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Orbicular oxides in carbonatitic kimberlites

STEPHEN E. HAGGERTY[1,*] AND AGNES FUNG[2]

[1]Center for Analytical Electron Microscopy, Department of Earth Sciences, Florida International University, Miami, Florida 33155, U.S.A.
[2]CF Mineral Research Ltd. Kelowna, B.C. V1X4L1, Canada

ABSTRACT

A renewed interest in mineral spheres that exhibit oscillatory layering is assured with the spectacular discovery of hematite beads in Meridiani Planum, Mars. The Martian "blueberries" are variously interpreted as sedimentary concretions, impact spherules, or accretionary lapilli. We draw attention to a possible alternative that the spheres of oxides are of igneous origin in carbonate-rich upper mantle systems. Two spherical oxide assemblages from cabonatitic kimberlites in the central Namibia (formerly South West Africa) volcanic field show similarities and contrasts to orbicular structures in carbonatites from Uganda, South Africa, Germany, Finland, and Russia. Structures have rapakivi appearances that bear upon the controversial issue of a relation between kimberlites and carbonatites. The first example is from a carbonatitic kimberlite adjacent to the Mukorob kimberlite (*sesu stricto*). Oblate spheroidal nuggets (2 × 1.5 × 0.5 cm) have olivine (now serpentine + calcite) cores with concentrically layered bands of ilmenite + calcite, and pedestals of ilmenite ($Ilm_{55}Geik_{40}Hem_5$) with intergranular calcite; the spheres are matrix-supported in massive ferroan calcite + serpentine, with minor apatite (F = 1.5 wt%) and barite. The matrix contains xenocrystic Mg-Al chromite (55 wt% Cr_2O_3) mantled by ilmenite ($Ilm_{51}Geik_{43}Hem_6$) but the dominant groundmass oxide mineral is Mg-Al-Ti (MAT) magnetite. The second example is from the carbonatite facies of the Hatzium kimberlite, and is also in a dike. MAT-magnetite pellets (2–3 cm) have alternating wide (~1 mm), and thin (~0.25 mm) bands of magnetite nucleated on a complex mixture of calcite (FeO = 0.1 wt%) and serpentine (4 wt% FeO, 0.5–1 wt% Al_2O_3). The matrix assemblage of serpentine + calcite + MAT-magnetite has mineral compositions similar to the spheroids, but is distinguished by abundant Ba-phlogopite and lesser apatite (F = 2 wt%). Phlogopite is strongly zoned in patches and bands, is kink-banded and contains inclusions of MAT-magnetite + calcite. The assemblage is typical of phoscorites (also known as camaforites) in other carbonatites, but the ilmenite spheroids appear to be restricted to Mukorob. Our interpretation is that both types result from the nucleation of immiscible liquids (a high-density and high-viscosity ore-carbonate-silicate component in a low-viscosity phosphoric carbonatite melt) on olivine or olivine + magnetite, assisted by frothing and slow degassing on decompression and dike intrusion.

Keywords: oxide orbicules, kimberlite, liquid immiscibility, magnetite, Martian blueberries, carbonatite, ilmenite, Namibia

INTRODUCTION

Water has become a driving force in the search for extraterrestrial life. The stratigraphic horizons with abundant oxide spherules of hematite discovered by the Martian rover explorers are seemingly permissive lines of evidence that aqueous environments once existed in this currently barren terrain (Herkenhoff et al. 2004; Yen et al. 2005), a view supported by at least one example in the Jurassic Navajo Sandstone of Utah (Chan et al. 2004). But there are other alternatives: one posits hematite spherule formation in basaltic tephra (also under aqueous conditions), as in Hawaii (Morris et al. 2005); and another draws attention to sphere similarities in base surge and accretionary lapilli, and impact-related iron condensates (Knauth et al. 2005). We note that oxide spherules may also form in upper-mantle-derived carbonate-rich liquids. If not an alternative to the origin of Martian "blueberries," these terrestrial oxide bodies do bear on the genesis and evolution of complex mantle melts.

There are considerable uncertainties in the origin of kimberlites and in the genesis of carbonatites, but the issue that has fueled the most vigorous debate is on the potential relation, if any, between kimberlitic and carbonatitic magmas in petrologic models: one perspective is consanguinity (e.g., Bell 1989); whereas the alternative view favors a complete separation in the evolution of these two exotic rock types from the upper mantle (e.g., Mitchell 1986). Liquid immiscibility between conjugate carbonate and silica-undersaturated liquids has both supporters and detractors among experimentalists, but also among petrographers in studies of alkaline igneous provinces and mantle xenolith samples (Pyle and Haggerty 1994 and references therein). A fundamental criterion for the recognition of liquid immiscibility is the formation of mineral spheres, or spheres of mineral aggregates.

Spheres have the lowest surface to volume ratio of any enclosed geometric form. This minimal condition, although simple and unique, must be non-specific in the context of the origin of spherical bodies in nature, given the variety that exists.

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* E-mail: haggerty@fiu.edu

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From rapakivi granite to Pele's basaltic tears, from meteoritic chrondrules to glass beads in the lunar regolith, and from Martian "blueberries" to ooliths and mini-concretions, the environments and mechanisms of formation cover an extraordinarily wide spectrum, and possible origins. Is there a common and underlying physical process linking this diversity, or is it the case that all are surface tension related, and origins branch from this basic principle? In carbonatites, the formation of spherical bodies is evidently a two-stage process: The first involves liquid immiscibility of phoscorite (magnetite + apatite + forsterite, diopside, or phlogopite), also known as camaforite (*ca*-calcite, *ma*-magnetite, *fo*-forsterite), from a parent carbonatite (Zeitsev and Bell 1995; Wall and Zaitsev 2004); and the second is the formation of spherical bodies from the separated liquid (Keller 1981; Moore 1984). The second stage, although limited to magnetite, and not ilmenite, is fully articulated into an interesting model of formation by Lapin and Vartiainen (1983). The process of fluidization (i.e., gas-expansion and droplet formation in liquids), as outlined by Sutherland (1980), deserves recognition and is revisited in light of the present study and given the widespread distribution of orbicular structures in carbonatites worldwide.

We report on two unusual occurrences of spherical oxide assemblages that bear on the relation between kimberlites and carbonatites, and on oxide liquid immiscibility in phosphorous-rich (Philpotts 1967) and carbon-rich (Weidner 1982) systems. The assemblages show similarities and contrasts to orbicular, rapakivi-like structures in carbonatites, and to related rocks from Tororo, Uganda (Sutherland 1980), Kaisertuhl, Germany (Keller 1981), Palabora, South Africa (Moore 1984), Kovdor, Russia (Krasnova et al. 2004), Sokli, Finland, and Vuorijarvi, Russia (Lapin and Vartiainen 1983).

GEOLOGICAL SETTING

Kimberlites are widespread in Namibia (Fig. 1) but the setting is off-craton, and the province is therefore one in which no diamonds have yet been reported. This is consistent with higher than normal heat flow and thinner continental crust at the edges of schematically modeled sub-continental lithospheric keels (Haggerty 1986 and refs. therein). Known as the Gibeon Province, at least 46 kimberlite pipes and 16 dikes (Janse 1975), as well as many closely associated carbonatites (Janse 1969; Lorenz et al. 1997) are reported in a NW-SE trending corridor between Keetmanshoop and Mariental (Fig. 1). An unpublished, 1984 exploration database lists 125 localities (De Beers pers. comm.). Intrusions at Mukorob and Hatzium are the subject of this study. The former is 90 km N, and the latter 180 km NNW of Keetmanshoop (Fig. 1). The enormous Gros Brukkaros carbonatite complex dominates the area, 50 km to the SW of Murorob. Kimberlites and carbonatites are Mid-Cretaceous in age (80–120 Ma), and are intruded into Nama Group Cambrian sediments, and into the Karoo (Carboniferous-Jurassic) sequence of volcanics and sediments (Fig. 1).

SAMPLES AND ANALYTICAL PROCEDURES

The Mukorob samples were collected in 1986 during a University of Cape Town field trip led by John Gurney, and the Hatzium material in 1988 during a De Beers trip led by Hennie van der Westhuizen. Good solid blocks of the Mukorob dike were obtained, but the host rock from Hatzium was highly fragmental and difficult to sample.

Polished and polished-thin sections were prepared for binocular and petrographic (air and oil-immersion) microscope study of individual oxide spheres and of spheres embedded in host rocks.

Mineral compositions were obtained on a CAMECA SX50 electron microprobe at the University of Massachusetts, Amherst, using the standards and procedures given in Fung and Haggerty (1995), and on a JEOL SEM with EDAX in the Florida Center for Analytical Electron Microscopy, Florida International University.

PETROGRAPHY AND MINERAL CHEMISTRY

Ilmenite nuggets at Mukorob

This locality has a diatreme outcrop (~100 m in diameter) of kimberlite *(sensu stricto),* and a closely associated, but non-intersecting dike (Janse 1975). The dike pinches and swells, has a maximum width of ~0.5 m (Fig. 2a), and is exposed for ~200 m. Frankel (1956) described the dike rock as a calcitized olivine melilitite. Our interpretation of the mineralogy and petrochemistry is that the term carbonatitic kimberlite is more appropriate. Olivine, now serpentine, is abundant, euhedral laths of calcite after aragonite, and remnants of dendritic calcite in this dike are similar to fresh, skeletal calcite in the Benfontein Sills, Kimberley, South Africa (Dawson and Hawthorne 1973). The calcite is magmatic and is compositionally and texturally distinct from secondary calcite in veins. The groundmass, moreover, has apatite + barite, two minerals typical of carbonatites (Kapustin 1980).

The most interesting feature of the buff-colored rock (the color is typical of weathered carbonatites) is the great abundance (10–20 vol%) of black, oblate spheroidal nuggets, 1 × 2 cm in diameter and ~0.5 cm in thickness (Fig. 2b). The nuggets are remarkably uniform in size and are matrix-supported, with cores of olivine (replaced by serpentine), calcite, and traces of quartz, concentrically overgrown by calcite, and by a thin (~0.25 mm) inner layer and a thicker (~0.5 mm) outer band of ilmenite (Figs. 3–4). Linked by delicate whiskers (Figs. 4a–4e), or coarser pedestals of ilmenite (Figs. 3 and 4a), these connecting oxides are radial in cross-section and normal to the concentric bands. The enclosed volumes consist of granular calcite and finely dispersed ilmenite (Figs. 3–4). The inner layer and the outermost band are intimately intergrown with calcite and finely disseminated ilmenite, making the layers dark and virtually opaque (Figs. 4a–4c). Granular calcite lends a smooth wavy appearance to the ilmenite (Fig. 4c), whereas vestigial skeletal calcite results in a jagged montane profile (Figs. 4b and 4d). The enhanced reflectivity is an artifact of differential hardness, curvature to the ilmenite surface, and oblique lighting. Ilmenite is polycrystalline and exhibits equilibrated, 120° dihedral angles within the ilmenite zones (Fig. 3) and between ilmenite and interlocking calcite; the outer band shows stress-induced twinning. A cuspate texture of ilmenite with intervening granular calcite marks the very edge of the sphere in contact with the groundmass (Fig. 3d).

Some spheres have internal orbicular bodies with cores of trapped polycrystalline calcite and haloes of coarse equilibrated ilmenite (Fig. 4f with details in 4g and 4h). The spherical structures are internally delicate yet remarkably robust; the disrupted inner band shown in Figure 4a (with details in Fig. 4b), is rare in the samples from our study. More commonly are the indented (soft shell), outer rims of spheres, some portions of which were sufficiently rigid to have undergone brittle fracture and fragmentation (Fig. 4i).

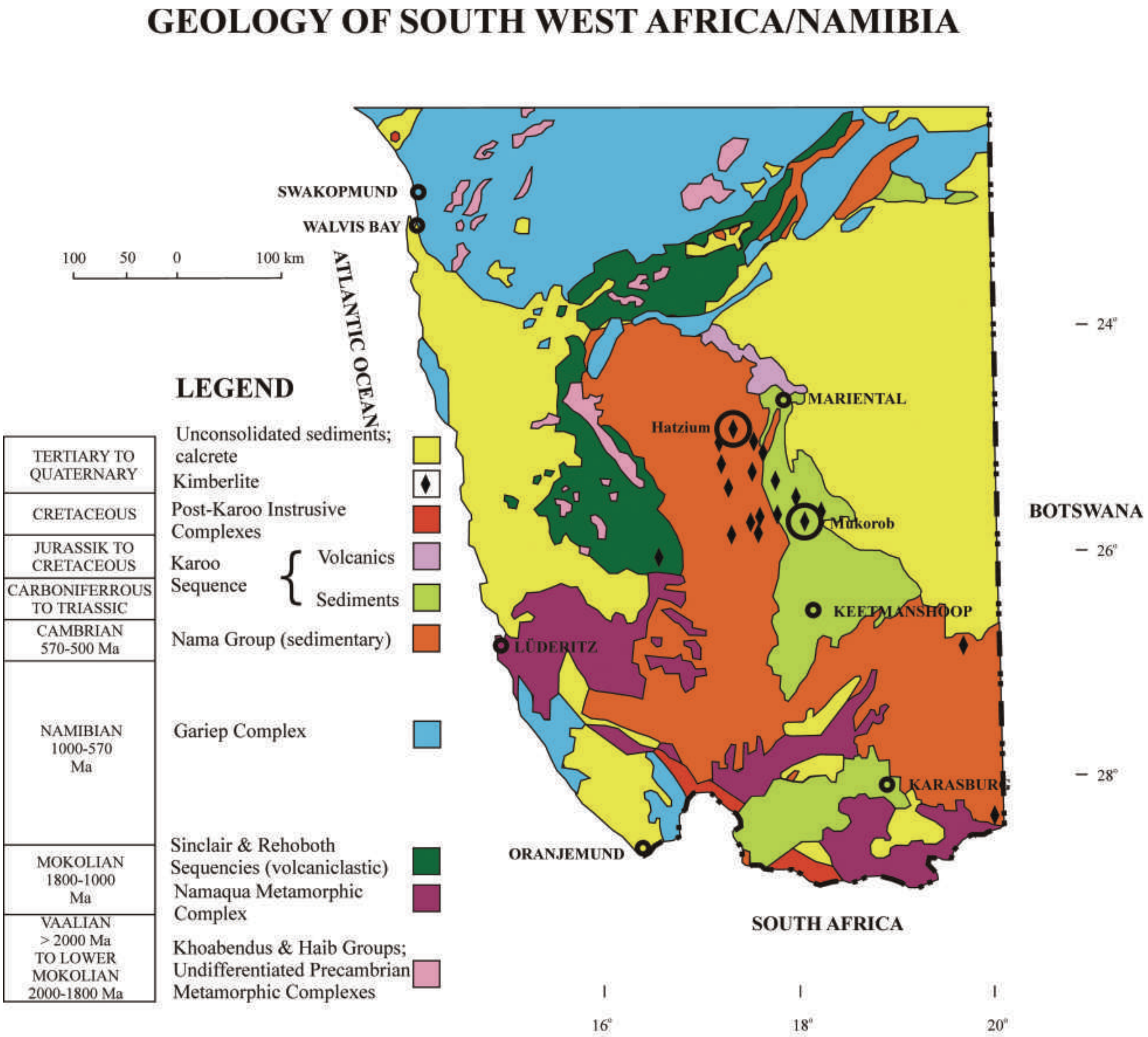

FIGURE 1. Generalized geology of the southern section of Namibia showing the centralized cluster of kimberlites and associated carbonatites in a broad NW-SE trend. Mukorob and Hatzium are the subjects of this study. Modified from the "The Geology of Southwest Africa – Nambia" (Geological Survey 1982).

Primary brown spinel is widespread in the groundmass and in some grains is overgrown by a second spinel that is also brown in oil-immersion reflected light; xenocrysts of gray spinel cores, on the other hand, are overgrown by polycrystalline ilmenite. Relatively minor magnetite and hematite are secondary minerals in serpentinized olivine. The groundmass assemblage of spinel + calcite + serpentine is typical of kimberlite (Mitchell 1986, 1995).

Mineral chemistry

Ilmenite, whether from inner or outer bands, or in dispersed blebs, is remarkably similar in composition (Table 1), with 11–12 wt% MgO, 2–3 wt% Cr_2O_3, and minor Mn and Nb. Compositions are essentially a solid solution between $FeTiO_3$ (ilmenite) and $MgTiO_3$ (geikielite), with minor Fe_2O_3 (hematite). From low contents of Fe^{3+}, and the distribution of ternary f_{O_2} isobars (Fig. 5a), we conclude that ilmenite crystallized in a relatively reducing (~FMQ-1) environment. Compositions fall close to the base of the ternary, and well within the megacrystic ilmenite field for kimberlites worldwide (Fig. 5a). Groundmass ilmenite is rare in kimberlite, and where present, the host rock has affinities to carbonatites, and ilmenite is enriched MnO (Tompkins and Haggerty 1985). A further test of the kimberlitic affinity of ilmenite is illustrated in a plot of MgO-Cr_2O_3 (Fig. 5b). The Mukorob data fall on the MgO-rich side of the parabolic distribution, typical of kimberlites (Haggerty 1991); compositions of megacrystic ilmenite from the adjacent Mukorob kimberlite have 7–12 wt% MgO, but are depleted in Cr_2O_3 (Mitchell 1987), relative to ilmenite in the dike spheres.

Spinel cores are of two distinct compositions: brown spinel is enriched in MgO (~15 wt%), Al_2O_3 (~13 wt%), and TiO_2 (~11

FIGURE 2. (**a**) Spherule-laden, carbonated kimberlite dike at Mukorob intruded into Nama Group Cambrian sediments. Note the pinch and swell structure to the dike and the parallel jointing in sediments at the baked contact. The dike trend is NW-SE. Lens cap diameter is 5 cm. (**b**) Dislodged block of spherule-laden ilmenite bodies in the Mukorob kimberlite dike. Note that the spherules are matrix supported and that no flow pattern is apparent.

TABLE 1. EMPA of ilmenite, spinel, and phlogopite in oxide orbicules

	Locations																	
	Mukorob								Hatzium									
	Ilmenite				Spinel				Spinel					Phlogopite				
wt%	ilm rim (spin)	ilm outer band	ilm blebs	ilm inner band	spinel rim	spinel core	spin core (ilm)	spinel gmass	spin outer band	spinel blebs	spin inner band	spinel in gmass	wt%	phlog in sp	phlog in sp	phlog in gmass	phlog in gmass
Nb_2O_5	nd	0.26	0.25	0.12	nd	nd	0.13	0.01	0.09	0.05	nd	nd	SiO_2	29.56	34.01	30.09	34.03
SiO_2	0.02	nd	0.01	0.03	0.03	0.23	0.06	0.03	0.15	0.31	0.13	0.23	TiO_2	1.82	0.88	1.92	1.53
TiO_2	52.67	53.86	53.62	53.41	17.26	11.38	1.13	18.05	11.00	10.38	11.62	11.66	Al_2O_3	17.81	17.19	17.08	17.21
ZrO_2	nd	0.06	0.04	nd	nd	nd	nd	0.05	0.08	0.15	0.50	0.00	Cr_2O_3	0.00	0.02	0.00	0.00
Al_2O_3	nd	nd	nd	nd	4.34	13.20	7.12	4.49	11.57	9.65	10.38	7.28	MgO	24.08	24.12	22.80	24.13
Cr_2O_3	3.28	2.13	2.00	1.98	0.50	0.01	54.13	0.58	nd	nd	nd	0.04	CaO	0.31	0.12	0.06	0.01
MgO	12.10	11.72	11.25	11.28	14.07	15.50	11.18	11.20	16.82	15.61	15.82	15.57	MnO	0.04	0.08	0.02	0.10
CaO	0.09	0.08	0.19	0.21	0.72	0.91	0.02	0.51	0.06	0.12	0.03	0.12	FeO	2.65	3.05	3.09	3.40
MnO	0.25	0.24	0.23	0.36	0.42	0.57	nd	0.41	0.64	0.95	0.69	0.71	BaO	10.04	5.35	8.67	5.85
FeO	25.47	27.51	28.00	27.45	29.31	18.78	17.24	30.43	17.06	17.93	18.91	18.69	Na_2O	0.06	0.07	0.08	0.05
Fe_2O_3	6.75	3.57	4.96	5.80	35.55	39.62	9.34	34.20	42.76	45.28	41.68	45.66	K_2O	6.77	8.10	7.27	8.55
Total:	100.62	99.43	100.53	100.64	99.19	100.20	100.37	99.96	100.23	100.42	99.76	99.90	Total:	93.16	92.88	91.07	94.85
	2 cats/3oxy				**24 cats/32oxy**				**24 cats/32oxy**					**Based on 22 oxy**			
Nb	nd	0.003	0.003	0.001	nd	nd	0.016	0.002	0.010	0.006	nd	nd	Si	4.641	5.132	4.796	5.076
Si	0.000	nd	0.000	0.001	0.007	0.059	0.015	0.010	0.038	0.080	0.034	0.060	Al	3.296	2.868	3.204	2.924
Ti	0.911	0.944	0.934	0.929	3.563	2.175	0.226	3.696	2.106	2.018	2.262	2.301	total	7.936	8.000	8.000	8.000
Zr	nd	0.001	0.000	nd	nd	nd	nd	0.006	0.010	0.019	0.062	0.000	Al	0.000	0.019	0.005	0.102
Al	nd	nd	nd	nd	1.404	3.952	2.230	1.442	3.473	2.940	3.167	2.250	Ti	0.215	0.099	0.230	0.172
Cr	0.060	0.039	0.037	0.036	0.109	0.002	11.372	0.124	nd	nd	nd	0.009	Cr	0.001	0.003	0.000	0.000
Mg	0.415	0.407	0.388	0.389	4.531	5.871	4.434	4.543	6.385	6.016	6.104	6.066	Mg	5.633	5.427	5.416	5.366
Ca	0.002	0.002	0.005	0.005	0.211	0.248	0.006	0.148	0.018	0.032	0.010	0.033	Mn	0.005	0.010	0.003	0.012
Mn	0.005	0.005	0.004	0.007	0.098	0.123	nd	0.096	0.138	0.205	0.151	0.158	Fe	0.348	0.385	0.411	0.424
Fe^{3+}	0.117	0.063	0.086	0.101	7.346	7.576	1.868	7.006	8.192	8.807	8.118	9.018	VI total	6.202	6.113	6.066	6.075
Fe^{2+}	0.490	0.536	0.542	0.531	6.731	3.993	3.831	6.929	3.633	3.877	4.092	4.103	Ca	0.052	0.020	0.011	0.002
Total	2.000	2.000	2.000	2.000	24.000	24.000	24.000	24.000	24.000	24.000	24.000	24.000	Ba	0.618	0.316	0.542	0.342
													Na	0.020	0.021	0.023	0.013
													K	1.356	1.559	1.478	1.627
													A total	2.045	1.916	2.054	1.984

Note: Each oxide column is the average of at least 10 virtually identical analyses; each phlogopite column is a selected single analysis because of extreme zoning. nd = not detected.

wt%), and is classified as an MAT spinel, or more completely as an MAT magnetite; gray spinel is a magnesian chromite with magnetite in solid solution (Table 1, Figs. 6a and 6b), typical of xenocrysts in kimberlites (Haggerty 1991). Spinel overgrowths on brown spinel have lower MgO (14 wt%) and Al_2O_3 (~4 wt%) contents and higher concentrations of TiO_2 (~17 wt%). These compositions are indistinguishable from brown spinel in the groundmass (Table 1, Figs. 6a and 6b), and from some kimberlites (Haggerty 1991). Ilmenite rims on gray spinel are slightly enriched in MgO, Cr_2O_3 (by about 1 wt%), and Fe_2O_3 (by 1–3 wt%), relative to ilmenite in polycrystalline bands and discrete blebs (Table 1). Both spinel (Fig. 6a) and ilmenite parageneses and mineral compositions are remarkably similar to those in the carbonate-rich kimberlite of the Benfontein Sills in Kimberley, South Africa (McMahon and Haggerty 1984; Gaspar and Wyllie 1984; Jones and Wyllie 1985).

Calcite compositions (expressed as oxides) in the dike have 0.2 wt% MgO, ~1 wt% MnO, and ~3.5 wt% FeO. This composition is similar to skeletal, magmatic calcite in the Benfontein Kimberlite Sills, but very different from secondary vein calcite with minor elements at or below the EMPA detection limit (<0.1 wt%).

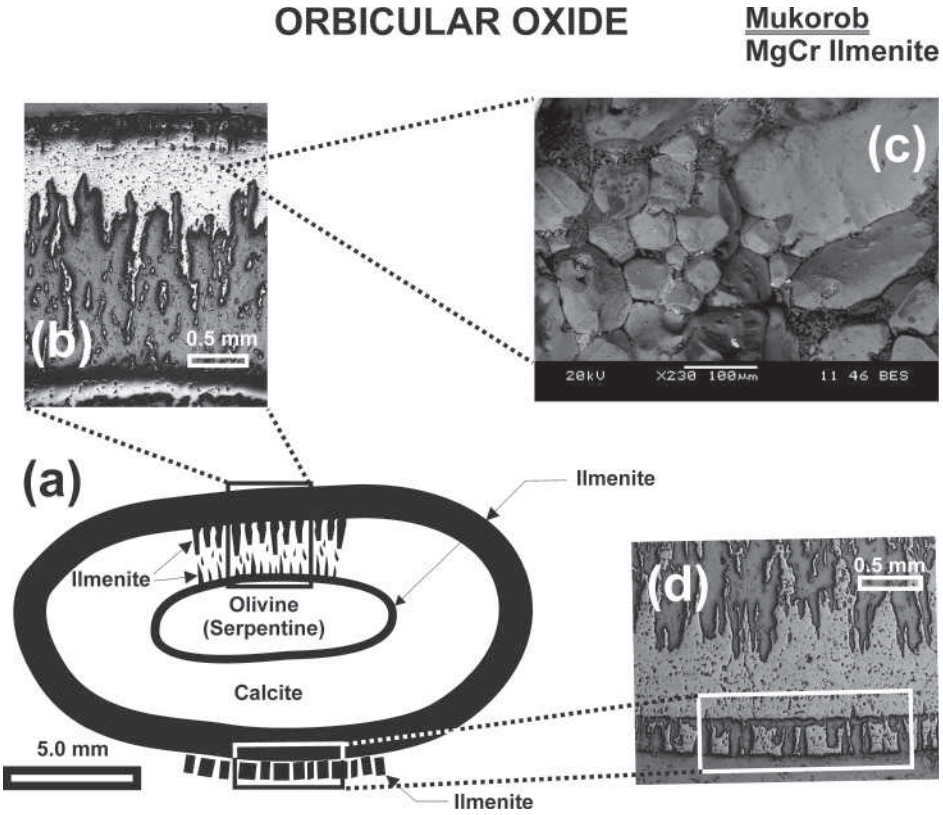

FIGURE 3. (**a**) Schematic representation of the most prominent textural features in spherule ilmenite assemblages from the Mukorob kimberlite dike. (**b**) Inner and outer bands of concentric ilmenite linked by pedestal ilmenite in a matrix of granular calcite; the outer band is polycrystalline and shows stress twinning (reflected light). (**c**) SEM image of a fractured surface of outer band ilmenite. (**d**) Extreme edge of a typical spherule showing dentate ilmenite in mosaic calcite.

Magnetite pellets at Hatzium

The Hatzium kimberlite intrusion (~100 m in diameter) is highly brecciated, and contains abundant country rock fragments. The area of specific interest, although poorly exposed and crumbly, is a ~2 m wide carbonatite dike on the SE section of the pipe (Janse 1975), where parallel veins (~20 cm wide) of barite also occur. The carbonatite facies has abundant black, magnetic spherules set in a macroscopic matrix of calcite, olivine, and magnetite, and microscopic phlogopite + apatite with minor barite. This is a classic phoscorite (Wall and Zaitsev 2004), typically associated with carbonatites, but not previously recorded in spatial and temporal proximity to kimberlite.

The spheres (2–3 cm diameter) have cores of olivine altered to serpentine, serpentine + calcite, and serpentine + calcite + magnetite (Figs. 7–8). Some cores have radial, spoke-like cracks (typical of syneresis), filled with calcite and serpentine; these fractures terminate abruptly on contact with the orderly, oxide-rich perimeter bands that follow (Figs. 8a–8c). The bands are rhythmically layered with alternating wider (1 mm), and thinner (~0.25 mm) concentric rings of magnetite (Figs. 7–8), nucleated on and dispersed within a complex mixture of calcite + serpentine. The oscillatory thicker and thinner bands are referred to here as *annular* and *diurnal*, respectively, in allusion to the banding in corals, but no time dependence is implied. Although rhythmic in individual spheres, there is some variation between and among spheres, as there is between core diameters, and core assemblages (Figs. 8a–8i). Oxide precipitation, with few exceptions, was sparse at the outset of nucleation (Figs. 8a–8c), continued rhythmically (Figs. 8a–8c, 8g–8h), but was also interrupted locally by the preferred crystallization of calcite + olivine (Figs. 8d–8f).

The groundmass is dominated by serpentine, calcite, and magnetite; neither ilmenite nor perovskite were identified. Apatite needles in calcite are developed prominently, as are euhedral to subhedral laths of phlogopite. The mica is kink-banded, is splayed by penetrative calcite, and contains inclusions of magnetite.

Mineral chemistry

Spinel in *annular* and *diurnal* bands, and in dispersed blebs, has a remarkably uniform composition characterized by significant contents of MgO (16–17 wt%), Al_2O_3 (7–11 wt%), and TiO_2 (10–11 wt%) and is classified as MAT-magnetite (Table 1, Figs. 9a–b). Although MAT-magnetite is a major constituent in the Hatzium spheres and a minor mineral in the Mukorob groundmass, both are similar in composition, with correspondingly close affinities to calcite-rich kimberlites and kimberlitic carbonatites as discussed above.

Calcite (~1 wt% FeO), serpentine (~4 wt% FeO, 0.5–1 wt%

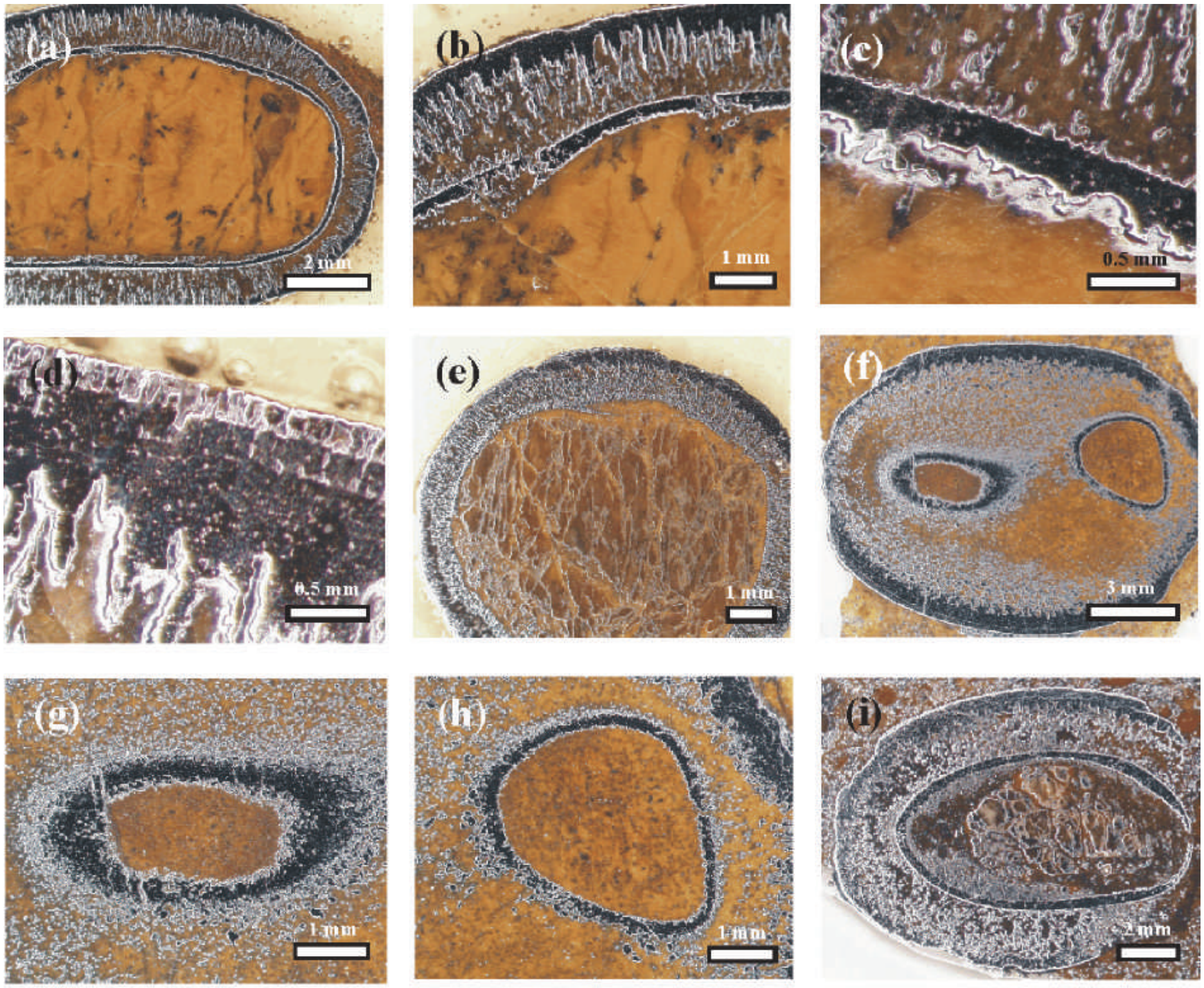

FIGURE 4. Examples of ilmenite-olivine (serpentine) orbicular bodies from the Mukorob carbonated kimberlite dike. The cores are dominated by rounded, probable xenocrysts of olivine (**a–e**) on which ilmenite, and ilmenite + calcite has crystallized. The jagged texture of ilmenite in (**b**) and (**d**) is due in part to precipitation on skeletal calcite. Some spherules have multiple cores of granular calcite (**f** with details in **g** and **h**) on which ilmenite + calcite has nucleated, both internally as well as at the external boundary; the dark haloes are aggregates of polycrystalline ilmenite + calcite. In **i**, the core is serpentine + calcite with successive concentric bands of ilmenite, ultra-fine ilmenite + calcite, coarser ilmenite + calcite, and finally polycrystalline ilmenite. Stereomicroscope images of polished thick sections in plane light. See Figure 3 and text for textural details in reflected light.

Al_2O_3), and apatite (~2 wt% F) have fairly uniform compositions. These are in stark contrast to phlogopite, which is spectacularly zoned (mainly in Ba, Si, and K), in irregular patches and in strips parallel to cleavage. Barium is most distinctive and phlogopite compositions range from 2–10 wt% BaO, whether in the groundmass, or as inclusions in spinel (Table 1).

Phlogopite compositions are compared with a selection of other barian and barian-titanian phlogopite from various undersaturated rocks in oxide and cation plots in Figures 10a–10c. These include Ba-Ti biotite in nephelinites from Hawaii (Mansker et al. 1979) and West Eifel, Germany (Edgar 1992), leucitites from Antarctica (Sheraton and Cundari 1980), alkali basalt from Italy (Thompson 1977), kimberlite from South Africa (Field et al. 1989), lamproite from Australia (Scott-Smith and Skinner 1984), and carbonatite from Brazil (Gaspar and Wyllie 1982, 1987). The spectrum of compositions covers Ba-phlogopite in ultramafic alkali rocks reviewed comprehensively in Mitchell (1995). The composition of the Hatzium phlogopite most closely resembles that of phlogopite in the Jacupiranga carbonatite (Figs. 10a and 10b), in the types of solid solution in large cations (Ba-K, Fig. 10b), and in coupled substitution $(Ba + 2Ti + 3Al) \leftrightarrow K + 3(Mg + Fe) + 3Si$ (Fig. 10c).

DISCUSSION

Excellent reviews of orbicular structures cover the compositional spectrum from felsic intrusions (Leveson 1966; Elliston 1985) to alkali ultramafic bodies (Wall and Zaitsev 2004) to sedimentary ooids (Simone 1981). Spheres in relation to minimum energies of formation are a common theme, but beyond that, models inducing precipitation that lead to orbicular bodies are notably varied. Of igneous interest is the crystallization of precursor, sedimentary colloidal hydrosilicates (Elliston 1985),

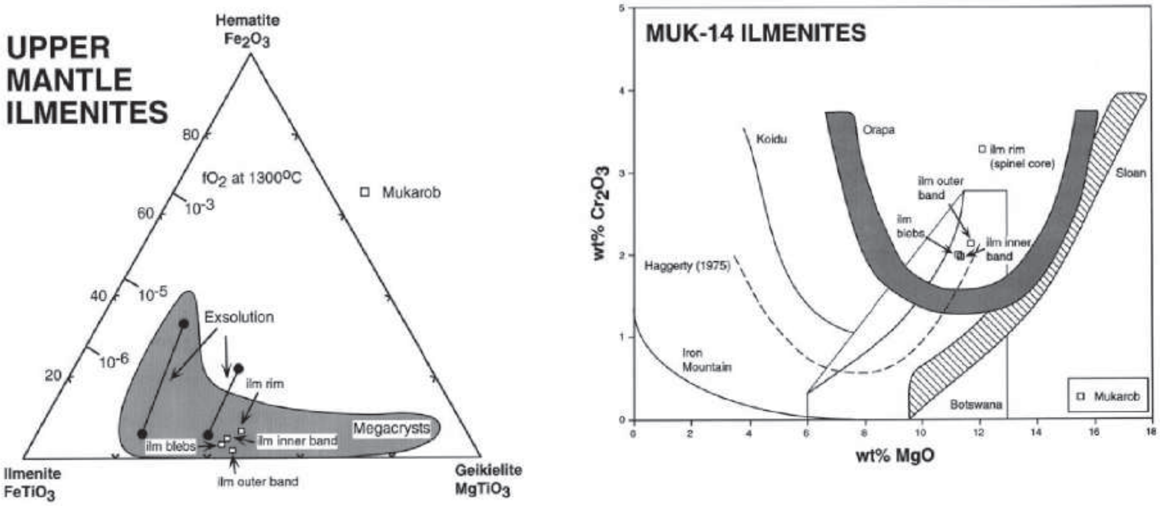

FIGURE 5. (**a**) Ternary diagram (mol%) for ilmenite compositions from Mukorob (Table 1). The mineral data plot well within the main compositional field of megacrystic upper-mantle ilmenite. Exsolution pairs are from the carbonated Koidu Kimberlite Complex, Sierra Leone. Oxygen fugacity estimates at 1300 °C, and the ilmenite megacryst field are from Haggerty (1991). (**b**) Ilmenite oxide compositions are from Mukorob. The data are typically kimberlitic, falling on the MgO-enriched side of the parabolic curves. Adapted from Haggerty (1991).

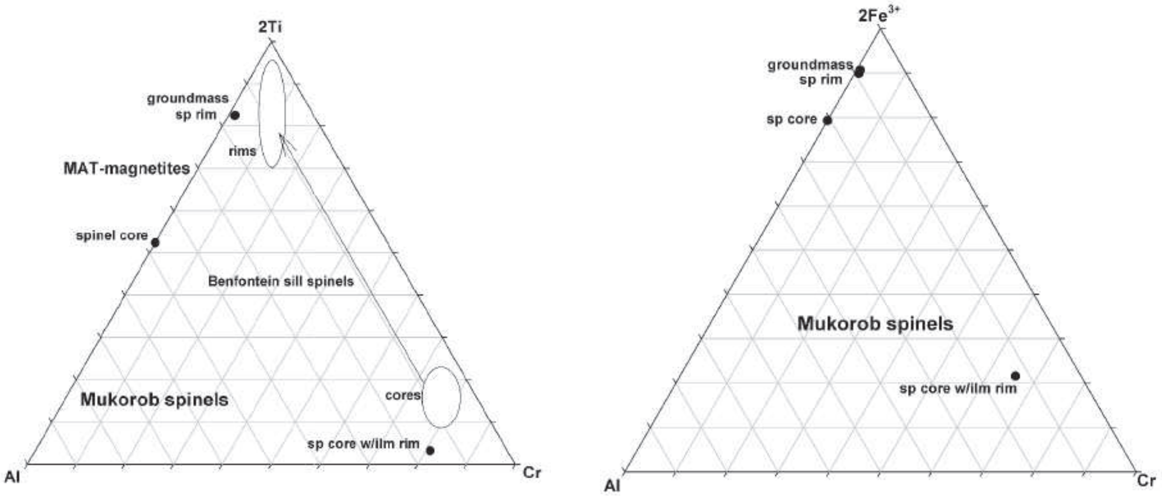

FIGURE 6. (**a–b**) Ternary diagrams showing the distribution of spinel compositions (in at%) in the Mukorob carbonatitic kimberlite. Benfontein Sill data are from McMahon and Haggerty (1984).

crystallization by feedback between concentration fronts and mineral reaction surfaces (Wang and Merino 1993), superheated magma (Vernon 1985), liquid immiscibility (Moore and Lockwood 1973; Lapin and Vartiainen 1983), and fluidization (Sutherland 1980).

Ilmenite spheroidal bodies, with cores of silicate + carbonate, have not been recognized outside of the Mukorob occurrence, in Namibia or elsewhere in carbonatite complexes. The closest in overall, but not detailed appearance, is rounded olivine (~10 μm in diameter) with spinel + perovskite overgrowths in rocks with kimberlite affinities from the Igwisi Hills, Tanzania (Reid et al. 1975); in kimberlites in NE Finland where olivines are overgrown by monticellite + layers of chromite, pleonaste, and titanomagnetite (O'Brien and Tyni 1999), and in pelletal lapilli from kimberlites and melilitites in Southern Africa where cores are typically olivine and overgrown mantles are phlogopite, spinel, perovskite, and olivine (Mitchell 1995). Bodies similar to, but not identical with, the Hatzium magnetite spheres have been described in carbonatites from Uganda (Sutherland 1980), Germany (Keller 1981), and Finland and Russia (Lapin and Vartiainen 1983; Krasnova et al. 2004). Although the mineral assemblage of magnetite + calcite + apatite is common to the last named localities, the internal textures differ from the concentric bands observed in the Hatzium dike (Figs. 7–8). In the magnetite dominant spheres from Sokli, Vuorijarvi, and Kovdor, the oxides are radial to the core; concentrically zoned spheres contain magnetite + calcite but are always accompanied by phlogopite. Examples from Uganda have inwardly protruding, triangular blades of magnetite in hosts of calcite, whereas the Rhine Graben lapilli have disseminated magnetite + apatite in a

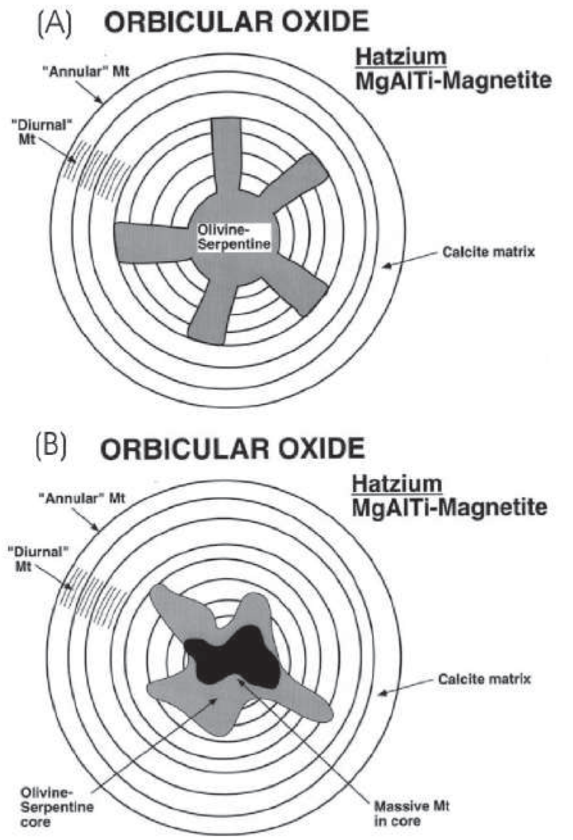

FIGURE 7. **(a–b)** Typical concentric textures and applied nomenclature are for MAT spinel in the Hatzium carbonatite dike. Core mineralogy is variable with other examples illustrated in Figure 8.

trachytic-textured host of calcite.

We consider the proposal by Lapin and Vartiainen (1983) to have most of the attributes necessary to model an origin for the Namibian orbicule structures. Firstly, we adopt their premise of liquid immiscibility (i.e., separation of a parent magma into two or more liquids), although mixing of two or more magmatic liquids (Zaitsev and Bell 1995) would be difficult to distinguish and cannot be ruled out. Magma mingling is possibly more appropriate for the two highly disparate liquids envisaged, namely a low-silica carbonatite and an ultramafic kimberlite. Kimberlites typically form at greater depths (>200 km) than carbonatites (60–80 km) and magma mingling is easy to envisage in a rising kimberlite. Immiscibility or emulsification of a carbonate-rich melt from a single magma could possibly occur at depths as shallow as 50 km (Pyle and Haggerty 1994), with constituent separation being triggered by the melt on P-T intersection with the C-H-O peridotite solidus (Haggerty 1989). Experimental work on the melting of eclogite + CO_2 (Dasgupta et al. 2006) more closely approximates the model we envisage. They show that immiscible carbonate and silicate liquids coexist between 1200–1400 °C at 3 GPa. In addition, the carbonatitic melts coexist with ilmenite (plus residual garnet and clinopyroxene) between 1000–1175 °C. The essential feature in our model is the separation of an ore-carbonate (magnetite or ilmenite) liquid from a carbonate + silica melt; the former is volumetrically smaller and in spherical droplets (minimum energy), and has a higher density and higher viscosity relative to its fluidal carbonate host. From ilmenite compositions, which are classically upper mantle in origin (Figs. 5a–b), we propose that the parental liquid at Mukorob was a carbonatitic kimberlite, whereas at Hatzium the liquid was close to a classic carbonatite. The former was Ti-enriched (ilmenite) and relatively reduced, the latter Fe-rich (magnetite) and moderately oxidized. The protoliths were possibly carbonated eclogite at Mukorob (Dasgupta et al. 2006), and carbonated peridotite at Hatzium (e.g., Presnell and Gudfinnsson 2005). On immiscible fractionation, the carbonate liquid became enriched in P, further assisting immiscibility (Phillpots 1967), and in the case of Namibia, also enriched in elemental constituents (K, Ba, Al, Mg, Si) essential to the crystallization of phlogopite of unusual composition (with up to 10 wt% BaO). With contrasts in density, gravitational separation was possible, and we suggest was necessary to account for the oblate spheroidal forms (not the cores but the overgrowths) in the ilmenite nuggets from Mukorob (Fig. 4). Stacks of flattened spheres, with carbonate liquid largely expelled by filter pressing, are suggested in the bodies from Finland. These bodies were rigid but not solid, with exterior rims that were highly polymerized.

This brings us to the second important point in the Lapin and Vartiainen (1983) model, which is that crystallization of the spherical bodies they described was initiated at the margin of spheres. Crystallization along a thermal gradient alleviates the difficult problem of having to form layers within spheres (Figs. 4 and 8) by diffusion and accretion. The thickness and compositions of layers are remarkably uniform for ilmenite in Mukorob, and less so within the oscillatory concentric zones of magnetite in Hatzium, but both imply that rates of crystallization were buffered externally. This not withstanding, we differ in the interpretation of the direction of crystallization, which seems justified in the examples studied by Lapin and Vartiainen (1983), but not in the samples from Namibia. Whereas entire spheres of ore + calcite are deemed immiscible liquids, it was nucleation of these melts on cores of olivine (Mukorob) or olivine + massive magnetite (Hatzium) that took place. The case is clearly defined texturally and mineralogically in the assemblage from Mukorob where olivine could not possibly have been a liquidus phase; but we do suggest that the subsequent mantles of successive overgrowths of pedestal and layered ilmenite (Figs. 3 and 4) or magnetite were from an immiscible ore-carbonate liquid. The ilmenite pedestals crystallized outwardly from the core, and inwardly from the groundmass (Fig. 3); magnetite by contrast was sequentially deposited. Phosphorous (Phipotts 1967) and C (Weidner 1982) were critical to the lowering of oxide liquidii to geologically reasonably temperatures of 900–1200 °C or less. This range would correspond to 2–3 GPa (60–90 km) if the C-H-O peridotite solidus is invoked, as suggested above.

There was little variation in the bulk chemistry of the ore-silicate-carbonate liquid at Mukorob. This is reflected in the similarity of minerals and mineral compositions, and minor variations in modal proportions (Fig. 4). The olivine cores are mafic and possibly of harzburgitic or dunitic ($Fo_{90–92}$) xenocrystic parentage; the mantling overgrowths are Ca-Fe-Ti and carbonate-rich. This is a typical late-stage composition in kimberlites

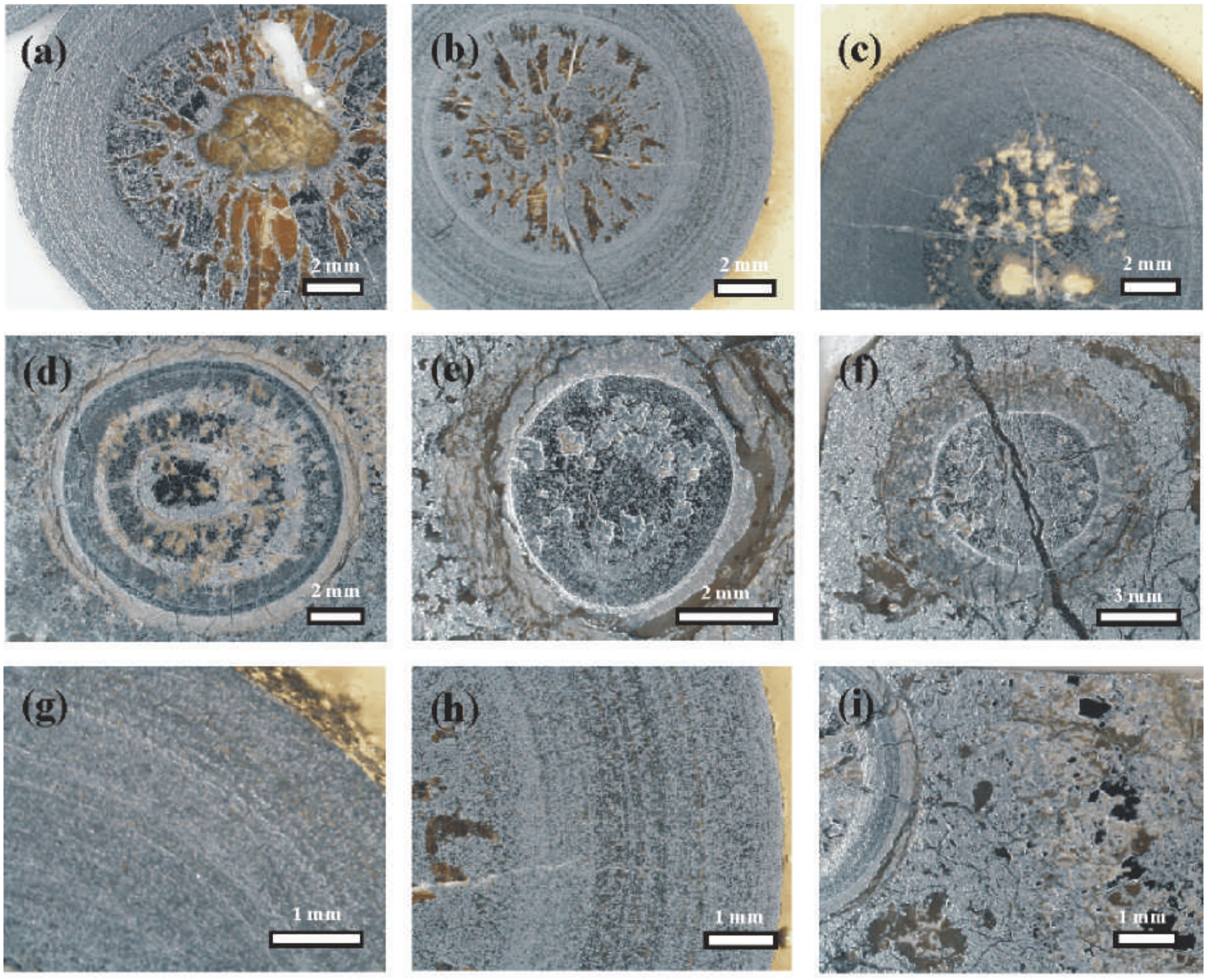

FIGURE 8. MAT = spinel spheres from the Hatzium carbonatite dike. Core assemblages are serpentine (**a–b**), serpentine + calcite (**c**), and serpentine + spinel + calcite (**d–f**). Details of the *"annular"* and *"diurnal"* (Figs. 6a–b) textural designations (**g–h**), and disruption of these concentric bands (**i**), show that quiescent precipitation and possible frothing took place during spherule formation. Stereomicroscopic photomicrographs of polished (**a–c**), and polished-thin sections (**d–f**) are in plain light.

where groundmass crystallization is perovskite + titanomagnetite or rutile (e.g., Haggerty 1991 and references therein), rather than the spherule assemblage of ilmenite + calcite. Some drops encapsulated larger amounts of carbonate liquid than equilibrium immiscibility permitted (Figs. 4f–4h). These internal bodies mimic the sequence of crystallization of their spherical hosts. The oblate spheroidal bodies are dense and may have settled into a gravitationally induced ore-carbonate layer (as suggested above). This represented a distinct magmatic episode that was subsequently sampled by a later pulse of erupting carbonatitic kimberlite.

At Hatzium, the liquids show a range in composition (Figs. 8d–8f), crystallization was punctuated between core and rim assemblages, and there is evidence for volatile loss from metastable colloidal gels (syneresis), prior to the onset of main-stage, magnetite crystallization (Figs. 8a–8c). These observations imply

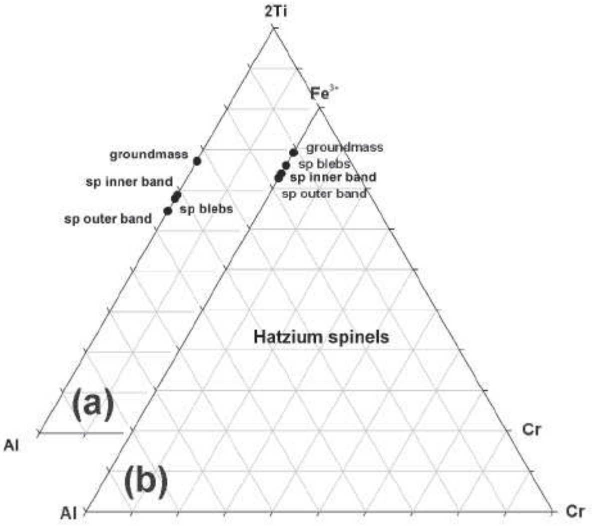

FIGURE 9. **(a–b)** Ternary diagrams showing the distribution (at%) of MAT-spinel compositions in spherules from the Hatzium carbonatite.

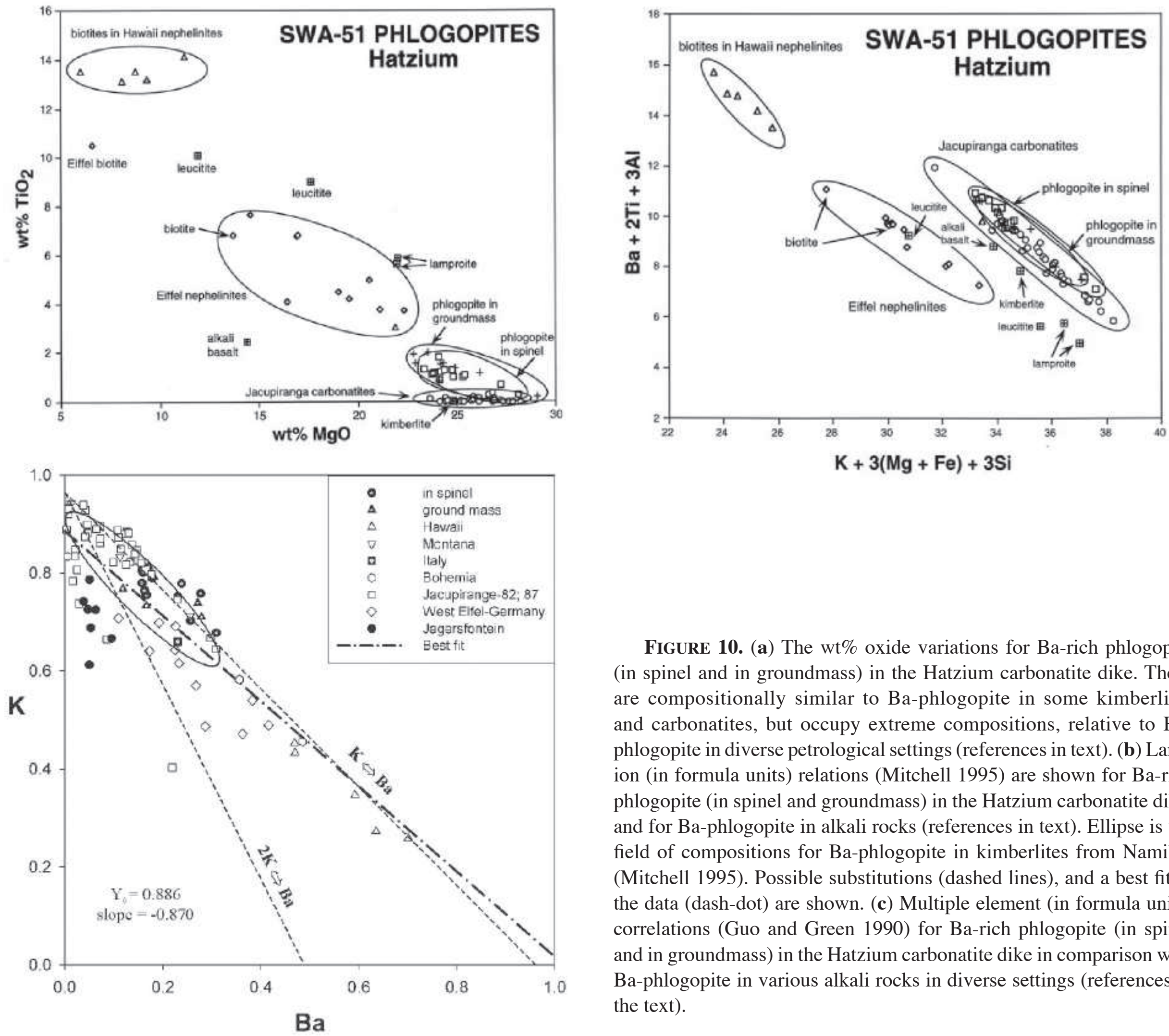

FIGURE 10. (**a**) The wt% oxide variations for Ba-rich phlogopite (in spinel and in groundmass) in the Hatzium carbonatite dike. These are compositionally similar to Ba-phlogopite in some kimberlites and carbonatites, but occupy extreme compositions, relative to Ba-phlogopite in diverse petrological settings (references in text). (**b**) Large ion (in formula units) relations (Mitchell 1995) are shown for Ba-rich phlogopite (in spinel and groundmass) in the Hatzium carbonatite dike, and for Ba-phlogopite in alkali rocks (references in text). Ellipse is the field of compositions for Ba-phlogopite in kimberlites from Namibia (Mitchell 1995). Possible substitutions (dashed lines), and a best fit to the data (dash-dot) are shown. (**c**) Multiple element (in formula units) correlations (Guo and Green 1990) for Ba-rich phlogopite (in spinel and in groundmass) in the Hatzium carbonatite dike in comparison with Ba-phlogopite in various alkali rocks in diverse settings (references in the text).

that the ore-carbonate liquid was modified modally possibly by reaction with its carbonatite host, clearly under non-equilibrium conditions because not all droplets were affected equally; MAT-spinel, however, whether in inner or outer bands, in core blebs, or the groundmass settings is remarkably similar in composition (Table 1, Figs. 9a–b).

That both localities in Namibia are dikes may be significant to the formation of immiscible liquids and spherical oxide bodies. There is no evidence for explosive activity, notwithstanding the fact that the magmas were hot (baked sedimentary contacts, Fig. 2a) and volatile-charged. But there is evidence for episodic mineral growth, volatile loss, and renewed crystallization. So we suggest that quiescent gas diffusion, with intermittent fluidization (gas expansion and droplet formation in liquids), through country rock cracks, prior to dike intrusion, was essential to droplet formation, nucleation, and growth of these exotic bodies.

CONCLUDING REMARKS

Although magnetite + olivine + calcite spheroidal bodies are known in carbonatites from several localities, worldwide, the objects from Namibia in our study show several important distinctions: ilmenite is currently known only from Mukorob; and is kimberlite-related; concentric layering of magnetite + calcite at Hatzium differs texturally from other classic carbonatite localities; both contain fluorapatite making these typical of phoscorites in carbonatites; phlogopite in Hatzium is enriched in Ba; both Mukorob and Hatzium are dikes, closely related to non-diamond-bearing kimberlite pipes; and both contain minor barite.

Ilmenite compositions are typical of ilmenite megacrysts in kimberlite, whereas spinel is MgO-Al_2O_3-TiO_2 (MAT)-enriched magnetite, similar to spinel in the carbonate-rich kimberlite sills of Benfontein, Kimberley, South Africa. The f_{O_2} of ilmenite crystallization was relatively reduced (10^{-8} bars at 1300 °C ~ FMQ-1), in contrast to MAT-magnetite that formed at moderately higher oxidation states (FMQ-MH).

Our model for the orbicular oxide bodies invokes liquid immiscibility of a carbonatitic kimberlite at Mukorob, and of a typical carbonatite at Hatzium. The protoliths were, respectively, carbonated eclogite and carbonate peridotite that underwent mantle melting at 2–3 GPa and 1000–1200 °C. Phase separation produced spherical, ore-carbonate liquid droplets of high density and viscosity in a magmatic fluid media of carbonate + silica. Immiscibility was enhanced by the fractionation of phosphorous + carbon into liquid carbonate. Gravity settling and compaction of the ilmenite-bearing bodies at Mukorob led to soft-shell deformation of the exterior overgrowths. This ore-silicate-carbonate cumulate was sampled, transported, and erupted in a carbonate-rich kimberlite. Other aspects of the model call for punctuated crystallization from the margins of spheres to core interiors, periodic frothing (fluidization), and slow degassing through dike systems.

Is it possible that some hematite Martian "blueberries" may be oxidized magnetite spherules from immiscible liquids that formed in the planet's interior? If not on Mars, Venus is surely a likely candidate for carbonatites and immiscible oxide orbicules.

ACKNOWLEDGMENTS

We appreciate the opportunity, at the invitation of the Associate Editors, to contribute in a small way to this volume. In recognition of Jim Papike's major contributions to the Earth and Planetary Sciences, and to his latest, innovative excursion into the realm of noble oxides, we dedicate our paper. In a career devoted to understanding the heralding call: "Don't deserve to play hard unless you work hard," all would agree that Jim has personified the ethic, and continues to do so. This splendid volume by friends and admirers pales in comparison to the volumes of wonderful tales, all true, that deserve to be told in a complete celebration of Jim's contribution to science, his achievements, and his endurance on the planetary playing field. As "quick-chip" Papike of lunar sample fame, we say…Keep it up, mate!

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