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Silicate-bearing rutile-dominated nodules from South African kimberlites: Metasomatized cumulates

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ABSTRACT

Rutile-dominated nodules from the Kampfersdam and Jagersfontein kimberlite pipes in South Africa contain euhedral olivines and spinels, graphic intergrowths of olivine and rutile, and massive rutile. In a similar nodule from the Pulsator dump in Kimberley, rutile and ilmenite are graphically intergrown with enstatite. These are interpreted as having crystallized from a Ti-rich silicate magma. The Jagersfontein samples contain Ca-Cr-Nb-Zr–bearing armalcolite that may also be magmatic or, together with Nb-rich titanite and calcite, may have formed by metasomatic reaction of the rutile-dominated cumulate assemblage with a Ca-rich fluid, possibly the residue from the magma that precipitated the cumulate assemblage. Despite some similarities in paragenesis and mineral chemistry, there are important differences between the rutile-olivine nodules and the MARID nodule–metasomatized peridotite group. These include the paucity of phlogopite and absence of K-richterite in the former, precluding a simple genetic relationship between the two groups.

INTRODUCTION

Evidence for metasomatism within the upper mantle has been documented in ultramafic xenoliths brought to the surface by kimberlites (e.g., Erlank and Rickard, 1977) and alkalic basalts (e.g., Wilshire et al., 1980). There is no agreement, however, as to whether the metasomatic minerals in the xenoliths are the result of alkalic igneous activity in the upper mantle, or whether the metasomatic event that formed the minerals was a precursor to the formation of the alkalic volcanic rocks (e.g., Wilshire et al., 1980; Menzies and Murthy, 1980; Boettcher and O'Neil, 1980; Jones et al., 1982).

Among ultramafic xenoliths found in kimberlites, a group of minerals, including phlogopite, amphibole, rutile, ilmenite, armalcolite, and members of the lindsleyite-mathiasite group (LIMA) have been proposed as indicators of metasomatic processes (e.g., Erlank et al., 1987). Although it has been suggested that rutile rich in Nb and Cr is likely a product of upper-mantle metasomatism (Haggerty, 1983; Erlank et al., 1987; Tollo and Haggerty, 1987), evidence is presented in this paper consistent with a magmatic origin for certain Cr- and Nb-rich rutile–dominated nodules. Incompatible element–rich fluids, which may have subsequently caused localized metasomatism, are inferred to have been liberated by the crystallization of rutile + olivine ± armalcolite and chromite from a Ti-rich silicate melt.

Rutile-dominated nodules have been described from kimberlites at Mbuji-Mayi in Zaire (Ottenburgs and Fieremans, 1979), Orapa in Botswana (Tollo et al., 1981; Tollo, 1982; Tollo and Haggerty, 1987), and Jagersfontein and Kampfersdam in South Africa (Haggerty, 1983; Schulze, 1983; Tollo and Haggerty, 1987). Unlike most occurrences of rutile in mantle-derived xenoliths such as eclogites (Shee and Gurney, 1979; Smith and Dawson, 1975), peridotites (Smith and Dawson, 1975), and MARID rocks (Dawson and Smith, 1977), in which it is present only as an accessory mineral, rutile from the localities cited above occurs in many nodules as the dominant mineral, associated with ilmenite, armalcolite, olivine, and other minerals. Graphic intergrowths of rutile and olivine (commonly serpentinized) occur at these localities and resemble nodules of intergrown ilmenite and clinopyroxene, which are important members of the Cr-poor megacryst suite of many kimberlites (e.g., Nixon and Boyd, 1973; Gurney et al., 1979).

This paper concerns two rutile-dominated nodules from the Jagersfontein kimberlite in South Africa, each of which contains graphic intergrowths of rutile and olivine. A third nodule, from the Kampfersdam kimberlite in Kimberley, resembles those from Jagersfontein, but the silicate is completely serpentinized and minor ilmenite is present.

After this study was virtually complete, a similar nodule was found at the Pulsator dump in Kimberley. It is dominated by rutile, but has substantial ilmenite, and, in places, rutile is graphically intergrown with enstatite. The results of a preliminary study of this nodule are included here for comparison with the three nodules studied in detail.

PETROGRAPHY

Small rutile-bearing nodules, ~0.8 cm in maximum dimension, occur in heavy-mineral concentrate at the

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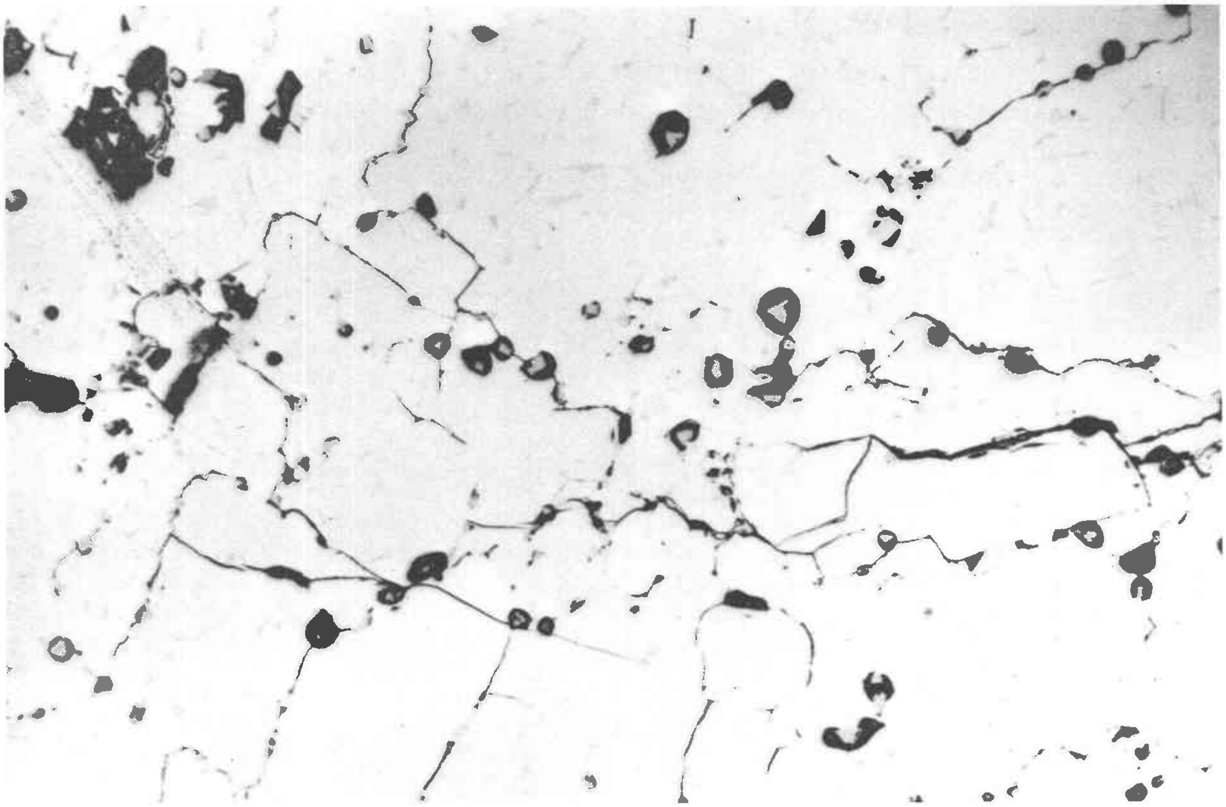

Fig. 1. Massive rutile (light gray) with included fine lamellae of ilmenite (medium gray rods) and interstitial euhedral grains of spinel, most of which are surrounded by black serpentine. Sample 12-96-3. Plane-polarized reflected light; field of view 350 μm across.

Kampfersdam pipe. One sample (12-98-4) is dominated by decussate-textured rutile grains as much as 0.5 mm across, with minor (<5 vol%) equant ilmenite grains interspersed throughout. A portion of this sample (~1 mm^2 exposed in polished section) consists of discontinuous lamellae of rutile (~0.5 mm long) in a matrix of serpentine.

Nodule 12-96-3 from Jagersfontein is ~4.0 cm in maximum dimension and also is dominated by decussate-textured rutile grains up to 0.5 mm across. Ilmenite occurs as sigmoidal lamellae up to ~2 × 10 μm in size within rutile grains (Fig. 1). Fresh olivine is present in this nodule as euhedra <1.1 cm long, with sharp boundaries against rutile-dominated domains. Also present are areas (~1 cm^2 in maximum size) of lamellar or graphic intergrowths of a single olivine crystal and a single rutile crystal, in places cored by rutile-free olivine grains (Fig. 2). The olivine in the core is in optical continuity with that in the surrounding intergrowth. Within the massive rutile are small (~5–10 μm) euhedra and subhedra of chromite (Fig. 1), generally along rutile grain boundaries and usually enclosed within serpentine. A small patch of phlogopite is in contact with rutile and olivine on one side of the nodule. Relatively coarse (1–2 mm) grains of strained and kinked mica dominate this patch that has locally recrystallized to smaller (~0.05 mm) strain-free euhedra. Armalcolite has nucleated on rutile in a thin (~20 μm) selvage between coarse euhedral olivine and massive rutile. Some massive rutile grains have undulose extinction, and some olivine euhedra have recrystallized in a few places into small (~0.05 mm) strain-free neoblasts. In only one graphic area is the olivine slightly deformed and recrystallized.

Another nodule from Jagersfontein (sample 12-96-23, ~3.0 cm across), also is dominated by rutile and contains euhedral olivine (Fig. 3), which, in places, is separated from rutile by phlogopite. Minor armalcolite occurs between the rutile and olivine + phlogopite, similar to its occurrence in 12-96-3. Interstitial to some rutile grains are assemblages in which the rutile is mantled successively by armalcolite, euhedral titanite, and calcite in the core (Fig. 4). One area of ~4 mm^2 consists of a graphic intergrowth of rutile and serpentine after olivine.

Lamellar intergrowths of rutile and ilmenite, similar to those described by Tollo et al. (1981), Tollo (1982), Haggerty (1983), and Tollo and Haggerty (1987) from Orapa and Jagersfontein, were found in the present investigation at both the Jagersfontein and Kampfersdam pipes. These authors have ascribed such intergrowths to subsolidus decomposition of armalcolite or a crystallographic shear–based structure. These intergrowths might instead repre-

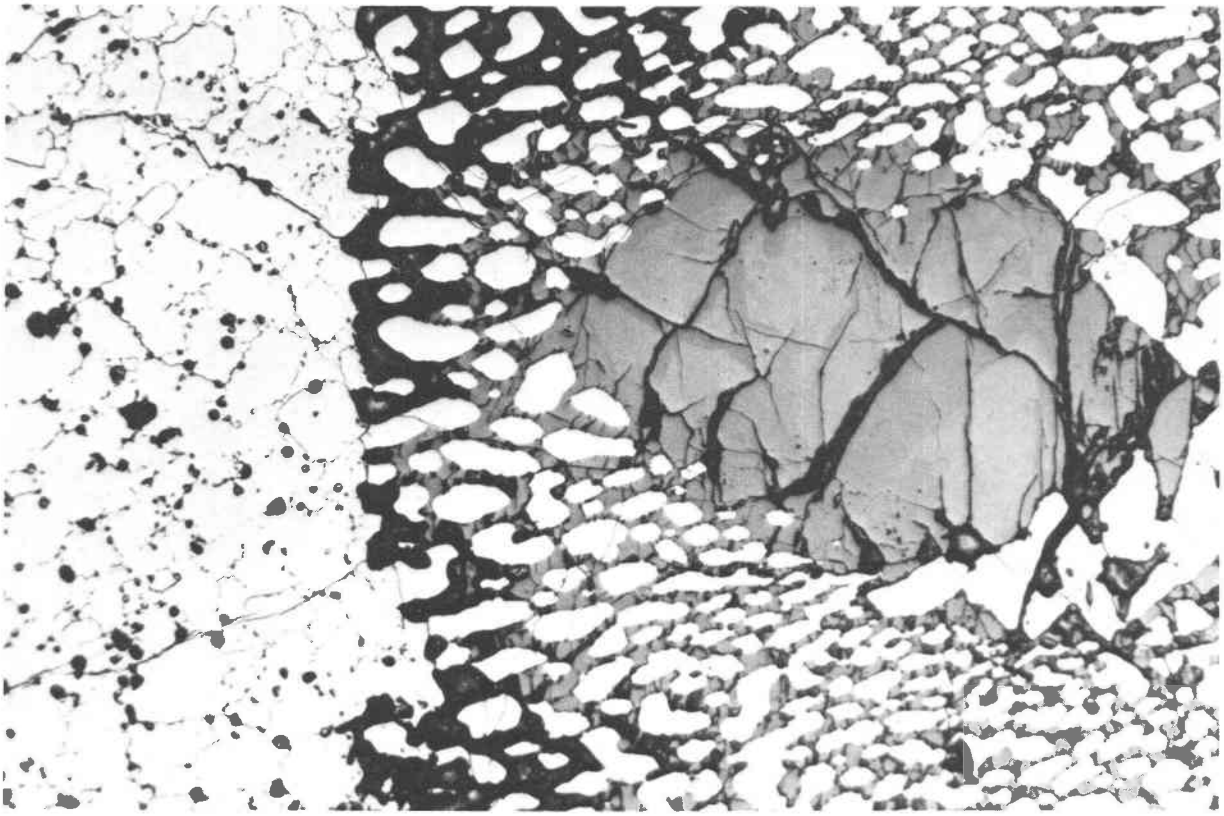

Fig. 2. Rutile-free olivine surrounded by graphic olivine-rutile intergrowth, in a matrix of massive rutile (left), with interstitial euhedral spinel. Sample 12-96-3. Plane-polarized reflected light; field of view 1.6 mm across.

sent replacement of ilmenite by rutile (Pasteris, 1980; Haggerty, personal communication, 1984). They are not considered further here.

MINERAL CHEMISTRY

Minerals in the Kampfersdam and Jagersfontein xenoliths were analyzed at the Geophysical Laboratory using a MAC 400 electron microprobe and procedures described by Boyd and Nixon (1978), with the exception that data for oxide minerals were reduced using ZAF correction procedures. The data for the Pulsator nodule was obtained with an ARL SEMQ electron microprobe at Queen's University using combined WDS and EDS methods and the correction procedures of Bence and Albee (1968). Each analysis in Tables 1–4 is an average of 3 to 13 individual analyses.

Rutile data represent analyses made with a fine (~2-μm diameter) electron beam to avoid ilmenite lamellae. The rutile in these samples is rich in Cr, Nb, and other minor elements, typical of rutile from other mantle xenoliths (Smith and Dawson, 1975; Wyatt, 1979; Tollo, 1982; Haggerty, 1983; Tollo and Haggerty, 1987), although not as enriched in these elements as are some rutiles from lamellar intergrowths with ilmenite from Orapa and Jagersfontein (Haggerty, 1983; Tollo and Haggerty, 1987). Ilmenite in the Kampfersdam sample does not contain detectable Nb, but is rich in Cr and Mg.

In the Jagersfontein samples, fresh olivines (Fo_{91}) in both textural domains (rutile-free and in graphic intergrowths with rutile) are uniform in most aspects of their chemical composition. Values for Mg/(Mg + Fe), MnO, CaO, Al_2O_3, and NiO do not serve to distinguish these olivines from most mantle-derived olivines (e.g., Boyd and Nixon, 1975). Both Cr_2O_3 and TiO_2 are low in coarse olivine, but higher in olivine in the graphic intergrowths. Within 10 μm of the rutile, the TiO_2 content of olivine reaches ~0.5 wt%, although these very high TiO_2 values near the rutile may be due to secondary fluorescence. The TiO_2 content at 100 μm from the rutile in the coarse olivine in Figure 2 is approximately that in the olivine not associated with rutile, ~0.05 wt%.

The phlogopite is hetereogeneous in composition, with neoblasts enriched in Fe, Ti, and Cr relative to coarse grains. Spinel euhedra in areas of massive rutile are magnesian chromites with high TiO_2 contents (8.24 wt%), and moderate ferric iron (4.24 wt% Fe_2O_3). Armalcolite, rich in minor elements, is the Cr-Ca-Zr-Nb (CCZN) variety and is chemically similar to those armalcolites analyzed by Haggerty (1983).

Titanites in 12-96-23 are strongly zoned. The magni-

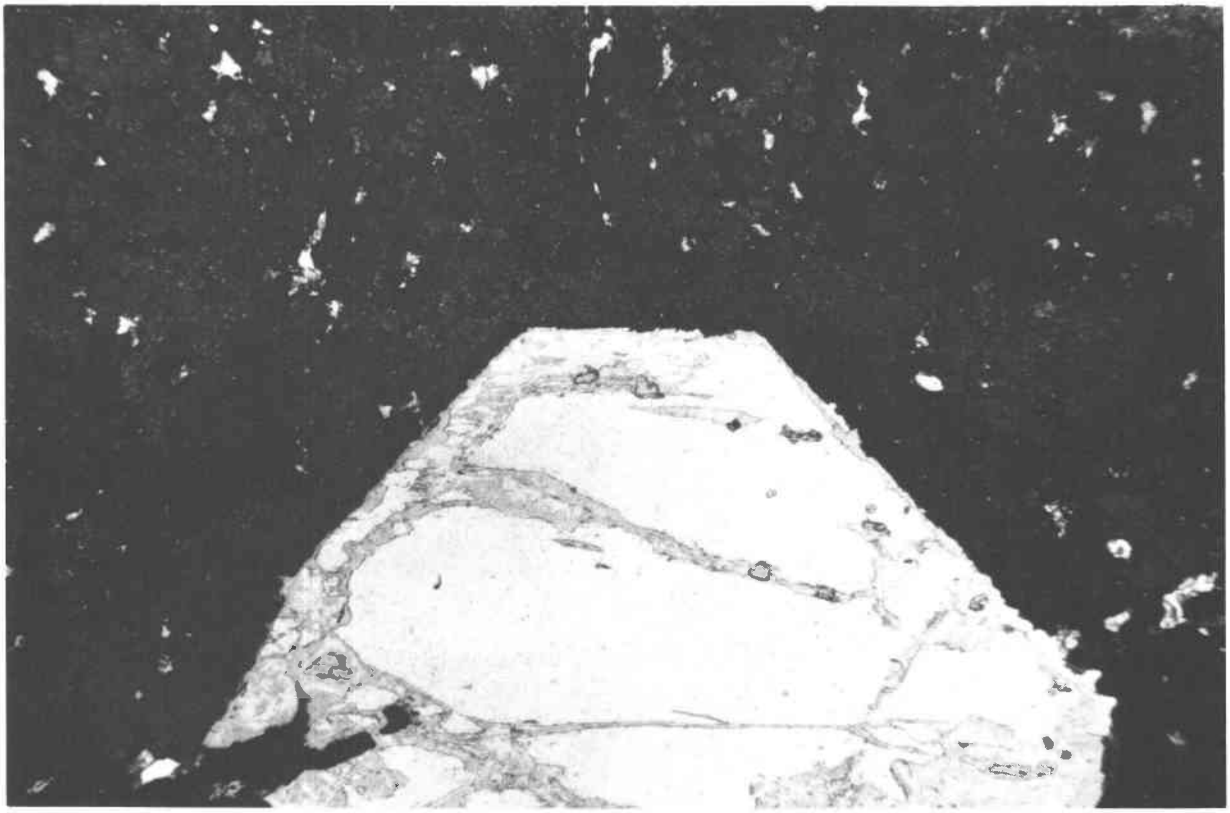

Fig. 3. Euhedral olivine grain, in matrix dominated by rutile. White patches in matrix are interstitial calcite + armalcolite + titanite assemblages. Sample 12-96-23. Plane-polarized light; field of view 4.5 mm across.

tude of chemical variation differs between grains, perhaps because of varying sections through different crystals, although certain changes are consistent. Extreme values occur in the cores and rims, respectively, in the following elements (expressed as wt% oxides) FeO, 0.1, 2.1; Al_2O_3, 0.7, 0.0; Nb_2O_5, 5.1, 0.2; Cr_2O_3, 0.0, 0.8. The average composition of titanite in this rock is listed in Table 3.

PETROGENESIS

Discussion of the origin of these nodules must take into consideration several important features that set this suite apart from most other mantle nodules. These features include the textural similarity of these nodules to graphic pyroxene-ilmenite intergrowths, the presence of euhedral olivine, and the unusual chemical composition of the phases.

TABLE 1. Compositions of minerals in rutile-ilmenite nodule 12-98-4 from Kampfersdam

	1	2	3
SiO_2	0.25	0.29	0.25
TiO_2	90.60	91.38	52.34
Al_2O_3	0.25	0.16	0.44
Cr_2O_3	5.22	4.99	6.11
Nb_2O_5	2.05	1.76	<0.03
FeO*	0.86	0.71	27.95
MnO	<0.03	<0.03	0.23
MgO	0.07	0.07	12.62
Total	99.31	99.36	99.96

Note: (1) Massive rutile. (2) Lamellar rutile in serpentine. (3) Ilmenite in rutile.

* Total Fe reported as FeO.

TABLE 2. Compositions of minerals in rutile-olivine nodule 12-96-3 from Jagersfontein

	1	2	3	4	5	6
SiO_2	<0.03	<0.03	41.16	41.86	41.19	0.13
TiO_2	91.63	92.07	0.04	0.76	1.99	8.24
ZrO_2	0.53	0.39	n.d.	n.d.	n.d.	n.d.
Al_2O_3	0.07	0.04	<0.03	11.88	12.18	2.75
Cr_2O_3	5.07	4.32	0.04	0.20	0.51	50.83
Nb_2O_5	1.64	1.03	n.d.	n.d.	n.d.	n.d.
FeO*	0.26	0.36	8.75	3.16	3.42	28.02
MnO	0.03	0.03	0.11	<0.03	0.03	0.19
MgO	0.04	0.17	50.84	26.04	25.37	10.44
NiO	n.d.	n.d.	0.33	0.16	0.16	n.d.
CaO	<0.03	<0.03	<0.03	<0.03	<0.03	n.d.
BaO	n.d.	n.d.	n.d.	0.03	0.06	n.d.
Na_2O	n.d.	n.d.	n.d.	0.17	0.26	n.d.
K_2O	n.d.	n.d.	n.d.	10.24	9.70	n.d.
Total	99.29	98.42	101.27	94.52	94.90	100.60

Note: n.d. = not determined. (1) Massive rutile. (2) Lamellar rutile intergrown with olivine. (3) Massive olivine. (4) Phlogopite (strained porphyroclasts). (5) Phlogopite (strain-free neoblasts). (6) Chromite euhedra [recalculation using the method of Finger (1972) yields Fe_2O_3 = 4.24 wt%, FeO = 24.21 wt%].

* Total Fe reported as FeO.

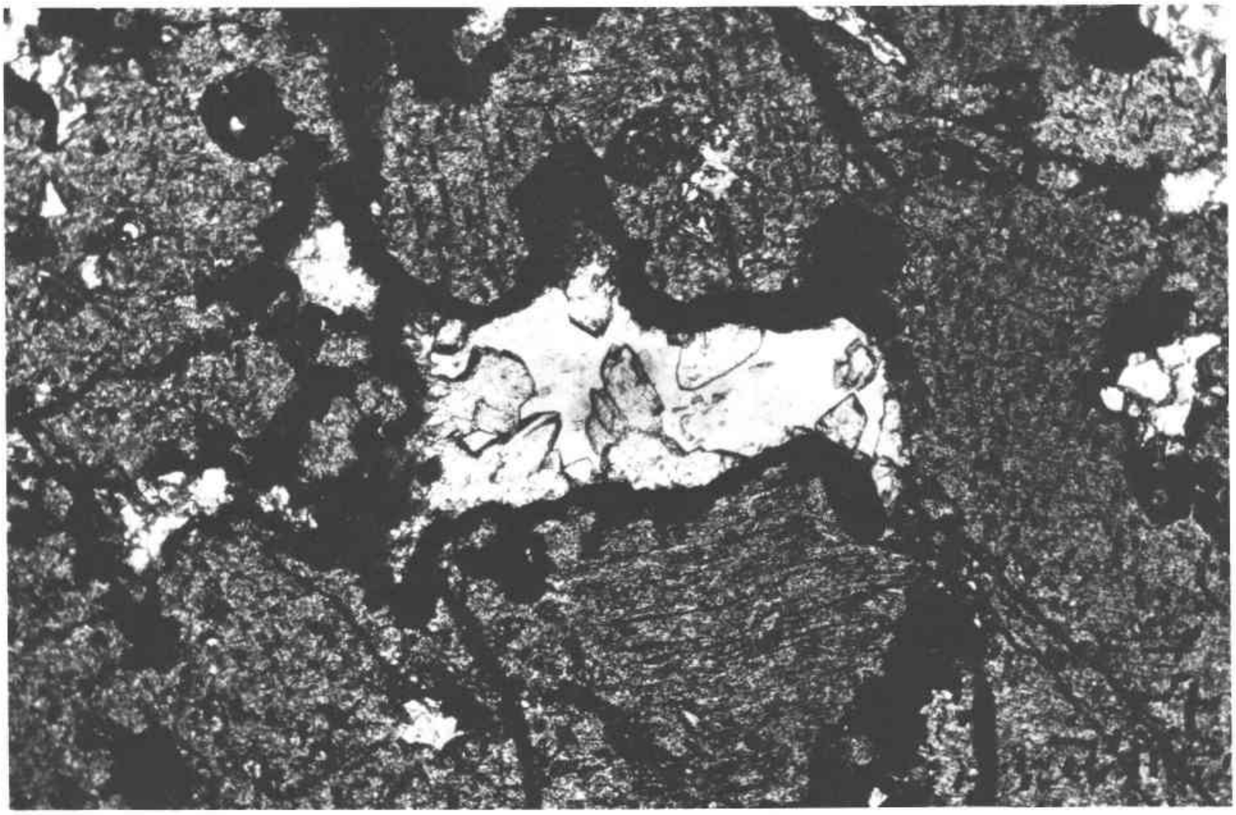

Fig. 4. Massive rutile (dark gray) with lamellae of ilmenite (fine black inclusions) and black armalcolite borders. The central interstitial assemblage consists of an armalcolite mantle on rutile, with euhedral high-relief titanite in low-relief colorless calcite. Sample 12-96-23. Plane-polarized transmitted light; field of view 1.1 mm across.

The textural similarity between the graphic intergrowths of rutile-olivine and pyroxene-ilmenite is somewhat superficial. In the rutile-bearing examples, the intergrowths form only a small portion of the nodules, which are mostly rutile. In graphic pyroxene-ilmenite intergrowths, each nodule is usually one crystal of pyroxene and one of ilmenite (McCallister et al., 1975), except in samples that obviously have been deformed and recrystallized (e.g., Pasteris et al., 1979). In addition, pyroxene-ilmenite nodules are only intermediate members of the texturally and chemically more extensive Cr-poor megacryst suite (e.g., Nixon and Boyd, 1973; Gurney et al., 1979; Robey and Gurney, 1979). Whether or not the rutile-olivine intergrowths are a part of a larger suite remains to be demonstrated, but they are not a part of the Cr-poor megacryst suite, as examples of rutile in these suites have never been demonstrated. The importance of the similarity between the rutile-olivine and pyroxene-ilmenite intergrowths lies in the fact that the latter have been shown to be of igneous origin, both experimentally (Wyatt, 1977) and by their intermediate position in a differentiation sequence (e.g., Gurney et al., 1979).

Table 3. Compositions of minerals in rutile-olivine nodule 12-96-23 from Jagersfontein

	1	2	3	4
SiO_2	40.98	<0.03	0.06	30.56
TiO_2	0.04	91.13	71.21	36.23
ZrO_2	n.d.	0.73	0.50	<0.03
Al_2O_3	<0.03	<0.03	0.76	0.19
Cr_2O_3	<0.03	3.98	6.20	0.35
Nb_2O_5	n.d.	2.13	0.82	1.72
FeO*	8.89	0.14	6.82	1.09
MnO	0.10	0.06	0.14	0.03
MgO	50.21	<0.03	6.05	0.15
NiO	0.32	n.d.	n.d.	n.d.
CaO	0.04	0.03	4.04	27.20
BaO	<0.03	n.d.	n.d.	n.d.
Na_2O	<0.03	n.d.	n.d.	n.d.
K_2O	<0.03	n.d.	n.d.	n.d.
Total	100.58	98.25	96.61	97.52

Note: n.d. = not determined. (1) Euhedral olivine. (2) Massive rutile. (3) Armalcolite. (4) Titanite.
* Total Fe reported as FeO.

Most mantle-derived olivines are anhedral, their shapes due to subsolidus processes, whereas euhedral olivines typically occur as phenocrysts in basic and ultrabasic magmas. The euhedral form of some of the coarse olivines in this study, as well as the graphic textures, is evidence in favor of crystallization of the rutile + olivine portions of these rocks from a magma. In the igneous-origin model of Schulze (1983), it was suggested that the minerals in sample 12-96-3 crystallized in the following

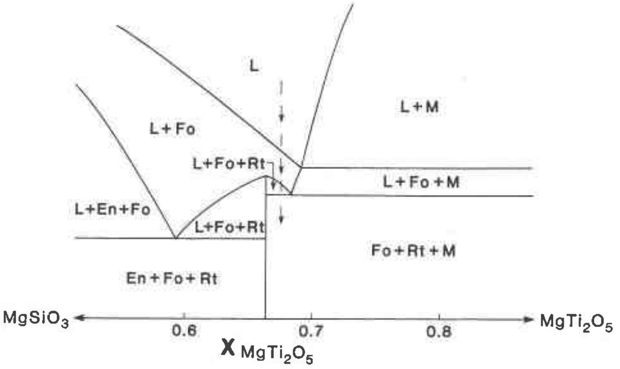

Fig. 5. Phase relations in a portion of the system $MgSiO_3$-$MgTi_2O_5$ at 40 kbar, between approximately 1400 and 1500 °C (MacGregor, 1969). The vertical dashed line represents a possible cooling path that could produce some of the features observed in the rutile-olivine nodules. L = liquid, Rt = rutile, Fo = forsterite, En = enstatite, M = $MgTi_2O_5$.

Table 4. Compositions of minerals in enstatite-rutile-ilmenite nodule from Pulsator dump, Kimberley

	1	2	3
SiO_2	56.11	0.28	0.21
TiO_2	0.57	89.50	52.97
ZrO_2†	n.d.	0.58	0.09
Al_2O_3	1.21	0.16	0.56
Cr_2O_3	0.50	4.59	3.31
Nb_2O_5†	n.d.	1.72	0.10
FeO*	5.98	1.08	27.17
MnO	0.11	0.04	0.21
MgO	34.30	0.19	14.18
CaO	0.87	0.00	0.04
Total	99.65	98.14	98.84

Note: n.d. = not determined. (1) Enstatite. (2) Rutile. (3) Ilmenite.
† Analyzed by WDS; all other elements by EDS.
* Total Fe reported as FeO.

order: (1) olivine, (2) olivine + rutile, (3) rutile + chromite. This sequence is similar to that in the slow-cooling experiments of Wyatt et al. (1975) on a melted clinopyroxene-ilmenite intergrowth (i.e., silicate followed by graphic silicate + oxide).

Further support for an igneous origin and for this crystallization sequence exists in experiments by MacGregor (1969) in the system $MgSiO_3$-$MgTi_2O_5$. At 15.5 kbar the field of liquid + forsterite is followed at lower temperatures by liquid + forsterite + rutile, at ~0.5 $MgSiO_3$, ~0.5 $MgTi_2O_5$. With increasing pressure, to 40 kbar, the field of liquid + forsterite + rutile expands and shifts toward $MgTi_2O_5$. The phase relations in a portion of this system at 40 kbar are shown in Figure 5. The dashed line represents a cooling path by which the sequence olivine, olivine + rutile, olivine + rutile + armalcolite could form. The textural relations in the titanite-bearing Jagersfontein nodule are consistent with a similar sequence, which may have continued with titanite + calcite. Whether these last two minerals are actually of igneous origin, or are metasomatic (see Haggerty, 1983), is uncertain, and is discussed below. As forsterite + rutile + enstatite is the subsolidus assemblage in this system at slightly more silica-rich compositions (Fig. 5), it is interesting to note that a rutile-dominated nodule that contains a graphic *enstatite*-rutile intergrowth has been found in the Pulsator dump suite from Kimberley (Table 4).

The constraints that can be imposed on the pressure and temperature conditions of formation of these rutile-rich nodules are very limited. Tollo (1982) and Tollo et al. (1981) suggested that the coarse lamellar intergrowths of rutile and ilmenite had originated by the breakdown of armalcolite and that the stability field of this mineral (maximum pressure limit = 14 kbar, Lindsley et al., 1974; Friel et al., 1977) could be used as one limiting possibility. The minor amount of ilmenite present in the Jagersfontein samples is not consistent with an armalcolite protolith, so its stability field cannot be considered as an appropriate constraint on the rutile-dominated parts of the nodules. Nevertheless, the presence of the CCZN armalcolite constrains the pressure of formation of the final sequence of minerals (armalcolite, titanite, calcite, phlogopite) to less than 14 kbar, if the experimental work on synthetic armalcolite bears on the unusual CCZN variety. The phase relations at 40 kbar expressed in Figure 5 are thus not likely to be directly applicable. Figure 5 represents a simplified system, however, and the addition of Fe and water, as well as minor components, would effect the position of boundaries in this system. The value of Figure 5 is that it illustrates that the rutile-olivine nodules *could* be magmatic rocks, although it does not impose restrictions on their pressures and temperatures of origin.

The nature of the parent magma that precipitated the rutile-olivine assemblage remains problematic. Although some lunar basalts have high TiO_2 contents (e.g., Papike et al., 1976), terrestrial magmas very rich in TiO_2 are not common. Boyd et al. (1984) postulated that Ti-rich pyroxenitic magmas (~13 wt% TiO_2 maximum) crystallized as ilmenite pyroxenites in the mantle, but in their search for crustal analogues found that only five Tertiary volcanic rocks were reported with >7 wt% TiO_2 (Boyd et al., 1984). The TiO_2 values of the liquids in the vicinity of the postulated crystallization path in Figure 5 are ~50 wt%! Such magmas are not likely to be either primary or common in the upper mantle, but might form by extreme fractional crystallization of less Ti-rich magmas.

In formulating the hypothesis that much of the rutile from Jagersfontein is due to metasomatic processes, Haggerty (1983) linked together the presence of minerals such as phlogopite, K-richterite, armalcolite, and "LIMA" (lindsleyite-mathiasite), most occurrences of which are considered to be products of mantle metasomatism (e.g., Erlank and Rickard, 1977; Erlank et al., 1982, 1987; Jones et al., 1982), with the unusual chemical compositions of the rutile and associated minerals, especially their high Nb and Cr contents. He had reservations, however, about including those rutiles that were lower in Nb and Cr in the metasomatic group. That interpretation is consistent with the model in this paper, in which the rutile-olivine intergrowths are considered to have formed by igneous processes. The enrichment of the armalcolite and titanite in Nb, Zr, and Cr is consistent with the metasomatic model suggested by Haggerty (1983), as is the presence of the volatile-rich minerals phlogopite and calcite. That the metasomatizing fluid was Ca-rich is supported by the strong gradient in Ca from the substrate rutile into the center of the interstitial spaces, filled successively by CCZN armalcolite, titanite, and calcite. The source of the metasomatizing fluid, the precursor to the titanate-bearing assemblages described by Haggerty (1983), is not known. The metasomatizing fluid may have been the residual fluid remaining after precipitation of the olivine-rutile assemblage, or it may have been $CaCO_3$-rich fluid associated with the kimberlite itself. Support for a cogenetic relationship between the metasomatizing fluid and the rutile-dominated cumulates is found in the textural relationship between the two assemblages. The "metasomatic" minerals occur interstitial to the rutile assemblage and generally do not occur along fractures or transgress grain boundaries, as might be expected from a "late" metasomatizing event unrelated to the cumulate assemblage.

There are similarities between the rutile-olivine nodules and both the MARID suite of nodules and the Kimberley metasomatized peridotites (e.g., Dawson and Smith, 1977; Jones et al., 1982; Erlank et al., 1987), including the common presence of rutile, phlogopite, and armalcolite and the high Nb, Cr, and Zr contents of these phases. More important, however, are the dissimilarities between the rutile-olivine rocks and the other groups. Although K-richterite and phlogopite generally dominate MARID nodules and are important constituents in metasomatized peridotites, phlogopite is only a minor constituent of the rutile-olivine nodules and K-richterite is absent. Furthermore, olivine was an early phase to crystallize in the rutile nodules, but is apparently not a magmatic or metasomatic component in the MARID nodule–metasomatic peridotite group. Finally, the metasomatic assemblage in the rutile-olivine nodules (armalcolite, titanite, calcite) points to a source fluid rich in Ca, unlike the K-rich fluid responsible for the metasomatic minerals in the peridotites. It is concluded that, in spite of some similarities, there is no simple genetic relationship between the rutile-olivine nodules and either the MARID nodules or the veined and otherwise metasomatized peridotites.

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