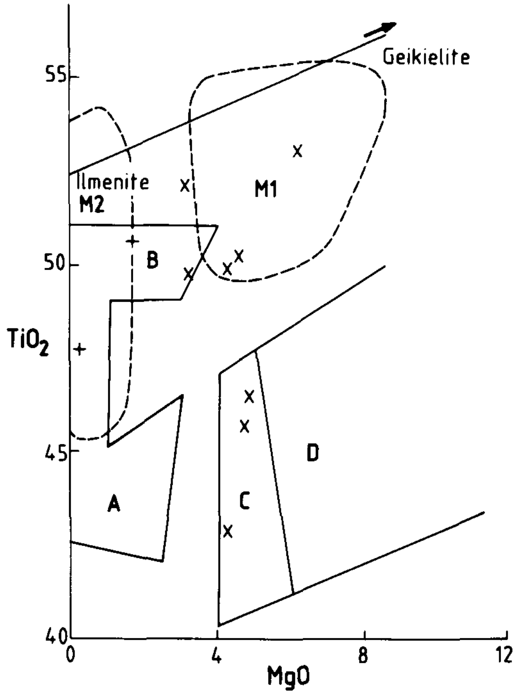


FIG. 4. Plot of MgO *vs.* TiO_2 in ilmenite. Crosses represent Lebombo picrites; pluses Lebombo low-MgO basalts. Fields M1 and M2 from Mount Ayliff are explained in Fig. 3. Note the high TiO_2 content of MgO-rich ilmenites from both localities. Typical fields of ilmenite compositions are from Haggerty (1976). A, ilmenites from felsic rocks; B, tholeiitic rocks; C, alkali basaltic rocks; D, kimberlites.

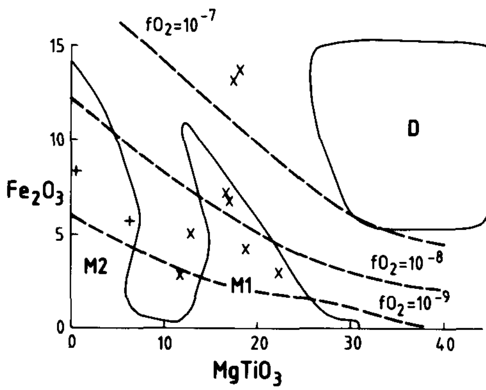


FIG. 5. Plot of Fe_2O_3 *vs.* MgTiO for ilmenite from Lebombo and Mount Ayliff; symbols as in Fig. 3. Some magnesian ilmenite compositions recalculate to give extremely low Fe_2O_3 contents. Oxygen fugacity curves are from Woermann *et al.* (1969).

Conclusions

Magnesian ilmenite occurs as microphenocrysts in picritic basalts in the Karoo Province. The observed crystallization sequence in some of these lavas is olivine, followed by ilmenite. This unusual sequence and mineral composition reflects crystallization of an atypical magma rich in MgO, TiO_2, and other incompatible elements, and possibly under a reducing environment. An analogous situation exists in the Mount Ayliff Intrusion, where near-liquidus ilmenite has a high MgO content (Cawthorn *et al.*, 1985). As the ilmenite in the picritic basalts represents a primary composition, it is suggested that the ilmenite compositions in the Mount Ayliff Intrusion are also primary, and that this intrusion has crystallized from a magma with similarities to that of the picritic basalt. These observations indicate that MgO-rich ilmenite may crystallize from high-MgO tholeiitic magma and is not necessarily the result of subsolidus reequilibration, as suggested the Lightfoot *et al.* (1987).

Acknowledgements

Most of the analyses from the Lebombo were obtained at the University of Cape Town. R.G.C. and J.W.B. acknowledge the financial support of the C.S.I.R. (South Africa). J.W.B. acknowledges the permission of Anglo-American Corporation to publish these results. We thank Dr John Bowles for his comments on this manuscript.

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[*Manuscript received 17 February 1988; revised 27 May 1988*]