# Olivine: a monitor of magma evolutionary paths in kimberlites and olivine melilitites

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Abstract. Petrographic and chemical criteria indicate that the overwhelming majority of olivines in kimberlites are probably cognate phenocrysts. The implied low volume of xenocryst olivines requires that primitive kimberlite magmas are highly ultrabasic liquids. Two chemically distinctive olivine populations are present in all of the kimberlites studied. The dominant olivine population, which includes large rounded olivines and smaller euhedral crystals, is Mg-rich relative to late-stage rim compositions. It is characterized by a range in 100 Mg/(Mg + Fe) and uniform Ni concentration, reflecting Rayleigh-type crystallization during magma evolution. The most Mg-rich of these olivines are considered to be similiar to those in the mantle source rocks. The second compositional population, generally very subordinate, though markedly more abundant in the megacrystrich Monastery kimberlite, is Fe-rich relative to rim compositions. This group of olivines crystallized from evolved liquids in equilibrium with iron-rich megacrysts, both entrained by the kimberlite magma during ascent. Differences between the chemical fields of Fe-rich olivines in Group I and Group II kimberlites point to relatively deeper derivation of the latter suite. Olivine chemistry can be used to characterize kimberlite magma sub-types, and may prove to be a useful tool for evaluating the diamond potential of kimberlites.

# Introduction

Olivines characterized by a wide compositional range invariably dominate phenocryst assemblages in kimberlites (Boyd and Clement 1977; Moore 1986). An understanding of the origin of these olivines is therefore critical in developing models for the genesis of kimberlite magmas. It is particularly important to establish whether a significant proportion of olivines are xenocrysts (c.f. Skinner and Clement 1979; Bailey 1984), or cognate phenocrysts (c.f. Moore 1986), as this question has bearing on the composition of primitive kimberlite melts. Further, compositional variation of kimberlitic olivines would be expected to provide a sensitive monitor of chemical evolutionary paths followed by the host magma.

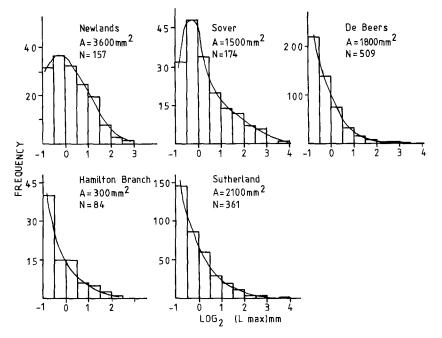
In an attempt to place constraints on the possible origins of kimberlitic olivines, a detailed petrographic study has been made on nine southern African kimberlites and two from Kentucky, USA (listed in Table 1). This was followed by a study of olivine compositional variation in seven of the South African kimberlites and both from Kentucky. The kimberlites from southern Africa for which chemical data data on olivine are available represent an essentially east-west transect across the sub-continent (Letseng-la-terai and Monastery in the east, de Beers, Sover, Bellsbank and Newlands in the centre, and the K35 kimberlite of the Gibeon cluster in the west).

The Sover, Bellsbank and Newlands kimberlites belong to the isotopically distinctive Group II variety recognized by Smith (1983). The remaining kimberlites studied belong to the Group I suite. Smith et al. (1985) suggest that the latter can be divided on bulk rock chemical criteria into two subgroups. The IA subgroup includes de Beers, Jagersfontein and probably also Letseng-la-terai, all located on the Archaean craton [data for Letseng-la-terai from Gurney and Ebrahim (1973)]. The Monastery kimberlite, which is also on the craton, is classified in the IB subgroup by Smith et al., largely on the basis of unusually high TiO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub>. However, with respect to most other components, this kimberlite is strikingly closer to the average composition of the IA sub-group. As the unusually high  $TiO_2$  and  $Fe_2O_3$  contents must in part reflect the particularly high proportion of ilmenite macrocrysts characteristic of this pipe (Smith et al. 1985), it is possible that the Monastery magma had closer affinities to the IA sub-group. Additional

Table 1. Localities of kimberlites studied

tes
Gibeon field
(approx. 25°30′S/17°45′ E)
Elliot County, Kentucky, USA
Elliot County, Kentucky, USA
kimberlites
28°44' S/24°45' E
28°44′ S/25°25′ E
29°02′ S/28°53′ E
28°50′ S/27°30′ E
28°04′ S/24°24′ E
28°22′ S/24°24′ E
28°14′ S/24°28′ E

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factors bearing on this question are discussed in a later section. Bulk rock chemical data are not available for the Gibeon and Kentucky kimberlites. However, their tectonic setting (off-craton) indicates that they are probably IB varieties.

In view of the uncertainty regarding subdivision of the Group I kimberlites studied, these have, for the purpose of this work, been arbitrarily sub-divided into diamondiferous Group I (de Beers, Jagersfontein, Letseng-la-terai and Monastery) and barren Group I (Gibeon and Kentucky pipes).

A close genetic link between kimberlites and olivine melilitites has been proposed by several authors (e.g. Taljaard 1936; McIver 1981). The petrographic and chemical data for kimberlitic olivines have therefore been compared with available and hitherto unpublished data for olivines from South African olivine melilitites.

#### Petrography

In the kimberlite specimens studied, olivine is characteristically present as discrete grains (i.e. lacks internal sub-grain boundaries). There is always a considerable range in relative size, from large, generally well-rounded grains, some in excess of 10 mm (macrocrysts), to smaller crystals, which are commonly idiomorphic, or show relic crystal outline. The tendency for the larger grains to be rounded and the smaller individuals to possess some degree of crystal outline superficially gives the impression of two discrete olivine size populations. This has been tested by measuring the longest axes of all olivine grains greater than 0.5 mm in all of the available thin sections of kimberlites from each of the three groups recognized in this study. Olivine size distribution data are summarized in Fig. 1a-d. In each of the samples studied, the olivines show a smooth continuous variation in size rather than distinct bimodal patterns. However, there are systematic differences between Group I and II kimberlites. The class frequencies in the former increases continuously to smaller crystal sizes while in the latter there is a modal maximum at 0.7 mm. Olivine size distribution in the Monastery kimberlite may differ from the patterns described above, as there are a significant number of grains which appear markedly larger than the rest of the phenocryst population. However, size distribution in this pipe could not be quantified because of strong marginal alteration of the olivines.

Fig. 1. Size frequency distribution of greatest dimensions (L max) (mm) of all olivines greater than 0.5 mm in kimberlites and the Sutherland olivine melilitite. Newlands and Sover are Group II kimberlites. De Beers is a diamondiferous Group IA kimberlite. Hamilton Branch is non-diamondiferous, and inferred to be a Group IB variety. A = Total area of thin sections studied. N=total number of olivines greater than 0.5 mm in area A. All samples are characterized by a smooth continuous size distribution in the size range studied. Attention is drawn to the frequency maximum at 0.7 mm for Group II kimberlites. This contrasts with the continuous increase in frequency towards smaller sizes in the Group I kimberlites and the olivine melilitite. None of the samples shows evidence of a polymodal size distribution

Olivines with undulose extinction are present in all of the diamondiferous Group I kimberlites studied, although there are differences in their proportions and in the degree of deformation. In the Letseng-la-terai, Jagersfontein and Monastery kimberlites, some of the macrocrysts (grains greater than 10 mm and generally rounded) show slight undulose extinction, although the majority lack optical strain features. The smaller euhedral crystals are characteristically strain-free in these samples. However, undulose extinction, often pronounced, is a characteristic feature of a majority of macrocrysts and also a significant proportion of the small idiomorphic grains in the de Beers kimberlite. In the deformed idiomorphic grains, bands of undulose extinction traverse core and rim of the crystal.

Both the de Beers and Letseng-la-terai kimberlites are characterized by the presence of rare small clasts composed of olivine neoblasts forming a mosaic texture. The outline of some of these mosaic-textured clasts suggests that they represent formerly euhedral grains that have been recrystallized. Small mosaic-textured selvages occur on the edges of some of the larger olivines in the de Beers kimberlite. Mosaic textures olivines appear to be absent in the Monastery kimberlite, and are very uncommon in the Jagersfontein sample studied.

Petrographic characteristics of olivines in the barren Group I kimberlites studied are broadly similiar to those described for the diamondiferous Group I pipes. Thus, slight to moderate undulose extinction is characteristic of a small proportion of the large, rounded olivines in the Hamilton Branch kimberlite, and a majority of those from Ison Creek. Undulose extinction is also typical of many of the euhedral olivines in the latter kimberlite. Rare mosaictextured olivines, similiar to those described earlier, are also present in the Ison Creek kimberlite. Some of the euhedral and macrocryst olivines from the Gibeon kimberlites contain carbonate inclusions that appear to be primary. In other respects these kimberlites are petrographically similar to Ison Creek.

Olivines in the Group II kimberlites studied very rarely show evidence of undulose extinction. Where present, it generally not marked. Olivines with mosaic recrystallization textures appear to be extremely rare or absent in this group of rocks. A small proportion of the Newlands macrocrysts contain rounded or oval inclusions of serpentine and subordinate carbonate.

A characteristic feature of many of the euhedral olivines (including those with well-formed prism and pyramidal faces) in all of the kimberlites discussed above, is the tendency to display slight oblique extinction. This is typically of the order of  $6-10^{\circ}$ , but may be as much as  $30^{\circ}$ . In thin sections of a majority of the kimberlites studied, garnet, orthopyroxene and other mantle-derived minerals are extremely rare or absent, either as discrete grains, or as intergrowths with olivine. Exceptions are the Ison Creek kimberlite, which has prominent garnet and ilmenite macrocrysts (both less than 3% by volume), and the Gibeon kimberlites, both characterized by the presence of prominent, though volumetrically subordinate (<5%) macrocrysts of orange spinel, garnet and orthopyroxene.

Peridotite microxenoliths are rare or absent in a majority of the samples studied. However, they form a conspicuous, though volumetrically minor (5-10%) proportion of the rock in one of the four Bellsbank specimens, one of the two Letseng-la-terai samples, one from Kentucky and in the two Gibeon specimens. The Bellsbank microxenoliths are of interest as they include a clast consisting of strongly corroded olivine surrounded by a carbonate – possibly a rare example of the mantle carbonation reaction predicted by experimental studies (e.g. Wyllie and Huang 1976).

Prominent ilmenite macrocrysts distinguish the Monastery kimberlite from the others studied.

A summary of olivine petrographic characteristics in the kimberlites studied is given in Table 2.

Descriptions of olivines in olivine melilitites from two pipe clusters in the Namaqualand-Bushmanland area are presented by Moore and Erlank (1979) and Moore and Verwoerd (1985). The following are the salient features: Four petrographically distinctive olivine groups can be recognized in the olivine melilitites:

1) A dominant population of euhedral and often skeletal (hopper) olivine phenocrysts.

2) Very rare olivines characterized by the presence of abundant yellow or green glassy blebs, often concentrated about the margin of the grain, but also present as trains traversing the grain. They are typically anhedral, but occasionally have what appear to be relic crystal faces or, in one instance, good crystal form. They have been referred to as HILN olivines (Moore and Erlank 1979) – an achronym for their distinctive High Iron, Low Nickel chemical signature.

3) Rare, rounded olivine megacrysts (discrete grains larger than 10 mm – Harte 1977). Such megacrysts are restricted to the pipes of the Bushmanland cluster, which are Mg-rich compared to melilitites from Namaqualand to the south-west (Moore 1987).

4) Olivines in rare peridotite xenoliths. These have been recovered from some of the Bushmanland pipes, but appear to be absent in those from Namaqualand (Moore AE, Lock NP, Boyd FR, Nixon PH: Mantle derived xenoliths from a suite of olivine melilitites and related rocks from Bushmanland, South Africa. In prep.).

Most olvines of the latter two populations are characterized by strain features (undulose extinction and mosaic recrystallization textures). Slight undulose extinction is also characteristic of many, though not all, of the HILN-type olivines, and some of the euhedral olivines of the first population. The proportion of euhedral olivines showing strain features varies from pipe to pipe, but appears to be more common where a majority of the rare HILN-type olivines show strained extinction. Undulose extinction in euhedral olivines, if present, typically occurs across the whole crystal (rather than being restricted to a deformed centre with an undeformed overgrowth). This is analogous to the relationship seen in kimberlites. A further similarity is the tendency for many of the perfectly idiomorphic olivines in the olivine melilitites to show slight oblique extinction. The proportion of grains showing this characteristic appears to vary from pipe to pipe.

### **Olivine chemistry**

#### Analytical procedure

Olivines were analysed on a Cameca microprobe in the Department of Geochemistry, University of Cape Town, using a natural olivine internal standard for Ni determinations. Olivines in the de Beers kimberlite were analysed at 15 kV. Those in the remaining samples were analysed at 25 kV and a beam current of 40 mA, giving a lower limit of detection of 100 ppm Ni.  $2\sigma$  at 3000 ppm Ni is 100 ppm.

Olivines in each of the kimberlites studied were subjectively divided on size basis into large, intermediate, small and micro categories. Olivines from the former two groups are commonly rounded, and would be broadly equivalent to the macrocrysts of Skinner et al. (1979). The small and micro olivines are commonly euhedral. However, where rounding or marginal alteration is pronounced (e.g. the Monastery and Sover kimberlites), crystal habit is not always preserved. Between 50–100 olivine analyses were made for each kimberlite, generally on two or more thin sections cut from the same hand specimen. (Exceptions were Bellsbank – 4 specimens, and Letseng-la-terai – 2 specimens). Where olivine margins were sufficiently preserved, centre and edge were analysed. A summary of Mg/(Mg+Fe) vs. Ni for olivines in the kimberlites studied is given in Figs. 2–4.

There are systematic chemical differences between the olivine populations in the three varieties of kimberlites (diamondiferous and barren Group I and Group II) recognized in this study.

#### Group I kimberlites

In all of the Group I kimberlites studied (Figs. 3 and 4) the centres of the majority of the olivines analysed in thin section define a continuous compositional field covering a range in Mg/(Mg + Fe) with essentially uniform Ni concentrations (0.3% Ni). The larger rounded olivines in general concentrate at the Mg-rich extreme of this compositional spectrum, while smaller euhedral olivines are typically more iron-rich. (The K35 kimberlite from Gibeon is the single exception to this generalization.) However, there is invariably a considerable overlap in the chemical ranges of large and small olivines in all samples. The most refractory olivine compositions are close to Fo<sub>94</sub> in the diamondiferous Group I kimberlites (Fig. 3). This contrasts with most refractory compositions close to or below Fo<sub>92</sub> in the nondiamondiferous Group I suite (Fig. 4). Rim compositions in all of the Group I kimberlites define narrow compositional fields characterized by decreasing Ni within a limited range in Mg/(Mg + Fe) (87–89). The dominant population of Mg-rich olivines shows normal core to rim zonation towards the field of edge compositions.

A second olivine compositional group, iron-rich relative to rim compositions and thus showing reversed core to rim zonation, is present in all of the Group I kimberlites studied. It includes both large rounded grains and small euhedral crystals. These iron-rich olivines generally form a very subordinate proportion of the total olivine population, except at Monastery, described in more detail below.

The Monastery kimberlite is characterized by the presence of an unusually large number of megacrysts relative to other kimberlites. Gurney et al. (1975) show that the olivine megacrysts at Monastery fall into two distinct compositional groupings – one relatively Mg- and Ni-rich and the other iron-rich and Ni-poor. Monastery megacryst olivines are shown by the solid diamond ornament in Fig. 3(b) [Data from Jakob WRO (1975) Geochemical aspects of the megacryst suite from the Monastery kimberlite pipe. Unpubl. M.Sc. thesis, University of Cape Town].

Olivines analysed in this study from thin sections cut from the Monastery kimberlite show a wide range in composition (Fig. 3b). The macrocrysts fall into three distinct groupings:

1) A population with a range in Mg/(Mg+Fe) from 94 to 88.3, and uniform Ni contents (close to 0.3% Ni).

# Table 2. Macrocryst/phenocryst characteristics in kimberlites studied

Kimberlite	No of thin sections	Olivine characteristics	Presence of discrete garnet, chromite, pyroxene	Possible micro-xenoliths	
Newlands	6	Predominantly discrete olivine grains. Rare individuals show slight undulose extinction but most are strain-free. Clasts with mosaic re- crystallization textures are absent. Rare macrocrysts with round or oval inclusions of serpentine and subordinate carbonate	Absent	4 macrocrysts with chromite inclu- sions and one with brown finger- print spinel. Macrocrysts with round pyrope inclusions seen in hand specimen, though not in thin section	
Bellsbank	5 cut from 4 hand specimens	Predominantly discrete grains lack- ing undulose extinction. Some eu- hedral olivines show oblique extinc- tion	Extremely rare (<1%) chromite xenocrysts	Olivine macrocryst with disloca- tion lamellae and a single round G10 garnet inclusion, and a clast with corroded olivine enclosed by carbonate in one specimen	
Sover	2	Predominantly discrete grains lack- ing undulose extinction. Minority with slight undulose extinction. Some euhedral crystals with oblique or undulose extinction.	Absent	1 microperiodotite clast	
de Beers	2	Dominantly discrete olivine grains. A majority of macrocrysts show moderate to pronounced undulose extinction, and some have marginal mosaic recrystallization selvages. Undulose extinction is typical of many euhedral crystals	Absent	2 composite olivine (dunite) clasts and very rare mosaic textured clasts.	
Letseng la Terai (NL 381)	2	Dominantly discrete olivine grains, many with slight undulose extinc- tion	Absent	Prominant microperidotite clasts (5–10%). Very Rare mosaic tex- tured clasts	
Letseng la Terai (NL 416)	2	Dominantly discrete strain-free grains. A minority show slight undu- lose extinction	Absent		
Jagersfontein	1	Dominantly discrete olivine grains, most strain-free, though some rounded and some euhedral olivines show slight undulose extinction	1 garnet grain	Single olivine with attached garnet. Minor crustal xenoliths	
Monastery	4	Mostly strain-free discrete olivine grains. Rare individuals show faint undulose extinction	2 small garnet grains (<1%). Ilmenite macrocrysts prominent.	2 micro-peridotite clasts	
Hamilton Branch	1	All olivine grains discrete. About 25% show undulose extinction. The remainder are stain-free. Oblique extinction in euhedral olivines is uncommon. Many large macrocrysts extinguish parallel to long crystal edges	Absent	Absent	
Ison Creek	3	About 10% of olivine clasts are composite grains, the remainder dis- crete. A majority of macrocrysts and many euhedral crystals have moder- ate to pronounced undulose extinc- tion	Garnet and ilmenite macrocrysts (both %) and rare opx and cpx macrocrysts prominent	Prominent (10%) micro-peridotite clasts	
Gibeon: K39	4	Dominantly discrete olivine grains. Some macrocryst and euhedral oliv- ines enclose carbonate, sometimes as round inclusions	Garnet, spinel, opx and phlogopite macocrysts present (1–2%)	Minor (5–10%) microperidotite clasts	
Gibeon: K35	4	As for K39	As for K39	As for K39	

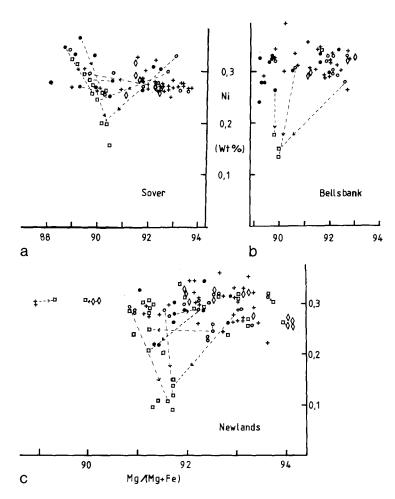


Fig. 2a-c. 100 Mg/(Mg+Fe) (atomic proportions) vs. Ni (wt%) for olivines in diamondiferous Group II kimberlites. ◊: Centres, large olivines (typically rounded); +: Centres, intermediate sized olivines (typically rounded); ○: Centres, small olivines (typically euhedral); □: Edges, all olivines. *Dashed lines* join analyses for individual grains

These correspond to the dominant Mg-rich olivines that occur in the other Group I kimberlites.

2) Olivines with Mg/(Mg + Fe) in the range 87–83 and greater than 0.19% Ni. This group overlaps the compositional field of the Mg-rich megacrysts, although extends to slightly more Fe-rich and Ni-poor compositions.

3) Olivines with less than 0.14% Ni and Mg/(Mg+Fe) ranging from 86–78.3. The iron-rich extreme of this range overlaps the field of the iron-rich megacrysts.

The small euhedral olivines at Monastery fall into two distinct compositional groupings. One overlaps the ironrich end of the field of the first olivine macrocryst population, and is separated by a distinct compositional gap ( $Fo_{88-86}$ ) from the remaining euhedral olivines. The latter cover much of the chemical range of the second population of macrocryst olivines. They straddle the Ni hiatus (0.14–0.19% Ni) that separates the two relatively iron-rich groups of olivine macrocrysts analysed in thin section, and overlap the Mg-rich end of the low-Ni olivine macrocrysts.

Strong marginal alteration of the Monastery olivines prevents analysis of rim compositions. However, it is noted that the iron-rich extreme ( $Fo_{88.3}$ ) of the Mg-rich Monastery olivines is within the range of rim compositions established for the other Group I kimberlites studied. It is therefore inferred that the field of late-stage compositions at Monastery was close to  $Fo_{88}$ .

#### Group II kimberlites

In the Group II kimberlites, a majority of the small euhedral and large rounded olivines collectively define a continuous chemical population characterized by essentially uniform Ni (0.3% Ni), and a range in Mg/(Mg+Fe). The most refractory compositions (close to Fo<sub>94</sub>) match those in the diamondiferous Group I kimberlites studied. Rim compositions of these olivines are however consistently more Mg-rich (Fo<sub>89-92</sub>) than in the Group I suite.

Euhedral and macrocryst olivines characterized by reversed zoning with respect to Mg-Fe are found in all of the Group II kimberlites studied. These are chemically analogous to the iron-rich olivines in Group I kimberlites. However, the compositional separation between normally- and reverse-zoned populations is not as marked as in the latter kimberlite suite. The reverse-zoned olivines in the Group II kimberlites also show a much more restricted compositional range (Fo<sub>91-88.2</sub>, uniform Ni close to 0.3%) compared to those in Group I kimberlites.

## South African olivine melilitites

Olivines in South African olivine melilitites define Mg- and Fe-rich compositional groupings that are analogous to those in kimberlites but differ in their chemical ranges (Fig. 5). The majority of the euhedral olivines show normal core to rim zonation with respect to Mg/(Mg+Fe) and Ni (e.g. Fig. 5b). In the Sutherland olivine melilitite (Fig. 5a), both the relatively large (though in general euhedral) and some of the small euhedral olivines are characterized by reversed zonation with respect to Mg/(Mg+Fe). They are thus compositionally equivalent to the Fe-rich olivine population in kimberlites.

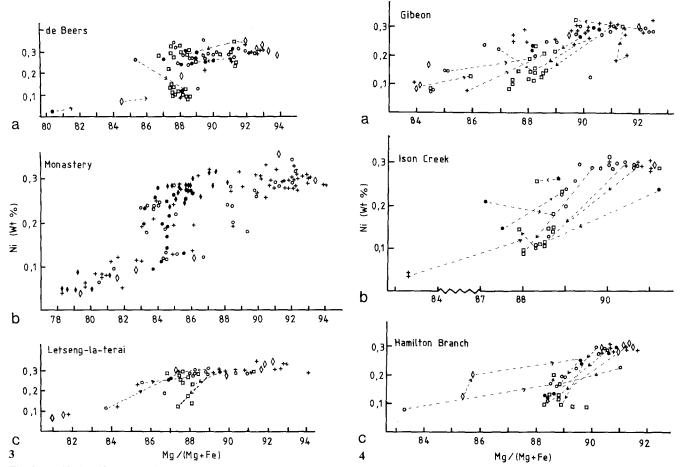


Fig. 3a-c. 100 Mg/(Mg + Fe) (atomic proportions) vs. Ni (wt%) for olivines in diamondiferous Group I kimberlites; •: Megacryst olivines. Remaining symbols as for Fig. 2

Fig. 4a–c. 100 Mg/(Mg+Fe) (atomic proportions) vs. Ni (wt%) for olivines in non-diamondiferous Group II kimberlites. Symbols as for Fig. 2

In the Dikdoring olivine melilitite from Namaqualand, (Fig. 5b) the rare, though petrographically distinctive olivines of the second population described in these rocks (Moore and Erlank 1979), range in composition from very iron-rich individuals towards the evolved (Fe-rich) extreme the dominant euhedral population. In this respect they are analogous to Fe-rich olivines in kimberlites. They are however characterized by very low Ni contents, generally below detection limit. In view of their distinctive <u>High</u> Iron, Low Nickel chemical signature, they have been referred to as <u>HILN</u> olivines (Moore and Erlank 1979).

A further characteristic of the olivine melilitites is the presence of some euhedral olivines, frequently large relative to associated euhedral phenocrysts, whose cores show a wide, non-systematic range in composition. Centre to edge zonation is towards the fields for the centres or edges of the majority of the euhedral olivines (e.g. Fig. 5b), resulting in aberrant zonation patterns with respect to Ni and Mg/ (Mg+Fe). Olivines showing such aberrant zonation patterns are for convenience referred to as "unusual hopper olivines".

Additional chemical data for olivines from South African olivine melilitites are given by Moore and Erlank (1979).

## General

Within each of the three groups of kimberlites discussed, olivines show qualitatively similiar variation patterns and compositional ranges. This generalization would also apply for olivine melilitites from the same pipe cluster [Moore AE (1979) The geochemistry of the olivine melilitites and related rocks of Namaqualand-Bushmanland, South Africa. Unpubl. Ph.D. thesis, University of Cape Town; Moore and Erlank 1979]. There is, further, a fairly systematic relationship between the average bulk rock Mg-number (100 Mg/(Mg + Fe)) of each group of kimberlites, or cluster of olivine melilitites, and the compositions of most Fe- and Mg-rich olivines in each case. This is shown in Table 3. In the table, the average composition for Group II kimberlites is from Smith et al. (1985). For the diamondiferous and non-diamondiferous Group I kimberlites respectively, average compositions for Group IA and IB kimberlites (Smith et al. 1985) have been used. Average bulk rock compositions for different clusters of olivine melilitites are from Moore (1979).

Average Mg-number and compositions of the most refractory olivines are closely similar for Group IA and Group II kimberlites. The data in Table 3 show that with



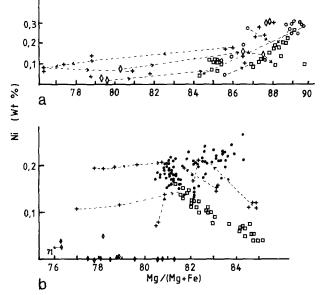


Fig. 5. a 100 Mg/(Mg + Fe) (atomic proportions) vs Ni (wt%) for olivines in the Sutherland olivine olivine melilitite. Symbols as for *Fig. 2.* Note however, that the olivines of the two larger size groups are mostly euhedral; b 100 mg/(Mg + Fe) (atomic proportions) vs Ni (wt%) for olivines in the Dikdoring olivine melilitite, Namaqualand;  $\circ$ : Centres of euhedral and sometimes skeletal olivines;  $\diamond$ : HILN olivines; +: Unusually zoned euhedral olivines. *Arrows* denote sense of centre to edge zonation;  $\Box$ : Edges, all olivines

decreasing average bulk rock magnesium number of the remaining groups of rocks, there is a sympathetic decrease in Mg/Fe of the most refractory olivine. With the exception of the Group IB kimberlites, the composition of the most iron-rich olivine in each group of rocks becomes more faya-litic in the same sequence.

# **Origin of olivines**

#### General

The kimberlites studied have a wide distribution. In addition, those within each of the three sub-groups recognized have olivines with similiar compositional ranges. It is therefore considered that the suite studied is sufficiently representative to permit generalizations regarding the origin and possible significance of kimberlitic olivines.

The presence of two chemically distinct olivine compositional groups (one relatively Mg-rich, and showing normal zonation, the other iron-rich and having reversed zonation) in all kimberlites studied, including those from Kentucky, points to the origin of both populations being intimately linked to kimberlite genesis. The qualitatively similiar chemical relationship between the two olivine populations in all kimberlites indicate the operation of broadly analogous processes in each case. However, the restricted chemical fields of the two olivine populations in Group II kimberlites relative to the Group I suite point to possible differences in detail. The origins of the two olivine compositional populations are considered separately.

## Mg-rich olivine population

The idiomorphic Mg-rich olivines are presumed to be cognate phenocrysts. The possibility that some may represent

**Table 3.** Relationship between average bulk rock Mg-number (100 Mg/(Mg + Fe) (atomic proportions) of kimberlite groups and South African olivine melilitite pipe clusters and compositions of the respective most Mg-rich and Fe-rich olivines

Average composition used	Mg- number	100/Mg(Mg+Fe) (olivine)	
		most Mg-rich	most Fe-rich
Group II kimberlite <sup>a</sup>	87.6	94.1	88.2
Group IA kimberlite <sup>a</sup>	86.1	94.2	78.3
Group IB kimberlite <sup>a</sup>	78.9	92.6	83.3
Sutherland olivine melilitite <sup>b</sup>	72.4	89.5	75.8
Gamoep olivine melilitite (Bushmanland) <sup>b</sup>	67.9	86.8	70.8
Garies olivine melilitite (Namaqualand) <sup>b</sup>	51.3	85.0	66.2

<sup>a</sup> Data from Smith et al. (1985)

<sup>b</sup> Data from Moore (1979)

Olivine data for the Namaqualand and Bushmanland olivine melilitites from Moore (1979)

overgrowths on exotic xenocrysts cannot be discounted, but is not supported by petrographic evidence. Thus, in instances where idiomorphic crystals show undulose extinction, this affects both core and rim of the crystal, indicating that deformation occurred at a late stage in the evolution of the kimberlite magma.

The cause of late-stage deformation in euhedral olivines is an intriguing problem. Kreston (1973) attributes latestage deformation features in the Lemphane kimberlite (Lesotho) to torsional forces applied to the magma during intrusion in a plastic or near solid state. It is possible that the deformation features in euhedral olivines is related to a similiar process.

The origin of the macrocryst (large rounded) Mg-rich olivines is more problematic. Bailey (1984) suggests that such olivines are xenocrysts derived from disaggregated periodotite xenoliths. Several lines of evidence support this interpretation:

1) The presence of peridotite xenoliths in kimberlites implies that some olivines are undoubtedly xenocrysts. The variation in olivine composition in sheared and granular lherzolite nodules recovered from South African kimberlites is generally in the range  $Fo_{88-94}$  [Nixon and Boyd 1973; Lock NP (1980) The geology of the Letseng kimberlites, Lesotho. Unpubl. Ph.D. thesis, Sheffield University]. This covers much of the compositional range of kimberlite macrocrysts, further supporting an exotic origin for such olivines.

2) Macrocrysts in some kimberlites show undulose extinction and recrystallization features resembling those seen in mantle-derived peridotites.

3) Consideration of Mg – Fe equilibria between olivine and liquid could be used to argue for an exotic origin for the macrocrysts. Thus, assuming  $K_D$  (as defined by Roeder and Emslie 1970) equals 0.4 and Fe<sub>2</sub>O<sub>3</sub>=0.85 FeO, subtraction of 29% olivine from an average Group II kimberlite (Smith et al. 1985) would be necessary to derive a composition that would be in equilibrium with a refractory mantle olivine (Fo<sub>94</sub>). Subtraction of even greater amounts of olivine would be required for a lower value of  $K_D$ . At face value, these considerations provide compelling reason for concluding that a large proportion of olivines in kimberlite are xenocrysts. There are however several lines of reasoning which throw doubt on this interpretation.

1) Olivine is clearly an early crystallizing phase in kimberlites. If kimberlite magmas were generated in equilibrium with refractory mantle phases, early crystallizing olivines would be expected to have compositions matching those in the source region, and would reasonable be expected to be large. They would be exposed to the evolving kimberlite magma for longer periods than later forming (smaller) crystals, and would be in chemical disequilibium with the late stage liquids. They would thus be more prone to rounding. In all of the Group I kimberlites other than the Gibeon sample, the Mg-rich olivine population shows a continuous compositional range from large rounded, generally more Mg-rich grains to more Fe-rich small euhedral crystals. This is the pattern that would be expected for continuous crystallization over a wide depth interval during ascent of the kimberlite magma.

Separation of the compositions of large and small olivines is less marked in the Group II kimberlites. This might in part be related to the more limited olivine compositional range in these rocks, which implies a correspondingly more limited temperature interval of crystallization.

A possible objection to the interpretation of rounded and euhedral olivines representing a continuous crystallization sequence is the uniform Ni (0.3% Ni) of the Mg-rich olivine population. Modelling of Ni in olivine is complicated by uncertainties regarding the olivine-liquid Ni partition coefficient in kimberlitic melts. Hart and Davis (1978) show that in basaltic liquids, the Ni partition coefficient can be calculated from the relationship:

$$K_{\rm Ni}^{\rm Ol-liq} = 124/{\rm MgO}_{\rm (lig)} - 0.9$$

However, Smith et al. (1980) suggest that for komatiitic magmas, the Ni partition coefficient varies with liquid composition according to the relationship:

$$K_{\rm Ni}^{\rm Ol-liq} = 124/{\rm MgO}_{\rm (lig)} - 2.85$$

Ni variation in kimberlite olivines has been modelled using both relationships, and assuming an original liquid composition equivalent to an average Group I kimberlite (MgO=28.5, Ni=1360 ppm; Smith et al. 1985). 5% olivine was subtracted from this composition and the residual liquid composition calculated (using the Rayleigh partitioning equation for Ni). A new Ni partition coefficient was then calculated and used to determine the expected Ni content of the equilibrium olivine. A further 5% olivine was subtracted, and the process repeated.

For removal of 35% olivine, predicted Ni contents of the phase decrease from 4690 to 2480 ppm Ni using the Hart-Davis equation. Using the relationship of Smith et al. predicts an *increase* in Ni in olivine from 2040 to 3300 ppm over the same range of crystallization. Averaging the two sets of results gives a small decrease in predicted Ni content of the olivines from 3370 to 2900 ppm. Thus, the behaviour of Ni in the Mg-rich olivine suite is consistent with a continuous crystallization model within the uncertainty in determining the Ni partition coefficient.

2) Average Ni contents of Groups I and II kimberlites are closely similiar (1360 and 1400 ppm Ni respectively – Smith et al. 1985). If it is assumed that these average compositions approximate those of primary liquids formed in equilibrium with mantle olivine (0.31% Ni), Ni partition coefficients in the range 2.21–2.28 are implied. This is intermediate between the extremes of 1.50–3.45 predicted on the basis of the equations of Smith et al. (1980) and Hart and Davis (1978), discussed above. Bulk rock Ni data do not therefore preclude the possibility that the average Groups I and II kimberlite compositions are similar to those of primitive liquids.

3) The continuous size distribution shown by olivines in several of the kimberlites studied suggests a single dominant population. Monastery is a possible exception to this generalization. However, the olivine chemical data show that this kimberlite is very atypical in having a significant proportion of large olivines with compositions that point to derivation from the megacryst suite. Olivines with comparable compositions are rare in those pipes with a unimodal size distribution. Further, the proportions of orthopyroxene, clinopyroxene, garnet and spinel are always very low in kimberlites. Extremely efficient resorption of these phases by the magma would be necessary if a significant proportion of kimberlitic olivines is exotic. The overwhelming dominance of olivine phenocrysts, and their size distributions are, on the other hand, consistent with formation during an extended period of crystallization. This may in turn be controlled by expansion of the olivine phase field with decreasing pressure (Wyllie and Huang 1976) during magma ascent.

4) Deformation features in large rounded olivines cannot be regarded as a reliable criterion for distinguishing xenocryst olivines, as similiar features are present in euhedral olivines.

5) A. Harwood (Kimberlite Symposium, University of Cape Town 1985) documents primary fluid inclusions in euhedral kimberlite olivines, with morphologies that can be related to temperature of olivine crystallization. Preliminary studies of inclusion morphology in macrocryst olivines suggest similarities with the highest temperature inclusions in the euhedral crystals. Such a relationship is consistent with the macrocrysts being early crystallizing cognate phenocrysts. Carbonate inclusions in macrocryst and euhedral olivines in the Gibeon pipes support this interpretation. The morphology of the rounded serpentine inclusions in some Newlands macrocrysts suggests that they may represent trapped liquids, which would also favour a cognate origin for these olivines.

7) The Mg-Fe distribution coefficient  $(K_D)$  for olivine and kimberlite magmas has not been established experimentally. However, it may be strongly influenced by the CO<sub>2</sub> content of the melt, as solution of CO<sub>2</sub> in silicate liquids results in polymerization reactions such as:

# $2 \text{CO}_2 + 2 \text{SiO}_4^{4-} = \text{Si}_2 \text{O}_6^{4-} + 2 \text{CO}_3^{2-}$ (Fraser 1975)

Mg/Fe ratios in (more polymerized) inosilicates in mantle lherzolites are higher than those of associated nesosilicates. By analogy, polymerized carbonate rich melts in equilibrium with mantle olivines would be expected to be Mg-rich relative to less polymerized carbonate-free liquids. This implies a higher  $K_D$  in carbonate-bearing systems. Caution is therefore necessary in using the distribution coefficient to estimate the proportion of xenocrystal olivines in kimberlites.

A decrease in  $K_D$  associated with decreasing pressure or CO<sub>2</sub> loss during magma ascent would lead to crystallization of more Mg-rich olivines. This might contribute to the overlap of the compositional fields of macrocrysts and euhedral olivines, particularly in Group II kimberlites.

8) Initial Fe<sub>2</sub>O<sub>3</sub>/FeO ratios of kimberlite magmas are not known. However, the presence of diamonds in deepseated mantle rocks implies that the source region for kimberlites might be relatively reduced. Equilibrium melts would be correspondingly reduced, and would have low initial Fe<sub>2</sub>O<sub>3</sub>/FeO ratios. This would in turn decrease the estimate of the proportion of xenocrystal olivines for any given value of  $K_{\rm p}$ .

The arguments presented above do not unequivocally demonstrate that a majority of kimberlite olivines are cognate phenocrysts. However, they do suggest that further investigation is needed to establish the proportion of exotic olivines in kimberlites. A more detailed characterization of primary inclusions in the olivines, and experimental studies on Mg-Fe and Ni partitioning between olivine and kimbelitic liquids at high pressures would place more rigorous constraints on this question.

#### Fe-rich olivine population

The iron-rich (relative to edge compositions) olivines are best represented at Monastery. Their origin is thus discussed in relation to the data for this kimberlite.

At Monastery, a significant proportion of the iron-rich olivines analysed in the thin section study overlap the chemical fields outlined for the two olivine megacryst populations in this kimberlite. This suggests an origin related to the formation of the low-Cr megacryst suite.

Harte and Gurney (1980) propose that the low-Cr megacrysts form as a result of injection of small fractions of kimberlitic liquid into fractures surrounding the main body of the kimberlite magma at depth. They envisage that these liquid fractions would cool and crystallize across the thermal aureole surrounding the magma reservoir, to form a pegmatitic vein network. This model would account for the observed isobaric range in megacryst equilibration temperatures. These authors do not account for the compositional hiatus separating the two megacryst olivine populations. A possible explanation would be non-crystallization of olivine over part of the temperature range of megacryst formation.

The Hart-Gurney model is supported by Shulze (1984), who notes however, that equilibration temperatures of the chemically most evolved megacrysts are above the kimberlite solidus. This implies ambient mantle conditions at the depth of megacryst formation above the kimberlite solidus. If so, pockets of evolved kimberlite-derived liquids would have remained in equilibrium with the most evolved (Ferich) megacrysts in the hypothetical fracture-fill network. Not all pegmatitic veins are likely to traverse the entire thermal aureole surrounding the magma at depth. In such veins, residual liquids would be less evolved.

It is suggested that during kimberlite eruption, both megacrysts and evolved liquids would be entrained by the kimberlite magma. Some of the iron-rich macrocrysts may represent megacryst fragments. It is suggest that the euhedral iron-rich olivines crystallized from the entrained liquids. These liquids would change in composition as a result of mixing with the main volume of kimberlite magma. Consequently, olivine rims would evolve in composition towards those of the dominant Mg-rich population crystallizing from the kimberlite magma. The iron-rich macrocryst olivine compositions (analysed in thin section) extend beyond the field established for the iron-rich megacrysts. This may indicate inadequate sampling of the latter population. However, the hypothetical entrained liquids would be expected to be volatile-rich, which might favour the growth of relatively large crystals. Some of the iron-rich macrocrysts may therefore have crystallized during magma ascent.

Reference to Figs. 3 and 4 suggests that iron-rich olivines in other Group I kimberlites have compositions analogous to those in the Monastery pipe. However, their rarity in these kimberlites suggests that only minor proportions of the megacryst suite and associated evolved liquids were entrained during eruption.

The Fe-rich olivine population in Group II kimberlites is considered to have an origin analogous to that suggested for the Group I kimberlites. Their restricted compositional range with respect to Ni and Mg (compared to those in Group I kimberlites) would then indicate that the entrained liquids from which they crystallized were less evolved than those incorporated by Group I kimberlites. This would in turn require higher ambient mantle temperatures at the depth of megacryst formation, and correspondingly smaller degrees of crystallization of liquids in the fracture network surrounding Group II kimberlites magma chambers at depth.

Limited crystallization of the fracture-fill liquids would account for the apparent rarity of the megacryst suite in Group II kimberlites (Smith et al. 1985). As ilmenite is commonly a late-crystallizing component of the megacryst suite (Gurney et al. 1979) such a model would also explain the rarity or absence of this phase in Group II kimberlites (Smith et al. 1985). As Group II and diamondiferous Group I kimberlites show a broad geographical overlap in their distribution, the inferred less evolved (higher temperature) character of liquids incorporated by Group II kimberlite magmas would suggest entrainment at greater depths than in the case of Group I magmas. A logical extension of this reasoning is that Group II kimberlites may be derived from deeper sources than the Group I suite.

## Olivines in olivine melilitites

The relationship between Fe- and Mg-rich olivine populations in the Sutherland olivine melilitites (Fig. 5a) is comparable to that established for kimberlites. This suggests an origin analogous to that presented for the kimberlites.

HILN olivines in the Dikdoring olivine melilitite (from the Namaqualand piper cluster) collectively show a range in Mg/(Mg+Fe) from very iron-rich compositions towards the low-Mg extreme of the field for the associated euhedral olivines. In this respect they are also analogous to the ironrich olivine population in kimberlites. However, a striking difference is the extremely low Ni content of the HILN olivines, which requires explanation.

Moore (1987) suggests that oxygen fugacities of liquids that were in equilibrium with evolved megacrysts may have been buffered by highly reduced mantle wall rocks. The low Ni contents of HILN olvines may therefore reflect crystallization from reduced liquids, entrained by the melilitite magma. An alternative possibility is that the entrained liquids were in equilibrium with a sulphide phase (possibly an immiscible sulphide liquid). Magmas in equilibrium with a given olivine composition would become more Mg-rich with increasing value of the distribution coefficient. The ultrabasic character of kimberlites may therefore be primarily determined by a high value of  $K_{\rm D}$  for carbonate-rich kimberlite magmas at the depth of melting.

Olivines within each of the three different groups of kimberlites recognized in this study (diamondiferous and barren Group I and Group II) are characterized by closely similiar zonation patterns. The differences in olivine zonation patterns between each of these kimberlite varieties suggests crystallization from chemically distinct magma sub-types. Bulk rock chemical data discussed by Smith et al. (1985) in general discriminate between the different kimberlite types recognized in this study (i.e. Group IB kimberlites are broadly synonomous with non-diamondiferous Group I kimberlites). However, discrimination on the basis of bulk roch chemistry should be applied with caution. In the Monastery kimberlite, incorporation of unusually large proportions of residual fracture-fill liquids (probably Fe- and Tirich) that were in equilibrium with evolved megacrysts, might, in part, account for the classification of this kimberlite as a Group IB variety. An implication is that olivine chemistry may be a more reliable discriminant than bulk rock chemistry in subdividing Group I kimberlites.

#### **Economic implications**

The economic grade of kimberlites is determined by the presence of diamonds in mantle rocks sampled during ascent, coupled with the degree of subsequent resorption in the kimberlite magma.

Peridotitic inclusions in diamonds point to a paragenesis involving extremely refractory source rocks. The compositional differences between the respective most refractory olivines in the diamondiferous and non-diamondiferous kimberlites studied, indicate that the former were in equilibrium with, or sampled relatively more refractory mantle rocks. Thus the composition of the most refractory olivines in kimberlites may reflect the likelihood that a richly diamondiferous portion of the mantle was sampled during magma ascent. Note, however, that this will not indicate whether eclogite-related diamonds were entrained by the kimberlite magma.

Octahedrons and cubes are the primary diamond growth forms in mantle rocks. Resorption in the inhospitable kimberlite magma characteristically produces dodecahedral crystal forms [Robinson DN (1979) Surface textures and other features of diamond. Unpublished Ph. D. thesis, University of Cape Town]. Statistical data for diamond habit are only available for a few kimberlites (Harris et al. 1979). It is interesting, however, that octahedron stones appear to be prominant in the Newlands kimberlite (Group II) (Wagner 1914), but are in contrast extemely rare at Letseng-la-terai (Group I) (Harris et al. 1979). It may be significant that the Mg-rich olivine population ranges to more evolved compositions in the latter kimberlite. In other words, the apparently greater degree of diamond resorption in the Letseng-la-terai kimberlite may be related to a greater degree of magma crystallization and evolution. Conversely, the relatively high grades of many Group II kimberlites (e.g.

4 ct/ton at Swartruggens) may in part be related to more limited diamond resorption in these kimberlites.

In a number of kimberlites (e.g. Letseng-la-terai and Monastery), economic viability is dependent on the recovery of a small proportion of large, high quality stones, whose abundance is extremely difficult to evaluate during a limited bulk sampling programme. Moore (1987) suggests that these large stones may be linked to the formation of the discrete nodule suite. More detailed characterization of the Fe-rich olivine population associated with such kimberlites (possibly in terms of compositional range and relative abundance) may offer insights regarding the likelihood of finding large stones, and their relative abundance. Better characterization of the megacryst suite, may also be instructive.

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