Unusual perovskite textural relationships in olivine melilitites from Namaqualand–Bushmanland, South Africa

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ABSTRACT. Textural relationships shown by oxide phases in olivine melilitites from Namaqualand–Bushmanland, South Africa, indicate early crystallization of picroilmenite from a relatively reduced magma. Evolution of a CO_2-rich vapour during magma ascent led to an increase in oxygen activity and liquidus temperatures, causing rapid crystallization of titanomagnetite. Higher oxygen activities, together with the increase in calcium activity following CO_2 loss from the magma, resulted in perovskite replacing ilmenite as the stable Ti phase.

PEROVSKITE is a scarce mineral in igneous rocks and, when present, indicates crystallization from a liquid having silica activity (a_{SiO_2}) on or below the perovskite–sphene–silica buffer. It is an ubiquitous phase in olivine melilitites and kimberlites, reflecting the TiO_2-rich, undersaturated nature of these alkaline rocks. In a suite of olivine melilitite pipes from the Namaqualand–Bushmanland region, South Africa (see Moore, 1976, for location), the mineral occurs in a variety of unusual textural relationships that have bearing on the evolutionary history of these volcanics.

Petrography. The Namaqualand–Bushmanland olivine melilitites (Moore and Erlank, 1979) are porphyritic rocks with euhedral and often skeletal olivines dominating the phenocryst assemblage. Very scarce picroilmenite phenocrysts (with up to 12% MgO) have been identified in several pipes. Titanomagnetite occasionally occurs as phenocrysts, although it is more typically a microphenocryst phase, frequently with skeletal habit, and always shows a continuous range in size down to a groundmass dusting, indicating an extended period of crystallization. Perovskite microphenocrysts are commonly present as equant, somewhat rounded anhedra with a rather limited size range (0.02–0.16 mm in diameter), and occasionally as characteristic interpenetrant twins. Fine scale poly-

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synthetic twinning in several orientations is generally present. Modal perovskite proportions in the olivine melilitites range from 1% to 9%; approximately. Melilite occurs as phenocrysts or microphenocrysts in many, though not all, of the rocks. Melilite-free rocks typically have a coarse-grained groundmass assemblage, indicating relatively slow cooling rates on emplacement. The absence of melilite in such coarse-grained rocks is ascribed to breakdown of the phase to clinopyroxene and nepheline at low temperatures and pressures (Moore and Erlank, 1979). The early crystallizing phenocryst and microphenocryst phases are set in a groundmass mosaic of interlocking clinopyroxene, interstitial nepheline, and a second generation of granular groundmass perovskite.

Unusual perovskite textural relationships. (i) Perovskite typically exhibits complex, though consistent textural relationships with the rare ilmenite phenocrysts in the olivine melilitites. The ilmenite phenocrysts, when present, are invariably mantled by a rim of elongate and angular skeletal titanomagnetite 'fingers' which are in turn surrounded by granular perovskite and an outer discontinuous rim of titanomagnetite anhedra (fig. 1). An unidentified colourless phase, probably a zeolite, is always intimately associated with these intergrowths. An unequivocal interpretation of the textural relationships described is not possible. However, the rim of skeletal titanomagnetite armouring the ilmenite shows that the associated perovskite did not crystallize as a result of a simple reaction relationship involving ilmenite + liquid → perovskite. Moreover, the angularity of the titanomagnetite 'fingers' is more readily interpreted in terms of rapid crystallization rather than resorption of this phase. It therefore also seems unlikely that perovskite crystallization was controlled by a reaction relationship between liquid and titanomagnetite. Rather, it would appear that perovskite simply crystallized about a suitable nucleus, and thus
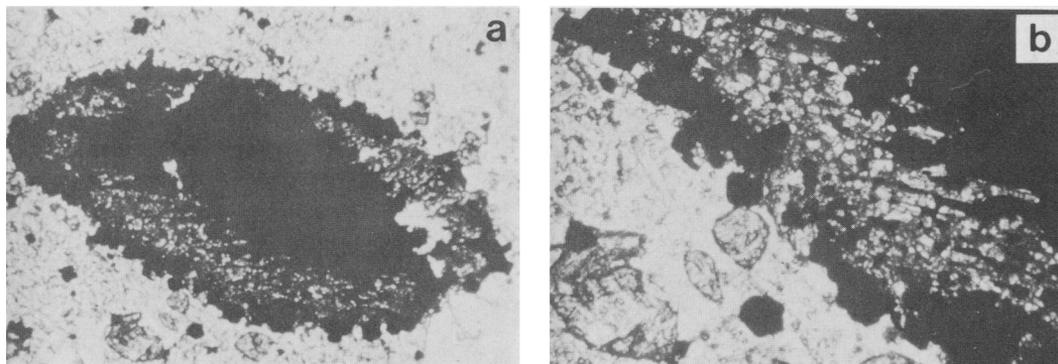
FIG. 1. (a) Picroilmenite phenocryst surrounded by skeletal fingers of titanomagnetite (both opaque), a mantle of granular perovskite (light grey), and an outer rim of anhedral titanomagnetite. The colourless phase in the intergrowth is a zeolite. Note the equant perovskite microphenocrysts (grey) adjacent to the intergrowth. (Plane polarized light; field length = 1.4 mm). (b) Detail of fig. 1a. Field length = 0.6 mm.

temporarily replaced titanomagnetite as a stable liquidus phase.

(ii) Perovskite has also been found associated with minor titanomagnetite as a smooth, thin rind (1 mm thick) armouring a rounded spinel harzburgite xenolith that was recovered from one of the olivine melilitite pipes. The rind appears to resemble closely the perovskite–spinel rims armouring large ovoid olivines in the Igwisi Hills 'kimberlite' (Reid et al., 1975). The textural relationships in both rocks indicate that crystallization of perovskite and spinel occurred subsequent to rounding of the enclosed host. In addition, the smooth contact between rind and enclosed host does not suggest that they are related by a reaction relationship with the liquid. A reaction relationship between olivine and liquid to form perovskite is, in any event, difficult to envisage. It therefore appears that the latter phase has once again simply crystallized about suitable nuclei.

(iii) A very unusual banded perovskite-rich inclusion (fig. 2) has been recovered from the Zwartheuwel pipe which is located 1 km to the north of the village of Gamoep (29° 52′ S., 18° 24′ E.). The lowermost layer of the inclusion (as illustrated) is dominated by elongate perovskite aggregates that appear to have grown radially outwards from a nucleating surface that is no longer preserved, producing a texture resembling comb layering (Drever and Johnson, 1957). Titanomagnetite, perovskite, olivine, and nepheline occur interstitially to the elongate perovskite aggregates and become more abundant towards the top of the first layer, which is marked by an abrupt change in modal mineral proportions. Elongate perovskite aggregates growing subnormally to this boundary surface dominate the second layer. These aggre-

gates are somewhat shorter than those in the first layer. Titanomagnetite, nepheline, olivine, and equant perovskite anhedra are once again present as interstitial phases. The second layer is succeeded by a narrow band rich in titanomagnetite, followed by a third layer dominated by elongate perovskite aggregates (shorter than those in the previous two), a second narrow band rich in titanomagnetite, and, finally, a layer (which is not shown in fig. 2) dominated by intergrown clinopyroxene and nepheline together with granular perovskite and titanomagnetite. Some of the clinopyroxenes in this final layer occur as branching crystals which, like the comb layering shown by the perovskite, are interpreted as rapid growth features.

The minerals present in the perovskite-rich xenolith described all occur in the host olivine melilitite. Those in the lowermost layers (as illustrated) are relatively early-crystallizing phases in these volcanics, while those in the final layer typically occur in the olivine melilitite groundmass assemblage. The layering sequence described thus closely mimics the order of crystallization in the host magma, and there can be little doubt that the inclusion is cognate rather than accidental. The nature of the nucleating surface for the inclusion is uncertain, but may have been the walls of the conduit or feeder channels to the magma at depth.

The origin of the mineralogical layering in the inclusion seems most likely to have been related to the interaction of crystallization and diffusion rates in the magma, as outlined by Drever and Johnson (1957). Thus rapid crystallization of one phase (for example perovskite) relative to ionic diffusion rates might starve the growth surface of those components required for further crystallization of that mineral and lead to saturation of the
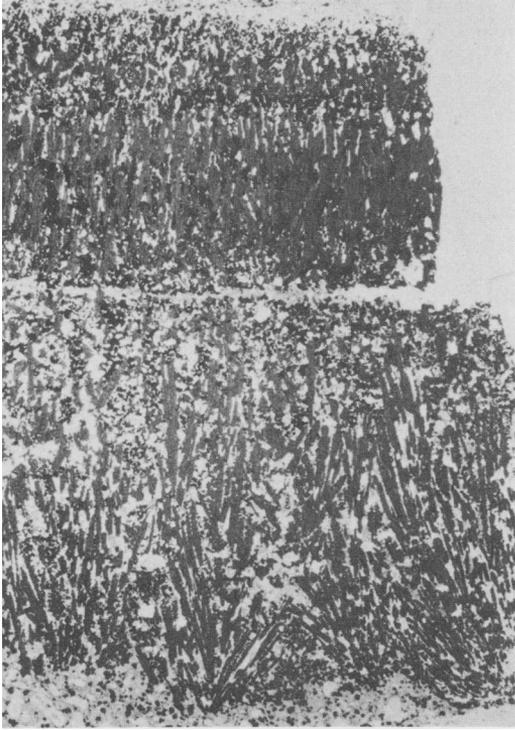
FIG. 2. Layered perovskite-rich nodule recovered from the Zwartheuwel pipe. Grey elongate needles are perovskite, opaque phase is titanomagnetite. The interstices are filled by olivine and nepheline (both colourless). (Plane polarized light; field length = 12 mm).

magma envelope adjacent to the nucleating surface by a second phase (for example titanomagnetite) which might in turn be precipitated. Saturation with respect to the first mineral might again occur, leading to renewed crystallization of that phase.

(iv) Rare pegmatitic segregations are found associated with some of the olivine melilitites and appear to be most abundant in the largest pipes. Such segregations vary in size from small rounded or ovoid bodies, less than 10 mm in diameter, to irregular schlieren, about a metre across. The pegmatites have essentially the same mineralogy as the groundmass assemblage of the olivine melilitites (i.e. clinopyroxene and interstitial nepheline with accessory titanomagnetite, perovskite, apatite, and phlogopite). Although coarse grained relative to the latter rocks, they have petrographic characteristics which indicate rapid crystallization: some of the smaller pegmatitic segregations have a narrow margin composed essentially of clinopyroxene and interstitial nepheline that is markedly finer grained than the central zone. Further, devitrified glass is present in some of the pegmatites, and

the apatite commonly occurs as hollow needles with a high length-to-breadth ratio (up to 54, fig. 3). Wyllie et al. (1962) have demonstrated experimentally that such skeletal apatite morphology is indicative of rapid crystallization. The perovskite in the pegmatites sometimes occurs as elongate aggregates along the grain boundaries of other phases (fig. 3). This will be referred to as 'dog-tooth' perovskite because of the serrated appearance resembling a row of dog's teeth. In view of the textural evidence presented for rapid crystallization of the pegmatites, the 'dog-tooth' habit is probably also related to rapid crystallization.

Discussion. Experimental evidence (e.g. Brey and Green, 1977; Wyllie and Huang, 1976) suggests that primitive olivine melilitite melts are CO_2-rich. The solubility of this component in silicate melts shows a positive pressure-dependance (Mysen et al., 1975) and, as a result, olivine melilitite melts might be expected to evolve a CO_2-rich vapour phase and possibly also an immiscible carbonate fluid (Moore and Erlank, 1979) during ascent to the surface. Moore and Erlank (1979) have proposed that oxygen activities in the olivine melilitite melts were initially low (between the Ni/NiO and Fe/FeO buffers), and that evolution of a CO_2-rich gas phase resulted in a marked increase in a_{O_2}. In addition, they suggested that loss of volatiles and the consequent increase in liquidus temperatures resulted in the rapid crystallization of near-liquidus phases, producing skeletal forms.

It is believed that the textural features described earlier can be interpreted in terms of the model
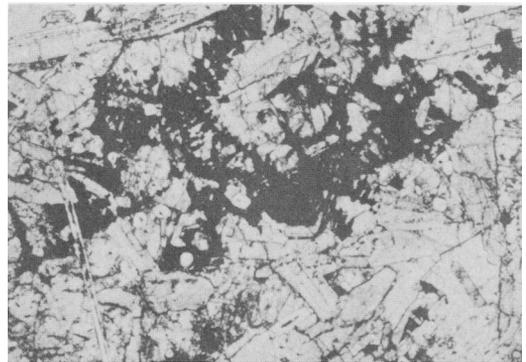

FIG. 3. Perovskite (opaque phase) with 'dog-tooth' habit in a nepheline–pyroxene pegmatite. The white skeletal needle at the left of the photomicrograph is apatite. The colourless equant grains are basal sections of skeletal apatite. Remaining phases are titaniferous clinopyroxene (broad grey laths) and interstitial nepheline. (Plane polarized light; field length = 5 mm).

proposed by Moore and Erlank (1979). It is envisaged that reduced phases such as ilmenite crystallized under conditions of relatively low oxygen activity. Rapid crystallization of more oxidized phases (e.g. the skeletal titanomagnetite fingers surrounding the ilmenite) would follow loss of a volatile phase as a result of the postulated increase in oxygen activity and liquidus temperature. Rapid titanomagnetite crystallization would deplete the residual magma in Fe, which might in turn trigger perovskite crystallization, with crystal growth favouring suitable nuclei such as ultramafic xenoliths or early-formed ilmenite–titanomagnetite intergrowths.

Textural relationships shown by the ilmenite–titanomagnetite–perovskite intergrowths indicate that ilmenite was the earliest-crystallizing Ti-rich phase. The reason for the initial suppression of perovskite stability is uncertain. However, one possibility is that complexing between Ca^{++} and carbonate anions in a CO_2-rich magma caused a decrease in calcium activity (a_{Ca}^{2+}) which, coupled with the high Fe contents of the olivine melilitites (13.5–22.9% Fe_2O_3—total Fe expressed as Fe_2O_3) and the low oxygen activities postulated, resulted in ilmenite initially being the stable Ti-rich phase. Loss of CO_2 during ascent may have led to an increase in a_{Ca}^{2+} and oxygen activity, resulting in perovskite replacing ilmenite as the stable Ti-rich phase.

The comb-layered texture of the layered perovskite inclusion could also be ascribed to rapid crystallization and a rise in liquidus temperatures following loss of volatiles from the magma. This would imply that perovskite was a near-liquidus phase in the olivine melilitites, which is not implausible as these rocks are extremely rich in Ca and Ti (10.1–16.9% CaO; 4.2–7.6% TiO_2).

The textures which characterize the nepheline–pyroxene pegmatites indicate rapid crystallization of these rocks. This may have been related simply to rapid chilling of late-stage pegmatitic liquids on emplacement in the upper crust. However, as the pegmatites are volatile-rich (over 6% weight loss on ignition at 1100 °C), it is possible that rapid crystallization was partly controlled by loss of volatiles and the resultant increase in liquidus temperatures at low pressures.

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