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# **Crystallization of Cr-poor and Cr-rich megacryst suites from the host kimberlite magma: implications for mantle structure and the generation of kimberlite magmas**

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Abstract Cr-poor and Cr-rich megacryst suites, both comprising of varying proportions of megacrysts of orthopyroxene, clinopyroxene, garnet, olivine, ilmenite and a number of subordinate phases, coexist in many kimberlites, with wide geographic distribution. In rare instances, the two suites occur together on the scale of individual megacryst hand specimens. Deformation textures are common to both suites, suggesting an origin related to the formation of the sheared peridotites that also occur in kimberlites. Textures and compositions of the latter are interpreted to reflect deformation and metasomatism within the thermal aureole surrounding the kimberlite magma in the mantle. The megacrysts crystallized in this thermal aureole in pegmatitic veins representing small volumes of liquids derived from the host kimberlite magma, which were injected into a surrounding fracture network prior to kimberlite eruption. Close similarities between compositions of Cr-rich megacryst phases and those in granular lherzolites are consistent with early crystallization from a primitive kimberlite liquid. The low-Cr megacryst suite subsequently crystallized from residual Cr-depleted liquids. However, the Cr-poor suite also reflects the imprint of contamination by liquids formed by melting of inhomogeneously distributed mantle phases with low melting temperatures, such as calcite and phlogopite, present within the thermal aureole surrounding the kimberlite magma reservoir. Such carbonate-rich melts migrated into, and mixed with some, but not all, of the kimberlite liquids injected into the mantle fracture network. Contamination by the carbonate-rich melts changed the Ca-

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ARC National Key Centre for Geochemical Evolution and Metallogeny of Continents, Department of Earth and Planetary Sciences, Macquarie University, NSW, 2109, Australia Mg and Mg–Fe crystal–liquid distribution coefficient, resulting in the crystallization of relatively Fe-rich and Ca-poor phases. The implied higher crystal-melt Mg–Fe distribution coefficient for carbonate-rich magmas accounts for the generation of small volumes of Mg-rich liquids that are highly enriched in incompatible elements (i.e. primary kimberlite magmas). The inferred metasomatic origin for the sheared peridotites implies that this suite provides little or no information regarding *vertical* changes in the thermal, chemical and mechanical characteristics of the mantle.

#### Introduction

Mantle-derived inclusions in kimberlites include a suite of mono-mineralic grains, termed megacrysts, that are often considerably larger than 10 mm in diameter (Harte 1977). Orthopyroxene, clinopyroxene, garnet, olivine and ilmenite are common megacryst phases, while subordinate phlogopite and zircon have been recorded at some localities. Eggler et al. (1979) showed that the cluster of kimberlites that straddles the Wyoming-Colorado border (USA) is characterized by the presence of chemically discrete Cr-poor and Cr-rich megacryst populations. Analogues of the former suite appear to be present in many, and possible most, of the isotopically distinctive Group I kimberlites recognized by Smith (1983), which broadly correspond to the basaltic variants of Wagner (1914). The chemical and textural characteristics of the Cr-poor megacryst suite are well documented for a variety of localities with a wide geographical distribution. Examples are the Lesotho kimberlites (Nixon and Boyd 1973a), Monastery (Gurney et al. 1979; Moore et al., 1992; Griffin et al. 1997), Orapa (Shee and Gurney 1979), the Gibeon kimberlites (Mitchell 1987) and Jagersfontein (Hops et al. 1989, 1992) in southern Africa. (The locations of these and other southern African kimberlites referred to in this

study are shown in Fig. 1.) In North America, this group of megacrysts is well documented for the Colorado-Wyoming kimberlites (Eggler et al. 1979) the Hamilton Branch kimberlite, Kentucky (Schulze 1984) and kimberlites of the Slave Province (e.g. Kopylova et al. 1999).

The Cr-rich megacryst suite has received far less attention. Nevertheless, there is growing evidence that suggests that it occurs in kimberlites with a wide geographic distribution. Thus, Shee and Gurney (1979) recognized both Cr-rich and Cr-poor clinopyroxene megacryst populations in the Orapa kimberlite in Botswana, and inferred that these are chemically comparable to the two megacryst suites in the Colorado-Wyoming pipes. Data presented by Mitchell (1987) indicate that compositionally discrete Cr-poor and Cr-rich clinopyroxene megacryst populations are also present in the Gibeon pipes in Namibia. Kharkiv (1995) reports the presence of both Cr-poor and Cr-rich megacryst suites from kimberlites in the Verkhnemunskoye field, Republic of Sakha.

Boyd et al. (1984) reported the occurrence of discrete apple green clinopyroxene megacrysts, which they termed "Granny Smith" diopsides, from Jagersfontein and a number of the Kimberley pipes, and noted that these are chemically comparable to the Orapa Cr-rich clinopyroxene megacrysts. The "Granny Smith" diopside suite is also present in the Nouzees kimberlite in North-West Province, South Africa (Jones 1987).

Paired high- and low-Cr ilmenite populations characterize many of the pipes in the Tsabong – Molopo kimberlite field in SW Botswana (Moore 1987; Moore and Lock 2001). Moore et al. (1992) have documented



Fig. 1 Localities of southern African kimberlites or kimberlite clusters referred to in the text. *1* Gibeon cluster (Mukurob 1 and 2 kimberlites); *2* Nouzees; *3* Weltevreden-Frank Smith; *4* Kimberley cluster (Bultfontein, de Beers, du Toitspan, Kimberley and Wesselton kimberlites); *5* Jagersfontein; *6* Swartruggens; *7* Lace (also sometimes referred to as Crown); *8* Kaalvallei; *9* Monastery; *10* Northern Lesotho cluster (Thaba Putsoa and Letseng kimberlites); *11* Tsabong-Molopo cluster (M4 kimberlite); *12* Kokong (or Kang) cluster (KN70 kimberlite); *13* Orapa cluster (Orapa or AK1, Letlakane or DK1 and BK4, BK7 and BK13 kimberlites)

two relatively Cr-rich ilmenite populations from the Monastery kimberlite, characterized by different and non-overlapping ranges in MgO. These are volumetrically very subordinate to the dominant Cr-poor ilmenite suite described by Gurney et al. (1979) from this kimberlite.

A study of garnet megacrysts from the Group II Swartruggens kimberlite, South Africa (De Bruin 1998) showed that the associated clinopyroxene inclusions can be grouped into chemically distinct Cr-rich and Cr-poor populations.

Analogues of the Cr-rich megacrysts are therefore present in kimberlites with a wide geographic distribution. Each of these localities is also characterized by the presence of Cr-poor megacrysts. Despite the evidence for the two megacryst populations in many kimberlites with a wide geographic distribution, the link between the two suites and their origin has long presented a conundrum, and remains controversial. However, as discussed below, resolving this question has important implications for understanding the processes responsible for generating kimberlite magmas.

Most attention has been focused on the Cr-poor megacrysts, and a variety of theories have been advanced to account for their origin. There is widespread consensus that this suite crystallized from a magma that was present either very shortly prior to or at the time of kimberlite eruption (e.g. Nixon and Boyd 1973a; Gurney et al. 1979; McCallister et al. 1979; Schulze 1984). However, there is little consensus as to the nature of this liquid.

Crystallization from the host kimberlite magma has been proposed by a variety of authors (e.g. Mitchell, 1973, 1986, 1987; Schulze 1987; Moore and Lock 2001; Schulze et al. 2001). In contrast to these essentially cognate models, the Cr-poor megacrysts have been linked to a variety of other primary mantle-derived liquids. Thus, Griffin et al. (1989) propose that they form from a proto-kimberlite, while Moore et al. (1992) suggest a meimechite parent (essentially a volatile, LIL element-enriched komatiite). Harte (1983) argued that the Cr-poor megacrysts crystallized from a basanitic melt, Jones (1987) suggested an alkalic or picritic ocean island basalt-like parent melt, while Tainton and McKenzie (1994) invoked a primary MORB-derived parental magma. A variation on this model was presented by Burgess and Harte (2004), who envisaged that the megacryst melt probably includes MORB and lithospheric components. Recently, Davies et al. (2001) argued that Cr-poor megacrysts represent high pressure crystallization products of plume-related magmas that in turn reacted with the sub-continental lithosphere to produce kimberlite magmas.

All the non-cognate models imply that the generation of kimberlite magmas followed, and may have been genetically linked to, precursor alkaline magmatism—in other words, that kimberlite genesis is at least a twostage process. These models are further complicated by evidence for Sr-isotopic differences between Cr-poor megacrysts from the same host kimberlite (Hops et al. 1992; Jones 1987; Davies et al. 2001) suggesting possible magma mixing or contamination processes (e.g. Jones 1987; Hops et al. 1992). Moreover, Eggler et al. (1979) suggest that the Cr-rich and Cr-poor megacrysts from the State Line kimberlites crystallized from different parental magmas. If true, the evidence, presented above for Cr-rich megacrysts being far more common inclusions in kimberlites than previously envisaged, raises the prospect that kimberlite magmas represent a most complex devil's brew. Understanding the origin of the megacrysts is clearly central to developing a model for kimberlite genesis. In this paper it is argued that both the Cr-rich and Cr-poor megacrysts are derived from the *host* kimberlite magma as part of a common magmatic process. This considerably simplifies models for kimberlite genesis, and implies that they represent primary mantle-derived melts, consistent with experimental evidence (e.g. Eggler and Wendtland 1979; Dalton and Presnall 1998; Wyllie and Lee 1999) as well as geochemical constraints (Tainton and McKenzie 1994; Le Roex et al. 2003).

The mode of formation of the megacryst suite also has important implications for understanding the origin of sheared peridotites (Harte 1977), which occur as xenoliths in kimberlites with a world-wide distribution [e.g. Lesotho (Nixon and Boyd 1973b), Russia (Boyd 1984), and Canada (Kopylova et al. 1999)]. Nixon and Boyd (1973b) originally suggested that this peridotite suite reflects shearing at the base of the lithosphere—in other words, a mechanical, thermal and chemical boundary, separating this unit from the underlying asthenosphere. This elegant early model has in large measure influenced views on the chemical and isotopic composition of the asthenosphere that persist to the present day.

Ehrenberg (1979) and Gurney and Hart (1980) drew attention to the overlapping compositional fields of corresponding phases from sheared peridotites and the Cr-poor megacryst suite, and suggested that the former rocks had been metasomatised by the megacryst parental magma. Mineral zoning in sheared peridotites has also been ascribed to interaction with the megacryst parental magma (Smith and Boyd 1987, 1989; Griffin et al. 1989; Burgess and Hart 1999, 2004). This interpretation was supported by Moore and Lock (2001), who demonstrated that mineral phases in sheared peridotites from kimberlites in northern Lesotho and the eastern Free State, South Africa, fall within compositional fields that are intermediate between those for corresponding phases in coarse granular lherzolites and the high temperature megacrysts.

A variety of recent proposals for the origin of the sheared peridotites can be viewed as essentially variants of the original Nixon-Boyd concept. Carlson et al. (1999) concluded that they represent asthenospheric material underplated on the lithosphere following destruction of the original deep keel. Kennedy et al. (2002) also envisaged that the sheared peridotites form at the asthenosphere–lithosphere interface. They suggested that the sheared textures are manifestations of the drag applied to the lithosphere by underlying asthenospheric flow. It was proposed that the shearing occurred as high strain-rate events along transient discontinuities, closely analogous to the formation of crustal mylonites. They suggested that these discontinuities provided permeable zones that would preferentially localize partial melts, and thus locally facilitate metasomatism by the megacryst magma. In contrast to these models, Pearson et al. (1999) postulated that the sheared peridotites from the Lac de Gras kimberlites are part of a fertile deeper section of a two-layer lithosphere.

While differing in detail, these models all envisage that the sheared peridotites in some manner reflect vertical mantle layering-in other words, vertical variations in the thermal, chemical and mechanical properties of the mantle. However, this interpretation was challenged by Mercier (1979), who demonstrated that the sheared textures are transient rather than steady-state features. He proposed that the shearing occurred in a narrow (50 m) thermal aureole surrounding the conduit to the kimberlite magma prior to eruption to the surface. This requires that the sheared peridotites represent a very localized chemical and mechanical perturbation of the mantle associated with the kimberlite magma. If correct, this implies that the sheared peridotites do not provide evidence for vertical mantle layering, and raises serious questions regarding the use of these rocks to infer asthenospheric compositions. Given these radically different models for the processes responsible for the deformation of the sheared peridotites, and the broad consensus that they have been metasomatised by the magma that crystallized the megacrysts, an understanding of the origin of the latter suite is clearly critical to the interpretation of compositional variations in the upper mantle.

The conclusion drawn in the present paper—that the megacrysts crystallized from the host kimberlite magma—is consistent with Mercier's model. Metasomatism of the sheared peridotites is therefore inferred to have occurred within the narrow thermal aureole surrounding a kimberlite magma chamber in the upper mantle.

# Mineralogical and textural characteristics of Cr-rich and Cr-poor megacryst suites

The mineralogical and textural characteristics of the Crpoor and Cr-rich megacrysts suites from different localities have been described in varying degrees of detail. Salient observations are summarized below.

Both Cr-poor and Cr-rich megacrysts occur either as single crystals, or intergrown with, enclosed by, or enclosing, other phases of the same suite. Co-existing mineral phases in the low-Cr-suite show that garnet, clinopyroxene, orthopyroxene and olivine crystallized together over a wide range in temperatures (Gurney et al. 1979; Eggler et al. 1979). With very rare exceptions, exsolution textures are absent in both suites at a microscopic scale, indicating that they did not experience post-crystallization thermal equilibrium in the mantle. However, sub-microscopic exsolution textures reported by McCallister et al. (1979) were interpreted to reflect cooling during rapid kimberlite ascent to the surface.

In the Colorado-Wyoming kimberlites (Eggler et al. 1979), both Cr-rich and Cr-poor garnet megacrysts are typically fractured. The former commonly have weakly developed shear planes. The Cr-rich clinopyroxenes are typically angular fragments of originally larger crystals. Partial alteration along cleavages or parting planes appears to have accompanied the shattering of these clinopyroxenes. The Colorado–Wyoming Cr-poor clinopyroxene megacrysts, in contrast, tend to be wellrounded and ellipsoidal to ovoid, although one or two cleavage planes may be present. Orthopyroxenes from both suites are less rounded than other megacrysts phases. This was inferred to reflect fragmentation along cleavages during kimberlite emplacement. Single crystal olivine megacrysts are rare in the Wyoming-Colorado pipes, but dunite nodules with an aggregate texture are inferred to be recrystallized olivine megacrysts (Eggler et al. 1979). These includes both Cr-rich and Cr-poor compositions.

The Granny Smith diopside megacrysts described by Boyd et al. (1984) are normally single crystals, around 2 cm in size, but may attain 10–15 cm. Many have been deformed, resulting in porphyroclasts of strained diopside in a recrystallized matrix, while some have coarse polycrystalline textures. A considerable proportion of the Granny Smith diopsides are intergrown with phlogopite and ilmenite and, less commonly, rutile. Irregularly shaped inclusions of fosterite, up to 1 cm in size, occur as very rare inclusions in the Granny Smith diopsides.

Mosaic-textured dunites, inferred to be part of the Cr-poor megacryst suite, are also more common than single crystal olivines at the Hamilton Branch kimberlite (Schulze 1984). However, megacryst olivines at Monastery do not show signs of recrystallization (Gurney et al. 1979). Fe-rich dunite xenoliths, which constitute an estimated 2% of the mantle-derived inclusions from Bultfontein, are interpreted to be related to the Cr-poor megacryst suite (Dawson et al. 1981). All are recrystallized to a greater or lesser degree, with textures varying from porphyroclastic to mosaic (terminology of Harte 1977).

Ilmenite occurs as both single crystals and polycrystalline aggregates in the Hamilton Branch and Monastery kimberlite. Polycrystalline ilmenites from the latter locality are systematically more Mg-rich than single crystals (Schulze 1984; Haggerty et al. 1979). In contrast, ilmenites from the Frank Smith kimberlite are virtually all polygranular (Pasteris et al. 1979).

The Cr-rich megacrysts described by Kharkiv (1995) from the Verkhnemunskoye kimberlites (Republic of Sakha) are characterized by the presence of branching stringers that are interpreted to reflect partial melting of the host. It is not clear whether similar textures also characterize the Cr-poor megacryst suites from these pipes.

Meyer et al. (1979) described a "unique" Cr-poor enstatite megacryst from the Weltevreden floors (South Africa) that has inclusions of Cr-poor orange pyropealmandine, which in turn enclose rounded pink Cr-rich garnets. The latter may contain inclusions of Cr-diopside, chromite and Cr-rich ilmenite. Abundant rounded ilmenites are associated with the narrow gradational chemical boundary zone between the two garnets, often increasing in size away from the pink garnet inclusion. This ilmenite-rich zone also contains diopside and olivine, with individual grains of these respective phases displaying optical continuity. Irregular patches of calcite, Ti-phlogopite and serpentine occur at the contact between the orthopyroxene host and the enclosed orange garnet. Ilmenite is a common inclusion within the host enstatite, varying in form from irregular blebs to angular lamellae that resemble those occurring as intergrowths with pyroxenes.

Two mantle-derived inclusions with co-existing red and orange garnets, recovered from the Kaalvallei kimberlite, South Africa by WL Griffin, were kindly made available for the present study. These samples (designated KLV-1 and KLV-2 respectively) display many of the petrographic features of the Weltevreden megacryst described by Meyer et al. (1979), although they differ in being pre-dominantly composed of garnets. In KLV-1, the orange garnet encloses or partially encloses a red garnet, which in turn contains inclusions of olivine, orthopyroxene and clinopyroxene. The boundary zone between the two garnets varies from 50 µm to 500 µm in width. White rounded inclusions (typically  $\sim 200 \ \mu m$  in diameter) in the red garnet may be a carbonate, but have not been positively identified. The interface between the two garnet color varieties is marked by a necklace of small (generally  $30-50 \mu m$ ) – irregular ilmenite blebs. Areas dominated by the red garnet have a spongy appearance, reflecting either alteration or incipient partial melting along the network of fractures traversing these grains.

The Kaalvallei sample KLV–2 is petrographically similar to KLV-1, although chemical evidence, discussed below, shows that the red garnet in this specimen consists of two compositionally distinct phases. Irregular-shaped ilmenite grains (up to 200  $\mu$ m in diameter) occur both enclosed by the red garnet, and irregularly along the contact with the orange garnet.

Lawless et al. (1979) and Wyatt and Lawless (1984) have described a suite of deformed polymict peridotites from the de Beers and Bultfontein kimberlites (Kimberley, South Africa.) The phase assemblage in these rocks includes megacrysts of garnet, and clinopyroxene. The latter may enclose, or be enclosed by, orthopyroxene. Phlogopite, ilmenite, rutile and chromite are ubiquitous inclusions within the clinopyroxeneorthopyroxene megacrysts, chiefly within the orthopyroxene-rich areas. Polyphase inclusions, interpreted to represent kimberlitic liquids, have been described from a number of Cr-poor megacrysts from the Monastery and Hamilton Branch kimberlites (Gurney et al. 1979; Schulze 1984). Haggerty et al. (1979) note that a small proportion (5%) of ilmenites from the Monastery kimberlite contain trapped ovoid inclusions of calcite + pyrrhotite + pentlandite. One of the Fe-rich dunites from Bultfontein, described by Dawson et al. (1981), contains serpentine-calcite-apatite-magnetite segregations, which were also interpreted to represent trapped liquids.

Van Achterberg et al. (2002) describe inclusions, varying from carbonatitic to kimberlitic in composition, in megacrystic Cr-diopsides recovered from pipes in the Slave province, Canada. They interpret the inclusions to represent the crystallization products of liquids trapped shortly prior to kimberlite eruption. Van Achterberg et al. (2002) infer a lherzolitic paragenesis for the Slave Province clinopyroxenes, as they enclose orthopyroxene, and garnet. However, textural characteristics of the megacrystic Slave clinopyroxenes, coupled with their chemistry, discussed in the following section, link them to the Cr-poor megacryst suite.

# Chemical characteristics of the Cr-poor and Cr-rich megacryst suites

Gurney et al. (1979) showed that the Cr-poor silicate megacryst phases at Monastery formed a co-genetic suite, characterized by a wide range in compositions, reflecting crystallisation over a range of temperature (1,400–950°C) under essentially isobaric conditions (45 kbar). These authors demonstrated that ilmenite is a relatively late-crystallizing phase at Monastery. Cr-poor megacrysts at Hamilton Branch kimberlite (Schulze 1984) and Jagersfontein (Hops et al. 1989, 1992) were also inferred to have crystallized over a range of temperatures under isobaric conditions (55 and 50 kbar respectively). The majority of megacrysts from Thaba Putsoa, Lesotho, also crystallized over a very limited pressure range (Nixon and Boyd 1973a).

Table 1 lists the ranges in composition of Cr-rich clinopyroxene and garnet megacrysts from different localities relative to the Cr-poor suite from the State line kimberlites. Included in Table 1 are compositional ranges of the Granny Smith diopsides, garnets enclosed by the Weltevreden orthopyroxene megacryst, and also clinopyroxene megacrysts associated with two polymict peridotites, JJG 513 from the de Beers kimberlite, and JJG 1414 from the Bultfontein pipe (Lawless et al. 1979).

Compositions of Cr-poor and Cr-rich megacryst suites from the State Line kimberlites are illustrated in Fig. 2. For reference, this diagram also shows the fields for minerals from coarse granular and sheared peridotites from the Lesotho kimberlites (Nixon and Boyd 1973b, c). Included in the diagram are the megacrystalline Slave clinopyroxene and associated phases (Van Achterberg et al. 2002), and garnets from the Weltevreden and Kaal Vallei nodules (KLV-1 and KLV-2).

In general, the Cr-rich megacryst suite partially overlaps the range in compositions for granular and sheared peridotites (Fig. 2). It is characterized by higher Mg# (100 Mg/(Mg+Fe)) (atomic proportions) and a range with more Cr-rich compositions relative to the Crpoor suite. Clinopyoxenes in the two suites show similar levels of TiO<sub>2</sub>, but the more sub-calcic clinopyroxenes are not represented in the Cr-rich suite. Garnets from the Cr-poor and Cr-rich suites are also characterized by similar ranges in TiO<sub>2</sub> (Table 1). The tie-lines in Fig. 2 connect compositions of the Cr-rich megacryst host and inclusions (data from Eggler et al. 1979). These tie-lines

Table 1 Comparison of clinopyroxenes and garnets from Cr-rich megacryst suits from different kimberlites with the state line Cr-poor megacryst suite

Kimberlite	Ca#	Mg#	TiO <sub>2</sub>	Cr <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>	CaO	References
Clinopyroxenes								
Cr-poor								
State line	36–47	82.6-90.8	0.18 - 0.48	0.08 - 1.0	1.0 - 1.7			Eggler et al. (1979)
Cr-rich								
State line	41–48	92.0-93.1	0.09-0.22	0.83 - 2.40	0.9–1.6			Eggler et al. (1979)
Orapa	43.6-46.9	86.1–93.8	n.d	0.71 - 2.88	n.d			Shee and Gurney (1979)
Weltevreden	42.8	90.5	0.33	2.5	2.06	2.13	17.8	Meyer et al. (1979)
Granny smith suite:								•
Kimberley and								
Jagersfontein	>45	>90	0.2-0.35	0.5-3.0	1.29-2.04	0.85-1.89		Boyd et al. (1984)
Polymict peridotites:								
Bultfontein(JJG1414)	43-45.1	91.4–92.3	0.27 - 0.46	2.32 - 2.70	2.37 - 2.77	1.64-2.13		Lawless et al. (1979)
de Beers (JJG 513)	41.3-48.4	88.7-92.0	0.13-1.22	0.98-3.24	0.91-2.98	0.51-3.03		Lawless et al. (1979)
Garnets								
Cr-poor								
State line	13-22	68.3-83.6	0.23-1.3	0.03-4.8	0.0-0.12			Eggler et al. (1979)
Weltevreden	20	81.0	$0.96 \pm 11$	$0.33 \pm 0.44$	$0.12\pm0.08$	$21.5\pm0.49$	$4.05\pm0.80$	Meyer et al. (1979)
Cr-rich								
State line	14–27	81.8-84.1	0.22-0.94	6.3-13.0	0.0009			Eggler et al. (1979)
Weltevreden	32	81.8	$0.71\pm0.21$	$9.9\pm0.56$	$0.1\pm0.06$	$15.5\pm0.48$	$8.36\pm0.61$	Meyer et al. (1979)

indicate that the relatively iron-rich orthopyroxenes and garnets (i.e. lower temperature, relatively evolved compositions) co-exist with the most sub-calcic and Mg-rich clinopyroxenes. It follows that the more calcic (i.e. lower temperature) clinopyroxenes in the Cr-rich suite did not crystallize in equilibrium with garnet and orthopyroxene. It is therefore difficult to estimate the range in equilibrium pressures and temperatures for this suite with any confidence.

Cr-poor megacryst phases from many other kimberlites with a wide geographic distribution show qualitatively comparable crystallization trends to those recorded at Monastery (Gurney et al. 1979) and the State Line kimberlites (Eggler et al. 1979). However, there are typically differences in the chemical fields for Cr-poor megacryst suites from different pipes (Eggler et al. 1979; Schulze 1987). The limited available data, discussed below, indicates that there are also systematic differences between the chemical fields for Cr-rich megacryst populations from different kimberlites. The distinction between the compositional fields of Cr-rich and Cr-poor megacryst populations therefore refers to individual kimberlites. It should be noted further that in the Sloan-Nix pipes from the Colorado–Wyoming



Fig. 2 Composition of megacrysts and garnet lherzolites phases plotted in a portion of the Ca–Mg–Fe ternary diagram (atomic proportions; total Fe as FeO). *Diagonal lined* and *stippled fields* denote Cr-rich and Cr-poor megacrysts from the State Line kimberlites, USA, respectively. Tie lines connect compositions of Cr-rich megacryst host and inclusions (microprobe data from Eggler et al. 1979). *Dashed lines* outline fields for granular mantle peridotites (GP) and sheared peridotites (SP) (data from Eggler et al. 1979). *Solid triangle:* Pink garnet, Weltevreden orthopyroxene megacryst; *open triangle:* orange garnet, Weltevreden orthopyroxene megacryst (microprobe data from Meyer et al. 1979), *Inverted solid triangles:* clinopyroxene and associated phases from van Achterbergh et al. 2002; *Open circles:* Garnets from Kalvallei nodule KLV-1; *Filled circles:* Garnets from KLV-2

cluster, there are anomalous clinopyroxene megacrysts with compositions intermediate between the Cr-poor and Cr-rich suites (Eggler et al. 1979). This suggests that the chemical distinction between the two populations may not always be clear cut.

Figure 3 illustrates the fields for clinopyroxene megacrysts from the Mukarob 1 and 2 pipes from the Gibeon kimberlite cluster in Namibia (data from Mitchell 1987). These data indicate that chemically distinct Cr-rich and Cr-poor suites are present in both pipes. Included in Fig. 3 are the compositions of Cr-rich and Cr-poor megacrysts from the Orapa kimberlite in Botswana (Shee and Gurney 1979). Note that in these three pipes, both the Cr-poor and Cr-rich suites define distinctive chemical fields that show only partial overlap.

Compositions of the Granny Smith clinopyroxenes described by Boyd et al. (1984) partially overlap the fields for Cr-rich megacrysts from the Mukarob 1 and 2 kimberlites (Fig. 4). This is consistent with the view expressed by these authors that the Granny Smith suite is chemically equivalent to the Cr-rich megacryst suite. Ilmenites associated with the Granny Smith clinopyroxenes are Cr-rich  $(1-3\% \text{ Cr}_2\text{O}_3, \text{ Boyd et al. } 1984)$ relative to the Cr-poor ilmenite suite from Monastery (typically < 0.4% Cr<sub>2</sub>O<sub>3</sub>, Gurney et al. 1979; Moore et al. 1992). This in turn links relatively Cr-rich ilmenite megacrysts in kimberlites to the Cr-rich suite. An olivine inclusion in a Granny Smith diopsides with a composition of Fo<sub>90.6</sub> (Boyd et al. 1984) falls within the range for olivines from the Cr-rich megacryst suite from the Colorado-Wyoming kimberlites (Eggler et al. 1979).

Compositions of the pink and orange garnets associated with the Weltevreden orthopyroxene megacryst, described by Meyer et al. (1979), are shown in Fig. 2 and listed in Table 1. These plot close to the fields for the Cr-



Fig. 3 Clinopyroxene megacrysts from Mukarob 1 and Mukorob 2 plotted in a portion of the Ca–Mg–Fe ternary diagram (atomic proportions; total Fe as FeO). (microprobe data from Mitchell, 1987). *Diagonal lined* and *stippled fields* are for the Mukorob 1 and 2 Cr-rich and Cr-poor suites respectively. *Filled triangles:* Orapa Cr-rich clinopyroxene megacrysts; *Filled circle:* Orapa Cr-poor clinopyroxene megacrysts (microprobe data from Shee and Gurney, 1979). Note that both Cr-rich and Cr-poor megacryst suites from each of the three kimberlites define distinctive chemical fields, with only partial overlap



**Fig. 4** Compositions of Granny Smith clinopyroxene megacrysts (*open triangles*) (microprobe data from Boyd et al. 1984) relative to the fields for Cr-rich clinopyroxene megacrysts from Mukarob 1 and 2 (data from Fig. 2), plotted in a portion of the Ca–Mg–Fe ternary diagram (atomic proportions; total Fe as FeO)

rich and Cr-poor garnet megacryst suites respectively from the State Line kimberlites, and well away from the fields for garnets from sheared and granular lherzolites. These two garnets in the Weltevreden orthopyroxene megacryst are separated by a narrow zone with abundant (20–30%) rounded globular ilmenites. Within this zone, the garnets show marked chemical zoning with a decrease in Cr/(Cr + Al) and Ca/(Ca + Mg) across the interface from the pink to the orange garnet, rather than an abrupt compositional break. This sense of garnet zonation is accompanied by a marked decrease in Cr contents of the associated globular ilmenites across the chemical interface. The ilmenites enclosed by the pink garnet, and within the garnet transition zone, are all Crrich  $(1.9-13\% \text{ Cr}_2\text{O}_3)$  relative to those  $(0.2\% \text{ Cr}_2\text{O}_3)$ within the surrounding Cr-poor orthopyroxene megacryst (Meyer et al. 1979). This provides further evidence for linking Cr-rich ilmenites to the Cr-rich megacryst suite.

Major-element contents of the two Kaalvallei garnet nodules (KLV-1 and KLV-2 were analyzed by NJ Pearson on a CAMEBAX SX50 electron microprobe at the GEMOC ARC National Key Centre, Macquarie University (http://www.es.mq.edu.au/GEMOC/An-Methods/anmeth.html). The analyses were performed with accelerating voltage of 15 keV, a beam current of 20 nA, and a beam size of less than 3 µm. The PAP matrix correction routine has been applied (Pouchou and Pichoir, 1984).

Representative analysis of phases in the Kaalvallei garnet nodules KLV-1 and KLV-2 are listed in Table 2, and included in Fig. 2. In both samples, the red garnet plots close to the field for Cr-rich megacrysts from the State Line kimberlites, while the orange garnets fall close to the field for the Cr-poor suite from these pipes. This chemical evidence indicates that the red and orange garnets in the Kaalvallei samples are respectively related to the Cr-rich and Cr-poor megacryst suites respectively. This conclusion is supported by the very coarse-grained textures of the two Kaalvallei samples, which distinguishes them from the sheared peridotites suite. A feature of both KLV-1 and KLV-2 is that there are marked differences in composition of both the red and orange garnets on the scale of each hand specimen (Table 2).

Scanning electron microprobe (EMP) images (Ca K $\alpha$  and Cr K $\alpha$ ) of KLV-1 show that the red and orange garnets are separated by a narrow (50–500 µm) boundary zone. Fig. 5a–d illustrate two lines of spot microprobe analyses that traverse this boundary, from the red to the orange garnet, with analytical positions approximately 400 µm apart. Along both traverse lines, Cr/(Cr+Al), Ca/(Ca+Mg) and Mg/(Mg+Ca) show sharp decreases across the boundary between the two garnets. This is closely analogous to the results reported by Meyer et al. (1979) from their study of the Weltevreden nodule. The ilmenites that occur at the contact between the two garnets are all relatively Cr-rich (3.7–4.9% Cr<sub>2</sub>O<sub>3</sub>, Table 2).

Within the portion of KLV-2 covered by the scanning EMP images, the red garnet contains irregular, patchily distributed Cr-rich and Ca-poor cores that are compositionally equivalent to G10 garnets (Table 2). These are surrounded by a more calcic, red garnet, with lower Cr contents, which in turn is separated from the orange garnet by a steep compositional gradient. Chemical differences between the red and orange garnets in this specimen are qualitatively similar to those documented for KLV-1 (Fig. 2; Table 2) The ilmenites enclosed by and concentrated along the margin of the red garnet are exceptionally Cr-rich (4.5-8.3% Cr<sub>2</sub>O<sub>3</sub>, Table 2.) This reinforces the link between Cr-rich ilmenites and the Cr-rich megacryst suite.

The Kaalvallei megacrysts are important because they show mineralogical characteristics and chemical zoning features that are very similar to those described for the Weltevreden sample. The latter can therefore no longer be considered "unique" (Meyer et al. 1979). The composite samples from Weltevreden and Kaalvallei also provide further evidence that both Cr-rich and Crpoor megacrysts are present in many kimberlites and further, that they can co-exist on the scale of individual hand specimens.

The megacrysts phases associated with the polymict peridotites are Ti-and Cr-rich, and have Mg# and Ca#, which fall within the range for the State Line Cr-rich megacryst suite of Eggler et al. (1979) (Table 1).

The megacrystic Slave clinopyroxene described by Van Achterberg et al. (2002) is chemically similar to those of the Cr-poor megacryst suite, and plots outside the field for coarse granular lherzolites (Fig. 2). The Ca# of the clinopyroxene (42.9) is low relative to compositions typical of coarse peridotites (e.g. Nixon and Boyd 1973b,c, but well within the range typical of the megacryst suite (e.g. Nixon and Boyd 1973a; Gurney et al., 1979). Garnets and orthopyroxenes enclosed by the Slave megacrystic Cr-diopsides plot close to and within the fields for Cr-poor megacryst, and away from those for coarse granular lherzolites respectively (Fig. 2). **Fig. 5** a–d. Spot microprobe analyses on traverses from a red (Cr-rich) to orange (Cr-poor) garnet in the composite garnet nodule KLV-1 from the Kaalvallei kimberlite. Average distance between analytical positions is approximately 400 µm





Kaalvallei Gt1, Traverse 1

0.40

0.35

0.30

0.25

0.20

0.15 0.10

0.05

0.84 0.82

0.80 Batio 0.78

Ratio

These chemical relationships all point to an affinity with the Cr-poor megacryst suite rather than coarse granular lherzolites.

A number of detailed studies of chemical characteristics of ilmenite suites from different kimberlites are relevant to the origin of the Cr-rich and Cr-poor megacryst suites. In the Monastery kimberlite, the dominant ilmenite population in concentrates is Cr-poor (generally < 0.4% Cr<sub>2</sub>O<sub>3</sub>) over a range of MgO contents (6.5–12%). The pipe also contains two subordinate, chemically discrete ilmenite populations, with similar elevated ranges in  $Cr_2O_3$  (0.6–1.2%  $Cr_2O_3$ ), but separated by a compositional hiatus between Mg# 32–36 (Jakob 1977; Moore et al. 1992). The majority of the ilmenites in the Mg-poorer of these latter two populations are intergrown with zircon (Moore et al. 1992).

Figure 6a-c illustrate ilmenite populations in kimberlites from the Kokong, Molopo-Tsabong and Orapa pipe clusters in Botswana (data from Moore and Lock 2001.) (See Fig. 1 for their locations). There are marked differences in the chemical fields of the ilmenite suites from these three pipes. KN70, from the Kokong kim470

	SiO <sub>2</sub>	TiO <sub>2</sub>	$Al_2O_3$	Cr <sub>2</sub> O <sub>3</sub>	FeO(tot)	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	NiO	Total
KLV-1												
Red garnet core, traverse 1	41.19	0.56	15.51	9.94	6.52	0.14	19.74	5.77	0.09	0.01	0.02	99.49
Red garnet core, traverse 2	40.60	0.64	15.58	9.65	8.39	0.27	18.15	5.98	0.09	0.01	0.00	99.35
Red garnet average $(n = 12)$	40.34	0.64	14.83	10.96	7.00	0.16	19.90	6.24	0.05	0.00	0.00	100.12
Orange garnet core, Traverse 1	41.52	0.58	21.50	0.73	12.37	0.47	18.53	4.51	0.07	0.01	0.02	100.30
Orange garnet core, traverse 2	41.60	0.54	21.62	0.65	12.38	0.49	18.34	4.48	0.07	0.00	0.01	100.19
Orange garnet core average $(n = 12)$	40.77	0.58	20.70	2.13	11.12	0.40	19.48	4.83	0.06	0.00	0.00	100.07
Ilmenite (interface between garnets)	0.04	46.90	1.02	3.71	37.75	0.36	7.46	0.02	0.01	0.00	0.09	97.36
Ilmenite (interface between garnets)	0.08	44.62	0.30	4.87	39.60	0.22	6.64	0.03	0.01	0.02	0.09	96.48
Ilmenite (interface between garnets)	0.05	47.20	0.90	3.92	35.68	0.32	8.71	0.01	0.02	0.04	0.11	96.95
KLV-2												
Cr-rich (G10) core to red garnet	42.03	0.01	17.45	8.30	5.94	0.19	22.55	3.27	0.00	0.00	0.12	99.84
Red garnet average $(n=2)$	40.59	0.60	15.03	10.60	7.50	0.18	19.64	6.21	0.08	0.00	0.00	100.43
Orange garnet core average $(n=6)$	41.02	0.50	20.23	2.70	11.45	0.40	19.34	4.46	0.06	0.00	0.00	100.16
Ilmenite (interface between garnets)	0.06	48.25	1.54	4.23	36.02	0.32	8.81	0.07	0.00	0.00	0.00	99.30
Ilmenite (interface between garnets)	0.08	45.92	0.45	4.23	34.10	0.05	9.77	0.02	0.01	0.00	0.00	98.68

berlite cluster in western Botswana has a very low diamond grade of the order of 1ct/100t, and is non-economic. The ilmenites from this kimberlite define a single population, characterized by a continuous, hyperbolic variation in MgO and  $Cr_2O_3$  (Fig. 6a). While the Mgand Cr-poor limb could be considered to be representative of the Cr-poor megacrysts, the Mg- and Cr-rich (up to 4%  $Cr_2O_3$ ) limb has a chemical affinity with ilmenites shown to be associated with the Cr-rich megacryst suite. Chemical characteristics of the KN 70 ilmenites therefore indicate a compositional continuum between the Cr-rich and Cr-poor suites at this locality.

M4 is a very low-grade pipe from the Molopo-Tsabong cluster from southwestern Botswana. A majority of the concentrate ilmenites from this pipe fall within one of two discrete compositional fields. One is relatively Cr-poor (generally < 0.5% Cr<sub>2</sub>O<sub>3</sub>), suggesting that it is related to the Cr-poor megacryst suite. The second is relatively Cr-rich (generally > 1.5% Cr<sub>2</sub>O<sub>3</sub>), pointing to an affinity with Cr-rich megacrysts. A few ilmenites have compositions that fall outside the fields of these two dominant populations (Fig. 6b). Ilmenites from many of the associated Molopo-Tsabong kimberlites define comparable paired Cr-poor and Cr-rich populations (Moore 1987).

The AK1 (Orapa) kimberlite from central Botswana, with a grade of 0.83ct/t, is currently being mined (de Beers Annual Report 2000). Ilmenites megacrysts (grains > 10 mm in size) and macrocrysts (grains ranging from 0.5 mm–10 mm in size) from this pipe are characterized by relatively high Cr contents (1–4%  $Cr_2O_3$ ) at rather low MgO contents (7–10% MgO) compared to the populations from the Kokong and Molopo-Tsabong kimberlites (Fig. 6c). These chemical characteristics link the AK1 ilmenites to the Cr-rich megacryst suite. It should be noted that a rare group of

Fig. 6 Microprobe analyses of ilmenites from kimberlites in three pipe clusters in Botswana (data from Moore and Lock 2001) a Ilmenites-KN-70 pipe (Kokong cluster). **b** Ilmenites—M4 pipe (Tsabong-Molopo cluster). c Ilmenites—AK1 (Orapa) pipe (Orapa cluster). d Ilmenites from the BK4, BK7, BK15 and DK1 (Letlhakane) kimberlites from the Orapa pipe cluster, Botswana. Note that these four pipes, together with AK1 (Orapa) (Fig. 6c) are characterized by different ilmenite compositional fields (chemical fingerprints), although there is partial overlap of these respective fields



ilmenites from AK1, characterized by very low Cr contents (generally <0.2% Cr<sub>2</sub>O<sub>3</sub>), and MgO ranging from approximately 9.5–11.5% (Fig. 6c), occur in eclogite xenoliths (Tollo 1982), and are thus unrelated to megacrysts. Ilmenite megacryst populations from other kimberlites of the Orapa cluster are each characterized by diagnostic, chemically distinct fields, which are also relatively Cr<sub>2</sub>O<sub>3</sub>-rich (predominantly in the range 1–4% Cr<sub>2</sub>O<sub>3</sub>; Fig. 6c,d). This also points to a chemical affinity with the Cr-rich, rather than the Cr-poor, megacryst suite.

The ilmenite data discussed therefore provides further evidence that the Cr-poor and Cr-rich megacryst suites co-exist in kimberlites with a wide geographic distribution. The silicate phases related to the latter population may have been overlooked at some localities because of their similarity in appearance to phases in garnet lherzolites.

## Evidence for a cognate origin for the Cr-rich and Cr-poor megacryst suites

It is necessary to consider the possible origins of the two megacryst suites. Several lines of field, petrographic, chemical and experimental evidence provide overwhelming evidence that both crystallized from the *host* kimberlite magma.

The following considerations argue for a cognate origin for the Cr-poor suite:

- 1. A number of Cr-poor megacrysts from the Monastery kimberlite contain poly-mineralic inclusions with bulk compositions that are very similar to the host Quarry kimberlite. These are interpreted to represent liquids trapped at the time of megacryst formation (Jacob 1977; Gurney et al. 1979). Polycrystalline inclusions, interpreted to represent high pressure trapped kimberlitic liquids, have also been reported from Cr-poor megacrysts from the Hamilton Branch kimberlite, USA (Schulze 1984). Similarly, Van Achterberg et al. (2002) describe inclusions, varying from carbonatitic to kimberlitic in composition, in megacrystic Cr-diopsides recovered from pipes in the Slave province, Canada. They interpret the inclusions to represent the crystallization products of liquids trapped shortly prior to kimberlite eruption. These observations demonstrate that kimberlitic megacrysts with a wide geographic distribution contain inclusions derived from trapped kimberlitic liquids.
- 2. Various authors (e.g. Harte 1983; Jones 1987; Moore et al. 1992; Davies et al. 2001) have proposed that kimberlitic Cr-poor megacrysts are non-cognate, and crystallized from parental magmas such as alkali basalts or basanites. However, none of these postulated parental magmas has been reported as occurring as inclusions in megacrysts. Neither have these alkaline rock varieties been found in southern Africa

as intrusions associated with kimberlites, nor as xenoliths entrained by kimberlite. There is, in other words, no field or petrographic evidence linking megacrysts to any of these alkaline magmas.

- 3. There are systematic differences in the compositional fields defined by Cr-poor megacryst suites from different kimberlites, including those from the same pipe cluster (e.g. Eggler et al. 1979; Mitchell 1987; Schulze 1987; Griffin et al. 1997; Kostrovitsky and Mitchell 1995). Examples of such differences are illustrated in Figs. 2 and 3). Griffin et al. (1997), Lee (1993) and Moore and Lock (2001) note that chemical fields for ilmenite, which is often a late-crystallizing megacryst phase (Gurney et al. 1979) may vary significantly between different pipe clusters, as well as between individual kimberlites from the same cluster. Thus, for example, there are clear differences between the ilmenite compositional fields for different kimberlite clusters in Botswana (Fig. 6a-c). There are also differences in the compositional fields for different pipes within the same cluster, as illustrated in Fig. 6c,d, for five kimberlites from the Orapa cluster. Such distinctive compositional fields provide "chemical fingerprints" that make ilmenite a particularly valuable tool for recognizing undiscovered pipes during prospecting operations. It is very difficult to account for the unique chemical characteristics of ilmenite populations in individual kimberlites from the same pipe cluster in terms of random sampling of mantle material. Rather, this relationship is more readily explained by a genetic link between the host kimberlite magma and ilmenite, and by extension, the Crpoor megacryst suite.
- 4. The majority of diamonds in kimberlites appear to be derived from either a peridotitic or an eclogitic paragenesis, and are considerably older that the host kimberlite (Richardson et al. 1984; Richardson 1986). They are thus mantle xenocrysts sampled at the time of kimberlite emplacement. Economic Group I kimberlites tend to be characterized by relatively Mg- and Cr-rich (less evolved) ilmenite suites relative to those in sub-economic pipes (Gurney and Zweistra 1995; Fig. 6a-d), although there are many exceptions to these generalizations. If ilmenite crystallised from a precursor magma, such as a proto-kimberlite, or other alkaline magma, and was subsequently randomly entrained by both economic and low-grade kimberlites, the link between chemistry of the phase and diamond grade is not readily explained. The relationships are more readily explained if ilmenite-and thus the megacryst suite-crystallized from the *host* kimberlite magma.
- 5. The chemistry of picroilmenites associated with kimberlites point to crystallization from a Mg- and Cr-rich parent, consistent with derivation from the host kimberlite. Ilmenite populations in the various alkali basalt magmas that have been proposed as parental to the megacryst suite never extend to the Mg- and Cr-rich compositions found in kimberlites.

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- 6. There are typically isotopic differences between Crpoor megacrysts and the host kimberlite (Kramers et al. 1981; Davies et al. 2001) as well as between individual megacrysts from the same kimberlite (Hops et al. 1992; Davies et al. 2001). Nevertheless, on a broader scale in southern Africa, megacrysts appear to broadly mirror the isotopic signature of the host kimberlite. Thus, Smith et al. (1995) showed that megacrysts from the Group II Lace kimberlite have Sr and Nd isotopic compositions that plot close to the field for isotopically distinctive Group II kimberlites of Smith (1983), which are broadly equivalent to the micaceous variety of Wagner (1914). In the Frank Smith kimberlite, which has a Sr/Nd isotopic signature intermediate between the fields for the Group I and Group II suites, megacrysts have isotopic compositions closely similar to that of the host kimberlite. The overall sympathetic variation in isotopic compositions of megacrysts and host kimberlite in southern Africa argues strongly for a genetic link. Factors that could account for the minor isotopic differences between megacryst and host, as well as between different megacrysts from the same kimberlite, are considered in a later section.
- 7. It has been demonstrated that kimberlite megacryst suites represent the products of isobaric crystallization over a wide range of temperatures (Gurney et al. 1979; Schulze 1984; Hops et al. 1989). The lack of microscopic scale exsolution textures in the megacryst suite, and failure to re-equilibrate to a constant ambient mantle temperature requires that the parent magma was present shortly before, or at the time of entrainment, by the host kimberlite. Given the common occurrence of inclusions of kimberlite composition in megacrysts, and lack of petrographic or field evidence for the presence of other alkali magmas, an a priori assumption must be that the kimberlite represents the parent liquid.
- 8. Sub-microscopic exsolution textures in clinopyroxene megacrysts from the Thaba Putsoa kimberlite (Lesotho) reflect thermal re-equilibration following entrainment to the surface by the host kimberlite. Prior to entrainment, the crystal was apparently in thermal equilibrium at approximately 1,300°C (McCallister et al. 1979), implying close proximity to the parent magma. Again, the a priori assumption must be that this was the host kimberlite magma.
- 9. Chemical zonation in garnets in sheared peridotites has been ascribed to interaction with the magma that crystallized the megacryst suite. However, in order to maintain the chemical gradients, it is necessary that this magma was present immediately prior to, or at the time of, entrainment, of the peridotite xenolith (Smith and Boyd 1987, 1989; Griffin et al. 1989). Again, the a priori assumption must be that this magma was the host kimberlite. By extension, the

latter was therefore also parental to the megacryst suite.

Several lines of evidence indicate that there is also a close genetic link between Cr-rich megacryst suites and the host kimberlite:

- 1. The common association of the Granny Smith diopsides with ilmenite and phlogopite suggests crystallization from a Ti- and K-rich parent liquid. The relatively high Ti contents of garnets from the Cr-rich suite (Table 1) also points to a Ti-rich parental liquid.
- 2. The limited available evidence suggests that, as is the case for the Cr-poor megacryst suite, there are differences in the chemical fields defined by Cr-rich megacrysts from different kimberlites, including those from the same cluster. This is illustrated in Fig. 3, which shows the fields for Cr-rich megacrysts from the Orapa kimberlite in Botswana, and two pipes from the Gibeon field, Namibia. Compositions of Crrich megacryst phases from different localities illustrated in Fig. 2 are consistent with this conclusion. It is also supported by the diagnostic chemical fingerprints of Cr-rich ilmenite populations in individual kimberlites in the Orapa field (Fig. 6c, d). As in the case of the Cr-poor megacryst suite, such differences are more readily explained in terms of a genetic link to the *host* kimberlite than random sampling of the mantle during ascent (Moore and Lock 2001).
- 3. Worldwide, megacrysts are not confined to kimberlites, and are relatively common in alkali basalts (Schulze 1987). However, the latter volcanics are characterized by a very different megacryst phase assemblage, which includes Al-rich augite, Al-orthopyroxene, and Ti-rich amphibole, considered to be genetically linked to the host magma (Schulze 1987). Any model for the origin of the discrete megacrysts should provide an explanation for the mineralogical differences between the suites in kimberlites and alkali basalts. High pressure experiments on liquids of alkali basalt compositions show that Al-augite and orthopyroxene and Ti-amphibole are liquidus phases (Schulze 1987). Experimental evidence (Eggler and Wendlandt 1979; Dalton and Presnell 1998) indicates that the early crystallizing kimberlite Cr-rich megacrysts (garnet, clinopyroxene, orthopyroxene and olivine) are high pressure liquidus or near-liquidus phases in liquids of kimberlitic composition. Compositions of the Cr-rich megacryst suite phases approach those of the equivalent phases in coarse, common mantle peridotites (Eggler et al. 1979), indicating that they could represent near liquidus compositions in a magma generated in equilibrium with a lherzolite source. This experimental evidence that the early-crystallizing megacrysts are liquidus or near-liquidus phases in their respective host magmas provides extremely compelling evidence for a cognate origin.

# Chemical processes linking the Cr-rich and Cr-poor megacryst suites

Field, petrographic, mineral chemistry and experimental studies collectively point to close a genetic link between the host kimberlite and both the Cr-rich and Cr-poor megacryst suites. It is therefore necessary to identify the processes responsible for the formation of these apparently different suites from the same magma. The key is provided by the occurrence of the two megacryst populations within individual hand specimens from the Weltevreden (Meyer et al. 1979) Swartruggens (de Bruin 1998) and Kaalvallei kimberlites (this study). The samples with co-existing Cr-rich and Cr-poor megacryst phases from these three localities show a number of consistent textural and chemical patterns. Crystallization of the Cr-rich garnets and associated megacryst phases always preceded that of the Cr-poor suite. The narrow boundary zone separating the two suites is characterized by consistent zonation patterns (decreasing Ca# and Mg# and Cr/(Cr + Al)) from the Cr-rich towards the later-crystallizing Cr-poor garnet. In the Weltevreden nodule, this appears to have been accompanied by fractional crystallization, involving abundant ilmenite, reflected in the marked decrease in Cr-contents of this phase outwards across the boundary between the pink (Cr-rich) and orange (Cr-poor) garnets. Ilmenite crystallization also marks the chemical boundary zone between the red (Cr-rich) and orange (Cr-poor) garnets in the Kaalvallei nodules.

Several studies (e.g. Gurney et al. 1979; Schulze 1984, Hops et al. 1992) have demonstrated that the Cr-poor megacryst suite crystallized from a melt under isobaric conditions over a wide temperature range, and that ilmenite is a relatively late-crystallizing phase. However, the textural evidence from the polyphase Weltevreden megacryst points to crystallization of a Cr-rich ilmenite prior to that of the Cr-poor megacryst assemblage at this locality. The abundance of ilmenites within the chemical transition zone separating the pink, Cr-rich and orange, Cr-poor garnets in the Weltevreden polyphase megacryst suggests sudden onset of rapid crystallization of the former phase. This would rapidly deplete the parental liquid in Cr. As a result, the orange garnets and ilmenites that formed later, enclosed by the (Cr-poor) orthopyroxene megacryst, would be depleted in Cr. The same mechanism can account for differences in Cr contents of the red and orange garnets in the Kaalvallei specimens. It is therefore proposed that compositional differences between the two megacrysts suites can, in part at least, be explained it terms of their relative positions in a common magmatic crystallization event. It is, however, necessary to identify the processes responsible for triggering rapid ilmenite crystallization. Recent experimental evidence suggests a possible mechanism.

Dalton and Presnall (1998) demonstrate that a small degree (0-1%) of partial melting of a carbonated peridotite at 6 GPa produces liquids that range from car-

bonatitic to kimberlitic in composition with increasing melt fraction. Such melts would have a very low viscosity, allowing them to coalesce and migrate to higher levels. Because of the positive slope of the solidus at pressures above approximately 2.5 GPa (Dalton and Presnall 1998), these liquids would become superheated if they did not equilibrate with the mantle during ascent. Interruption of the ascent of a kimberlitic liquid at a higher level, leading to magma accumulation in a temporary reservoir, would result in the development of a narrow thermal aureole in the surrounding mantle rocks. Small volumes of kimberlitic liquid, injected into fractures in this thermal aureole surrounding the main kimberlite magma reservoir, would crystallize down the temperature gradient to form the megacryst suite, with the earliest formed megacrysts represented by the Crrich suite. However, negligible crystallization would take place in the main kimberlite magma body. This could account for the range in megacryst compositions despite the fact that kimberlites show little evidence for fractionation. This is closely analogous to the model originally proposed by Harte and Gurney (1981), differing only because the host kimberlite is seen as the parental liquid of the megacrysts.

If carbonate phases were present in the mantle rocks within the thermal aureole, the superheated kimberlite magma would trigger local low degrees of partial melting. Ionov et al. (1993, 1996) document petrographic evidence from carbonate-bearing peridotite xenoliths from Spitsbergen that indicates that melting of the carbonate phase occurred either shortly before, or at the time of, incorporation of the xenolith in the host magma. This is compatible with melting in a thermal aureole surrounding the host magma in the mantle. Dalton and Presnall's (1998) experiments predict that carbonatitic liquids would be produced in the cooler outer margins of the thermal aureole, where the smallest degrees of melting take place. Where temperatures are higher, slightly higher degrees of melting would produce kimberlitic melts. The range in compositions of carbonaterich inclusions in clinopyroxene megacrysts documented by van Achterbergh et al. (2002) provides evidence of the range of melts generated within the thermal aureole where the megacrysts formed.

Mixing of carbonate-rich melts with kimberlitic magma injected into a fracture network would effectively "contaminate" the latter, leading to reactions such as:

$$Si_2O_6^{4-} + 2CO_3^{2-} \rightarrow 2SiO_4^{4-} + 2CO_2$$

This would in turn lead to an increase in oxygen fugacity, and consequently in the activity of  $(Ti^{4^+} + Fe^{3^+})$ . It is suggested that this was responsible for triggering the rapid ilmenite crystallization that is indicated by the textural evidence in the Weltevreden nodule described by Meyer et al. (1979). Calcite-phlogopite patches concentrated at the boundary between the orthopyroxene and orange garnet in the Weltevreden

nodule provide possible evidence for carbonate and volatile-rich melts that mixed with the kimberlite magma within the megacryst vein network. Mixing of secondary carbonatitic melts with small volumes of kimberlitic liquids injected into a fracture network surrounding the main magma body is essentially a variation on the magma contamination models proposed by Jones (1987) and Hops et al. (1992)

The ilmenite-dominated crystallization recorded at the interface between the two garnets in the Weltevreden nodule would be expected to result in an increase in the Mg# of the residual liquid, and thus the late-crystallizing orange garnet. However, the Mg# of the latter is in fact lower (Mg#=81.0) than that of the pink garnet (Mg#=81.9) (Meyer et al. 1979). Ilmenite dominated crystallization can also not account for the marked decrease in Ca# across the chemical boundary zone separating the Cr-rich and Cr-poor garnets This indicates that there are additional factors that influenced the marked change in mineral chemistry across the narrow interface separating these garnets.

Green and Wallace (1988) and Moore (1989) proposed that the Mg-Fe distribution coefficient (as defined by Roeder and Emslie 1970) between silicate minerals and a carbonate-rich melt is significantly higher than for a silicate melt, possibly as a result of complexing between carbonate and  $Mg^{2+}$  ions. The implied higher mineral-melt  $K_d$  for carbonate-rich liquids is supported by experimental evidence presented by Dalton and Wood (1993). Complexing of  $Mg^{++}$  with a carbonaterich contaminant melt, and the resulting changes in Fe-Mg distribution coefficients offers an explanation for the hiatus in Mg# between the Cr-poor and Cr-rich megacryst suites reported by Eggler et al. 1979). It would also explain the marked decrease in Mg# from the pink to the orange garnet across a narrow boundary zone in the in the Weltevreden orthopyroxene nodule described by Meyer et al. (1979), and the analogous compositional jump across a narrow interface in the Kaalvallei garnet nodules (Fig. 5).

The experimental evidence presented by Dalton and Wood (1993) also indicates that carbonatitic liquids reacting with silicate phases will become more calcitic. This provides an explanation for the marked decrease in Ca# across the interface from the pink to the orange garnet in both the Weltevreden orthopyroxene megacryst and the Kaalvallei samples KLV-1&2. In other words, the marked decrease in Mg# and Ca# across the chemical interface separating the Cr-rich and Cr-poor garnets is dominated by changes in distribution coefficients caused by mixing (contamination) with a carbonate-rich liquid, that are superimposed on fractional crystallization effects.

In summary, it is suggested that Cr-rich and Cr-poor megacrysts are linked to a common crystallization event, resulting from the injection of kimberlitic liquid into a fracture network surrounding a kimberlite magma body at depth in the mantle. The range in megacryst compositions reflects crystallization across the thermal aureole surrounding the kimberlite magma body. This closely follows the model proposed by Harte and Gurney (1981), except that the host kimberlite is seen as the parental magma for the megacryst suite. The least evolved members of the Cr-rich megacryst suite crystallized from a relatively primitive kimberlite liquid. If phases such as carbonate and phlogopite, with low melting temperatures, occur within the thermal aureole, these would form secondary liquids that would mix with (i.e. contaminate) the small volumes of kimberlitic magma injected into the fracture network. This, in turn, would trigger rapid crystallization of ilmenite. The experimental evidence suggests that differences in Mg# and Ca# between the Cr-rich and Cr-poor megacryst suites are dominated by the effects of changing distribution coefficients related to carbonate contamination of the parental liquid. However, fractional crystallization involving ilmenite and other Cr-rich phases would result in the rapid depletion of Cr in residual liquids and thus, later-crystallizing megacryst phases. In contrast to the range in fractional crystallization in the liquids filling the vein network within the thermal aureole surrounding the main volume of kimberlitic magma, the latter would undergo minimal fractional crystallization.

# **Isotopic and REE considerations**

The field, petrographic and experimental evidence presented points to a cognate origin for the Cr-rich and Crpoor megacryst suites. However, it is necessary to reconcile this conclusion with the evidence for isotopic disequilibrium between megacrysts and the host magma, as well as between different megacrysts from the same pipe (Jones 1987; Hops et al. 1992; Davies et al. 2001). These differences are readily explained if there are isotopic differences between the mantle source involved in kimberlite generation, and the mantle at higher levels, where the megacrysts crystallize from kimberlitic liquids contaminated by secondary, probably Sr-rich, carbonatitic melts. Mixing of variable proportions of such secondary melts with the kimberlitic magmas in the vein network would account for the Sr isotopic differences between different clinopyroxene megacrysts from the same kimberlite reported by Hops et al. (1992) and Davies et al. (2001).

A number of studies have used REE modelling to argue that the Cr-poor megacrysts could not have crystallized from the host kimberlite, but rather that they are derived from one of a variety of alkali-rich basaltic magmas (e.g. Harte 1983; Jones 1987; Davies et al. 2001). However, all of these modelling studies have a variety of inherent problems. Firstly, kimberlites are characterised by a wide range in REE concentrations. Thus, group 1A and group II kimberlites have average La contents 368 and 818 times chondritic respectively, and average La/Nd ratios of 1.0 and 1.38 (Smith et al. 1985). There is also a wide range in REE concentrations within individual pipes. Thus, four samples from Jag-

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ersfontein showed a range in La from 94 to 1,145 times chondritic, and La/Nd ratios ranging from 0.88 to 1.39 (Smith et al 1985). The Wesselton kimberlite has a range in La varying between 368 and 854 times chondritic (Mitchell and Brunfelt 1973; Mitchell 1986). Le Roex et al. (2003) report a comparable range for the Kimberley pipes as a group. This raises a major question mark over the appropriate kimberlite composition to use in modelling studies.

Of greater concern is that all the modelling studies are based on REE partition coefficients for basaltic systems. Kramers et al. (1981) suggested that for kimberlites, clinopyroxene-liquid partition coefficients could be up to an order of magnitude lower than for basaltic systems. While Davies et al. (2001) note that no experimental data have been presented to support this conclusion, a number of experimental studies must at least raise strong concerns regarding the use of basaltic REE partition coefficients for trace element modelling in (carbonatebearing) kimberlitic systems.

Hamilton et al. (1989) showed that over the pressure range of 1-6 Kb, and temperatures between 1,050 °C and 1,250 °C, partitioning of REE between carbonate liquids and phonolitic and nephelinitic magmas is strongly dependent on temperature, pressure and the composition of the silicate liquid. These authors demonstrated that increasing pressure, decreasing temperature and increased polymerisation of the silicate liquid led to a concentration of REE into the carbonate liquid, by as much as a factor of 10. Similarly, Jones et al. (1995) quote Fielding (1992) as reporting that trace element distribution between silicate and carbonate liquids is a complex function of temperature, pressure and composition in the range 2–8 kbar and 1,100–1,400°C. However, Jones et al. (1995) note that these effects were not observed in their experimental runs, which were carried out at higher pressures (7-20 kbar), although they did not include REE. Baker et al. (1995) demonstrate that there are marked changes in the clinopyroxene-liquid partition coefficient for Ti with increasing partial melting just above the solidus. They speculate that other high field strength ions, including the REE, may show similar effects. Blundy and Dalton (2000) show that in the systems diopside-albite, and diopsidealbite-dolomite, the clinopyroxene-liquid partition coefficient for the HREE is up to fivefold higher for carbonate-rich liquids compared to those for silicate liquids. They point out that such differences offer an explanation for the extreme LREE enrichment of carbonatites-and by extension, kimberlites.

The limited published values for partition coefficients between clinopyroxene and a carbonatitic melt are summarized in Table 3. For comparison, the table includes Cpx/melt partition coefficients determined by Hart and Dunn (1993), using a natural alkali olivine basalt composition. REE partition coefficients determined for silicate and carbonate-bearing systems show little similarity, and there are also significant differences in the ratios of the partition coefficients for the heavier and lighter REE elements reported in different studies. These observations raise very serious questions regarding the use of partition coefficients for basaltic systems to model megacryst equilibrium liquids if the latter are carbonate-bearing. This conclusion is underlined by the fact that trace element models using basaltic REE partition coefficients for garnets invariably give poorer fits than those for clinopyroxene (Jones 1987; Burgess and Harte 2004).

There are also geological problems associated with a number of models that use basaltic REE partition coefficients to invoke an alkali basalt parental magma for the megacryst suite. Thus Burgess and Harte (2004) interpret zoning in garnets in sheared peridotites from the Jagersfontein kimberlite to reflect metasomatism by a MORB-source melt that crystallized the Cr-poor megacrysts. They demonstrate that the highest temperature sheared peridotites from Jagersfontein (inferred to have been derived from the greatest depth) have the most pronounced metasomatic imprint. They interpret the progressively lower degrees of metasomatism in lower temperature rocks as reflecting the evolution of the megacryst magma with decreasing pressure and temperature over a mantle section of some 50–75 km. However, this interpretation is inconsistent with the evidence from a wide variety of localities, including Jagersfontein, that the megacryst magma evolved over a wide (300°C) temperature range under essentially isobaric conditions (Gurney et al. 1979; Schulze 1984; Hops et al. 1992).

An explanation for this apparent geological conundrum is provided by Fraser and Lawless (1978) and Lee (1997), who suggested that the sheared peridotites, like the megacrysts, formed over a very limited depth range. These authors proposed that the apparent pressure range indicated by many sheared peridotite xenoliths is an artefact of slower diffusion (by several orders of magnitude) of the trivalent Al, used for the geobarometers, relative to those for the divalent ions, used in the geothermometers. This interpretation would account for the fact that while some geothermometers indicate a "kinked" geotherm at depths of around 150–180 km within the lithosphere in many different cratonic terrains around the world, seismic studies of the Kaapvaal craton have failed to detect any discontinuity in the mantle at this depth (Fouch et al. 2003). If the "kinked" geotherm is indeed an artefact of the differing diffusion rates of divalent and trivalent ions, it follows that it does not reflect a horizontal thermal, chemical and mechanical boundary in the mantle. Diffusion problems would not be applicable to the megacrysts, as they crystallized from a magma, and with very rare exceptions, do not show evidence of subsolidus re-equilibration. The megacrysts would therefore provide more reliable pressure estimates over a range of temperatures.

Tainton and McKenzie (1994) suggested that the megacrysts could have crystallized from alkali-rich basaltic liquids like those inferred to be responsible for the modal mantle metasomatism documented by Erlank

	I. For basaltic	and carbonatitic m	helts				2. For synthetic melts	
	Cpx-Alk. Bs	Clinopyroxene-ca	urbonate melt partitio	n coefficients			Cpx-silicate	Cpx-carbonate
	Hart and Dunn (1993)	Klemme et al. (1995) (Range)	Klemme et al. (1995) (Average)	Green et al. (1992)	Sweeney et al. (1995)	Jones et al. (1995)	System (Di-Ab) (Blundy and Dalton 2000)	System Di-Ab-Dmt (Blundy and Dalton 2000)
La Ce	0.0536 0.0858	0.06-0.11 0.08-0.09	0.07 (0.02) 0.09 (0.007)	0.02		0.023	0.053	0.053
Pr Nd	0.1873	0.11-0.12 0.097-0.19	$0.11 (0.04) \\ 0.11 (0.052)$				0.136	0.26
Sm Fu	0.291 ND	0.13 0.16-0.28	0.13 (0.04)	0.08			0.174	0.35
Dy Gd	0.442 0.442	0.17-0.35 0.26-0.32	0.29 (0.03)					
Er Yb	0.38/	0.33-0.48	0.41 (0.06)				0.194	0.963
D(Sm)/D(La) D(Dy)/D(La)	5.43 8.25 7.22	2.17–1.18 4.33–2.9 5.82 4.35	1.85 4.14 5.05	4			3.28	6.6
D(Yb)/D(La)	77.1	00.400.0	0.0				3.66	18.17
Y Ba	0.467	0.22-0.37	0.30 (0.08)	0.22	0.28	0.00038		
Sr	0.1283	0.08-0.11	0.08 (0.015)	0.03	0.28	00000		
Ti	0.384	0.81 - 2.02	1.42(0.61)		0.17			
Nb	0.0077	0.07 - 0.13	0.10(0.03)	0.01	0.24	0.0046		
1a Zr	0.1234	0.12-0.10	(c000) c1.0 0.48 (0.37)	0.29	0.32			
Hf	0.256	0.16	0.16(0.05)					
٨	0.31	1.68 - 3.46	2.93(0.89)					

et al. (1987). However, isotopic studies (Hawkesworth et al. 1983, 1990) date the mantle metasomatism beneath southern Africa at  $\sim$ 150 Ma—i.e. shortly following the Karoo volcanic episode, and roughly coeval with the opening of the Indian Ocean. Given the evidence that the megacrysts were formed shortly before kimberlite emplacement (80–90 Ma for many Group I kimberlites), they cannot be linked to the Jurassic metasomatic event.

#### **Ilmenite chemistry**

The marked range in Cr-contents of the Mg-rich limb of the KN-70 ilmenite population (Fig. 6a) is interpreted to reflect early fractional crystallization of this phase. This would cause rapid Cr-depletion of the residual liquid in the pegmatitic (megacryst) vein network surrounding the main kimberlite magma body. The range in MgO in the Cr-poor limb of the KN-70 ilmenite field is interpreted to reflect continued crystallization of the phase together with Mg-silicates. The KN70 ilmenite population is therefore interpreted to reflect a chemical continuum from the Cr-rich to Cr-poor megacryst suites at this locality.

The relatively high Cr-contents of Orapa ilmenites (Fig. 6c) require that this phase crystallized prior to significant proportions of other Cr-rich phases such as garnet or clinopyroxene. As the ilmenite chemical field does not extend to very low Cr-contents (<1% Cr<sub>2</sub>O<sub>3</sub>) relatively low proportions of ilmenites and other Cr-rich phases such as garnet and clinopyroxene must have formed prior to cessation of crystallization of the oxide. The Orapa ilmenites are MgO-poor relative to those from the KN-70 kimberlite with comparable Cr-contents. The reasons for this difference are uncertain, but may reflect earlier crystallization of Mg-rich, Cr-poor megacryst phases such as olivine and orthopyroxene at Orapa. Factors influencing the order and proportions in which different megacryst phases crystallize in megacryst liquids from different kimberlites are also not well understood. However, they may include differences in the composition of the parental magma, temperatures and pressures of crystallization, or in the proportions of low-melting components contaminating liquids in the vein network where the megacryst suite crystallized.

The model linking the Cr-poor and Cr-rich megacryst provides a potential mechanism to explain paired highand low-Cr ilmenite populations, such as those that characterize many of the Molopo-Tsabong kimberlites (Fig. 6b) (Moore 1987) as well as the Monastery kimberlite (Griffin et al. 1997). It is suggested that such paired populations may be linked to an inhomogeneous distribution of carbonate within the mantle. As a result, carbonate-rich melts formed within the thermal aureole surrounding the kimberlite magma would migrate into and contaminate some, but not all, of the small volumes of kimberlitic liquid injected into the associated fracture network. This would lead to differences in the crystallization sequences of megacryst phases in different veins.

Cr-rich ilmenites would form where the phase preceded the crystallization of significant proportions of clinopyroxene and garnet. The Cr-poor ilmenite population would form where the phase crystallized after the Crrich phases following contamination by a carbonate liquid. In terms of this model, it would be anticipated that a carbonate melt would contaminate kimberlitic liquids within the vein network to varying degrees, resulting in ilmenite compositions intermediate between the Cr-rich and Cr-poor suites. This is indeed the case in the Molopo-Tsabong kimberlites (Fig. 6b) as well as at Monastery (Griffin et al. 1997). In the latter pipe, some of the Cr-poor ilmenites enclose ovoid carbonate-sulphide inclusion, consistent with crystallization of from a liquid contaminated by carbonate melts. The low proportion of Cr-rich ilmenites at Monastery suggests that, at this locality, only a very small proportion of the vein network did not experience such contamination.

## Implications for the genesis of kimberlite magmas

Previous models have invoked separate magmas to account for the Cr-rich and Cr-poor megacrysts (e.g. Eggler et al. 1979; Hatton 1998). The Cr-poor suite has been linked to proto-kimberlite liquids (e.g. Griffin et al. 1989) as well a variety of other alkaline magmas that are inferred to have been present at the time or shortly prior to kimberlite eruption (e.g. Harte 1983; Jones 1987; Moore et al. 1992; Hops et al. 1989; Davies et al. 2001). These models all imply that the generation of kimberlite magmas is a complex, multi-stage process. A far simpler (single stage) model of formation of kimberlites is possible if both megacryst suites are crystallization products of the host kimberlite, as argued in this study. In particular, it is unnecessary to invoke a link to magmas that are not recorded at the surface, and are not present as xenoliths in kimberlites, nor as inclusions in megacryst phases. It is noted in this respect that Eggler and Wendlandt (1979), Dalton and Presnall (1998) and Wyllie and Lee (1999) have demonstrated that primary kimberlitic liquids can form in equilibrium with garnet lherzolite. This conclusion is supported by bulk rock REE modeling studies (Tainton and McKenzie 1994; Le Roex et al. 2003) that demonstrate that kimberlites can be derived as primary melts from modally metasomatised garnet peridotites or garnet phlogopite peridotites such as those described by Erlank et al. (1987).

Experimental evidence (Dalton and Wood 1993) indicates that the mineral-melt Mg–Fe distribution coefficient ( $K_d$ ) is higher for carbonate-bearing liquids than silicate magmas. A corollary is that a carbonate-bearing magma generated in equilibrium with mantle phases will be more Mg-rich than expected for carbonate-free systems. Small degrees of partial melting involving a mantle carbonate would therefore produce ultrabasic liquids that are highly enriched in incompatible elements (i.e. kimberlite magmas). A quantitative evaluation of the influence of carbonate content of liquids on crystal-melt distribution coefficients would provide important constraints on the origin of kimberlite and other alkaline magmas.

LeChiminant et al. (1998) and Konzett et al. (1998) document complex zonation patterns and overgrowths in kimberlitic megacryst zircons. Both studies show that these zircons record a wide range of ages, ranging from that of kimberlite emplacement to 30-40 Ma earlier. Data presented by Davis (1977) and Zartman et al. (1997) suggest that in South African pipes, zircon ages are restricted to a range from the time of emplacement to only 5-10 years earlier. The zonation patterns, overgrowths and range in ages of these zircon from individual pipes appear to reflect mantle events that predate entrainment of the phase by the host kimberlite. This suggests that the kimberlite magma may have been interrupted at the depth of megacryst formation for a significant period of time prior to eruption. It is possible that during this halt, periodic re-fracturing of the vein network surrounding the magma chamber may have resulted in the injection of later pulses of kimberlitic liquid from the main magma body. This could account for the complex zonation patterns recorded, and the range in zircon ages. The study of zircon ages from individual pipes therefore offers a potentially important window on both the nature and timing of mantle processes that predate kimberlite eruption.

# Implications for mantle composition and structure and kimberlite diamond potential

Zoning of garnets in the sheared peridotites, and the range in compositions of this suite of mantle xenoliths has been ascribed to interaction with (metasomatism by) the megacryst magma (Gurney and Harte 1980; Smith and Boyd 1987 and 1989; Griffin et al. 1989; Burgess and Harte 1999 and 2004). The deformation textures that characterize many megacrysts of both the Cr-rich and Cr-poor suites are consistent with their formation in close proximity to the sheared peridotites. Nevertheless, field, petrological and experimental evidence individually and collectively argue that the megacryst suites are ultimately derived from the host kimberlite. This in turn requires that the kimberlite was responsible for the metasomatism of the sheared peridotites. This is consistent with the model put forward by Mercier (1979) for the origin of this suite -i.e. that the sheared peridotites formed in a narrow envelope ( $\sim 50$  m wide) surrounding the kimberlite conduit prior to eruption. The identification of megacrysts in polymict peridotites, interpreted to have formed in the kimberlite conduit (Lawless et al. 1979; Wyatt and Lawless 1984), is also consistent with the Mercier model.

The conclusion that sheared peridotites are the products of metasomatism and thermal re-equilibration within the thermal aureole surrounding the kimberlite magma in the upper mantle implies that they provide no information whatsoever on vertical variations in the mechanical and thermal structure of the mantle and, in particular, cannot be used to infer compositions of the asthenosphere.

The link between kimberlites, megacrysts and sheared peridotites has important implications for evaluating the prospectivity of an area for the discovery of economic kimberlites. Griffin and Ryan (1995) have proposed that the inferred depth of equilibration of the least metasomatised sheared peridotites reflects the maximum depth of diamond stability within the lithosphere. The depth interval between the intersection of the geotherm with the graphite-diamond inversion curve, and this inferred maximum depth of diamond stability within the lithosphere is defined as the "diamond window." In general, regions characterized by a broader diamond window would be considered to be more prospective than those with a narrow window. However, if the sheared peridotites are the product of matasomatism by the host kimberlite in a narrow thermal aureole surrounding a magma reservoir in the mantle, rather than a horizontal mechanical, thermal and chemical boundary, such metasomatism may have occurred at different depths in different kimberlites within the same cluster, and probably at greater depths for diamond-rich pipes relative to associated diamond-poor pipes. The diamond window, as determined from a single or even several non-economic kimberlites within a cluster, would then have little relevance for the prospectivity of the area as a whole.

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#### References

- Baker MB, Hirshmann MM, Ghiorso MS, Stolper EM (1995) Compositions of near-solidus peridotite melts from experiments and thermodynamic calculations. Nature 375:308–311
- Blundy J, Dalton J (2000) Experimental comparison of trace element partitioning between clinopyroxene and melt in carbonate and silicate systems, and implications for mantle metasomatism. Contrib Mineral Petrol 139:356–371
- Boyd FR (1984) Siberian geotherm based on lherzolites xenoliths from the Udachnaya kimberlite, USSR. Geology 12:528–530
- Boyd FR, Dawson JB, Smith JV (1984) Granny Smith diopside megacrysts from the kimberlites of the Kimberley area and Jagersfontein, South Africa. Geochim Cosmochim Acta 48:381–384
- Burgess SR, Harte B (1999) Tracing lithosphere evolution through the analysis of heterogeneous G9/G10 garnets in Peridotite xenoliths, I: major element chemistry. In: Gurney JJ, Gurney JL, Pascoe MD, Richardson SH (eds) Proceedings of VIIth international conference, vol 1 (Red Roof Design). pp 66–80
- Burgess SR, Harte B (2004) Tracing lithosphere evolution through the analysis of heterogeneous G9/G10 garnets in Peridotite xenoliths, II: REE chemistry. J Petrol 45:609–634

- Carlson RW, Irving AJ, Hearn BC (1999) Chemical and isotopic systematics of peridotite xenoliths from the Williams kimberlite, Montana: clues to processes of lithosphere formation, modification and destruction. In: Gurney JJ, Gurney JL, Pascoe MD, Richardson SH (eds) Proceedings VIIth international conference, vol 1 (Red Roof Design) pp. 90–98
- Dalton JA, Presnall DC (1998) The continuum of primary carbonatitic-kimberlitic melt compositions in equilibrium with lherzolite: Data from the system CaO-MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>-CO<sub>2</sub> at 6 Gpa. J Petrol 39:1953–1964
- Dalton JA, Wood BJ (1993) The composition of primary carbonate melts and their evolution through wallrock reaction in the mantle. Earth Planet Sci Lett 119:511–525
- Davies GR, Spriggs AJ, Nixon PH (2001) A non-cognate origin for the Gibeon kimberlite megacryst suite, Namibia: Implications for the origin of Namibian kimberlites. J Petrol 42:159–172
- Davis GL (1977) The ages and uranium contents of zircons from kimberlites and associated rocks. Extended bstracts, 2nd International kimberlite conference, Sante Fe (New Mexico)
- Dawson JB, Hervig RL, Smith JV (1981) Fertile iron-rich dunite xenoliths from the Bultfontein kimberlite, South Africa. Fortschr Mineral 59:303–324
- De Bruin D (1998) Inclusion bearing Cr-poor and Cr-rich garnet megacrysts from the Group II Swartruggens kimberlite. Extended Abstracts, 7<sup>th</sup> International Kimberlite Conference, Cape Town, South Africa, pp 181–183
- De Beers Consolidated Mines Limited and De Beers Centenary AG (2000) Annual Report pp. 156
- Eggler DH, Wendlandt RF (1979) Experimental studies on the relationship between kimberlitic magmas and partial melting of peridotite. In: Boyd FR, Meyer HOA (eds) Proceedings 2nd International Kimberlite Conference, vol 1 (AGU). pp 330–338
- Eggler DH, McCullum ME, Smith CB (1979) Megacryst assemblages in kimberlites from northern Colorado and southern Wyoming: petrology, geothermometry-barometry and areal distribution. In: Boyd FR. Meyer HOA (eds) Proceedings 2nd international kimberlite conference, Vol 2 (AGU). pp 213–226
- Ehrenberg SH (1979) Garnetiferous ultramafic inclusions in minette from the Navajo volcanic field. In: Boyd FR, Meyer HOA (eds) Proceedings 2nd International kimberlite conference, vol 2 (AGU). pp 330–344
- Erlank AJ, Waters FG, Hawkesworth CJ, Haggerty SE, Allsopp HL, Rickard RS, Menzies M (1987) Evidence for mantle metasomatism in peridotite nodules from the Kimberley pipes, South Africa. In: Menzies M, Hawkesworth CJ (eds) Mantle metasomatism. Academic, New York, pp. 221–311
- Fielding KD (1992) Element partitioning between coexisting carbonate and silicate liquids. Unpublished PhD thesis, University Edinburgh
- Fouch MJ, James DE, Vandecar JC, Van Der Lee S, the Kaapvaal Seismic Group (2003) Mantle seismic structure beneath the Kaapvaal and Zimbabwe cratons (In press). South Afr J Geol
- Fraser DG, Lawless PJ (1978) Palaeogeotherms: implications of disequilibrium in garnet lherzolite xenoliths. Nature 273:220– 222
- Green DH, Wallace ME (1988) Mantle metasomatism by ephemeral carbonatitic melts. Nature 336:459–462
- Green YH, Adam J, Sie SH (1992) Trace element partitioning between silicate minerals at 25 kbar and application to mantle metasomatism. Contrib Mineral Petrol 46:179–184
- Griffin WL, Ryan CG (1995) Trace elements as indicator minerals: area selection and target evaluation indiamond exploration. Hour Geochem Explor 53:311–338
- Griffin WL, Smith D, Boyd FR, Cousens DR, Ryan CG, Sie SH, Suter GF (1989) Trace element zoning in garnets from sheared mantle xenoliths. Geochim Cosmochim Acta 53:561–567
- Griffin WL, Moore RO, Ryan CG, Gurney JJ, Win TT (1997) Geochemistry of magnesian ilmenite megacrysts from southern African kimberlites. Russ Geol Geophys 38:421–443
- Gurney JJ, Harte B (1980) Chemical variations in upper mantle nodules from southern African kimberlites. Phil OS Trans R Soc Lond A 297:273–293

- Gurney JJ, Zweistra P (1995) The interpretation of the major element compositions of mantle minerals in diamond exploration. J Geochem Explor 53:293–310
- Gurney JJ, Jacob WRO, Dawson JB (1979) Megacrysts from the Monastery kimberlite pipe, South Africa. In: Boyd FR, Meyer HOA (eds) Proceedings 2nd internaional kimberlite conference Vol 2 (AGU). pp. 227–243
- Haggerty SE, Hardie RB III, McMahon BM (1979) The mineral chemistry of ilmenite nodule associations from the Monastery diatreme. In: Boyd FR, Meyer HOA (eds) Proceedings of 2nd international kimberlite conference vol 2 (AGU). pp. 249–256f
- Hamilton DL, Bedson P, Esson J (1989) The behaviour of trace elements in the evolution of carbonatites. In: Bell K (ed) Carbonatites – Genesis and evolution. Unwin Hyman, London, pp 405–427
- Hart SR, Dunn T (1993) Experimental cpx/melt partitioning of 24 trace elements. Contrib Mineral Petrol 113:1–8
- Harte B (1977) Rock nomenclature with particular relation to deformation and recrystallisation textures in olivine-bearing xenoliths. J Geol 85:279–288
- Harte B (1983) Mantle peridotite and processes the kimberlite sample. In: Hawkesworth CJ, Norry MJ (eds) Continental basalts and mantle xenoliths. Shiva Publishing, England, pp. 46–91
- Harte B, Gurney JJ (1981) The mode of formation of the Cr-poor megacryst suite from kimberlites. J Geol 89:749–753
- Hatton CJ (1998) The kimberlite-megacryst link at Monastery mine. Extended abstracts 7th international kimberlite conference, Cape Town pp. 314–316
- Hawkesworth CJ, Erlank AJ, Marsh JS, Menzies MA, van Carlsteren P (1983) Evolution of the continental lithosphere: evidence from volcanics and xenoliths in southern Africa. In: Hawkesworth CJ, Norry MJ (eds) Continental basalts and mantle xenoliths. Shiva Publishing, England, pp. 111–138
- Hawkesworth CJ, Erlank AJ, Kempton PD, Waters FG (1990) Mantle metasomatism: isotopic and trace element trends in xenoliths from Kimberley, South Africa. Chem Geol 85:19–34 Hops JJ, Gurney JJ, Harte B, Winterburn P (1989) Megacrysts and
- Hops JJ, Gurney JJ, Harte B, Winterburn P (1989) Megacrysts and high temperature nodules from the Jagersfontein kimberlite pipe. In: Ross J, (ed) Proceedings 4<sup>th</sup> international kimberlite conference Geological Society Australia Special Publications Blackwell, VIC vol 14.759–770
- Hops JJ, Gurney JJ, Harte B (1992) The Jagersfontein Cr-poor megacryst suite – towards a model for megacryst paragenesis. J Volcanol Geothermal Res 50:143–160
- Ionov A, Dupuy C, O'Reilly SY, Kopylova MG, Genshaft YS (1993) Carbonated peridotite xenoliths from Spitsbergen: implications for trace element signature of mantle carbonate metasomatism. Earth Planet Sci Lett 119:283–297
- Ionov DA, O'Reilly SY, Genshaft YS, Kopylova MG (1996) Carbonate-bearing mantle peridotite xenoliths from Spitsbergen: phase relationships, mineral compositions and trace-element residence. Contrib Mineral Petrol 125:375–392
- Jakob WRO (1977) Geochemical aspects of the megacryst suite from the Monastery kimberlite pipe. MSc thesis, (unpublished), University Cape Town, pp. 81
- Jones RA (1987) Strontium and Neodymium isotope and rare earth element evidence for the genesis of megacrysts in kimberlites of southern Africa. In: Nixon PH (ed) Mantle Xenoliths. Wiley, NewYork, pp 711–724
- Jones JH, Walker D, Pickett DA, Murrell MT, Beattie P (1995) Experimental investigations of the partitioning of Nb, Mo, Ba, Ce, Pb, Ra, Th, Pa and U between immiscible carbonate and silicate liquids. Geochim Cosmochim Acta 59:1307–1320
- Kennedy LA, Russell, JK, Kopylova MG (2002) Mantle shear zones revisited: The connection between the cratons and mantle dynamics. Geology 30:419–422
- Kharkiv AD (1995) Two series of megacrysts from kimberlites of the Verkhnemunskoye field (Republic of Sakha). Extended abstracts, 6th international kimberlite conference, Novosibirsk, Russia pp 271–272
- Klemme S, van der Laan SR, Foley SF, Günther D (1995) Earth Planet Sci Lett 133:439-448

- Konzett J, Armstrong RA, Sweeney RJ, Compston W (1998) The timing of Marid metasomatism in the Kaapvaal mantle, An ion probe study of MARID xenoliths. Earth Planet Sci Lett 160:133–145
- Kopylova MG, Russell JK, Cookenboo H (1999) Petrology of peridotite and pyroxenite xenoliths from the Jerico kimberlite: implications for the thermal state of the mantle beneath the Slave Craton, northern Canada. J Petrol 40:79–104
- Kostrovisky SI, Mitchell RG (1995) The trends of variability of garnet megacryst composition from diamond-bearing and diamond-devoid pipes (Yakutia, Russia). Extended abatracts, 6th international kimberlite conference, Novosibirsk, Russia pp 301–302
- Kramers JD, Smith CB, Lock NP, Harmon RS, Boyd FR (1981) Can kimberlite be generated from ordinary mantle?. Nature 291:53–56
- Lawless PJ, Gurney JJ, Dawson JB (1979) Polymict peridotites from the Bultfontein and de Beers mines, Kimberley, South Africa. In: Boyd FR, Meyer HOA (eds) Proceedings 2nd international kimberlite conference, Vol 2 (AGU). pp 149–155
- Le Roex AP, Bell DR Davis P (2003) Petrogenesis of group I kimberlites from Kimberley, South Africa: evidence from bulk-rock chemistry. J Petrol 44:2261–2286
- Le Chiminant AN, Heaman LM, Kretchmar U, LeCouteur PC (1998) Complex origin and multiple ages of mantle zircon megacrysts from Canadian and South African kimberlites. Extended abstracts, 7th international kimberlite conference, Cape Town, pp. 486–488
- Lee JE (1993) Indicator mineral techniques in a diamond exploration programme at Kokong, Botswana. Prospectors and developers association of Canada, diamonds: exploration, sampling and evaluation Toronto, Canada, pp. 213–236
- Lee CT (1997) Are inflected geotherms real? Extended Abstracts, VIIth International Conference, Cape Town, South Africa 489– 491
- McCallister RH, Meyer HOA, Aragan R (1979) Partial thermal history of two exsolved clinopyroxenes from the Thaba Putsoa kimberlite pipe, Lesotho. In: Boyd, FR, Meyer HOA (eds) Proceedings of the 2nd international kimberlite conference, vol 2 (AGU). pp 244–248
- Mercier J-CC (1979) Peridotite xenoliths and the dynamics of kimberlite intrusion. In: Boyd FR, Meyer HOA (eds) Proceedings 2nd International kimberlite conference vol 2 (AGU). pp 197–212
- Meyer HOA, Tsai H, Gurney JJ (1979) A unique enstatite megacryst with co-existing Cr-poor and Cr-rich garnet, Weltevreden floors, South Africa. In: Boyd FR, Meyer HOA (eds) Proceedings 2nd International kimberlite conference vol 2 (AGU). pp 279–291
- Mitchell RH (1973) Magnesium ilmenite and its role in kimberlite petrogenesis. J Geol 81:301–311
- Mitchell RH (1986) Kimberlites: Mineralogy, geochemistry and petrology. Plenum, New York
- Mitchell RH (1987) Megacrysts in kimberlites from the Gibeon field, Namibia. Neues Jahrbuch Mineral Abh 157:267–283
- Mitchell RH, Brunfelt AO (1973) Rare Earth geochemistry of kimberlite. Phys Chem Earth 9:671–686
- Moore AE (1987) A model for the origin of ilmenite in kimberlite and diamond: implications for the genesis of the discrete nodule (megacryst) suite. Contrib Mineral Petrol 95:245–253
- Moore AE (1989) Olivine: a monitor of magma evolutionary paths in kimberlites and olivine melilitites. Contrib Mineral Petrol 99:238–248
- Moore AE, Lock NP (2001) The origin of mantle-derived megacrysts and sheared peridotites – evidence from kimberlites in the northern Lesotho – Orange Free State (South Africa) and Botswana pipe clusters. S Afr J Geol 104:23–38
- Moore RO, Griffin WL, Gurney JJ, Ryan CG, Cousens DR, Sie SH, Suter GF (1992) Trace element geochemistry of ilmenite megacrysts from the Monastery kimberlite, South Africa. Lithos 29:1–18

- Nixon PH, Boyd FR (1973a) The discrete nodule (megacryst) association in kimberlites from northern Lesotho. In: Nixon PH (ed) Lesotho Kimberlites. Cape and Transvaal Printers, Cape Town, pp. 67–75
- Nixon PH, Boyd FR (1973b) Petrogenesis of the granular and sheared ultrabasic nodule suite in kimberlite. In: Nixon PH (ed) Lesotho Kimberlites. Cape and Transvaal Printers, Cape Town, pp. 48–56
- Nixon PH, Boyd FR (1973c) Deep-seated nodules. In: Nixon PH (ed) Lesotho Kimberlites. Cape and Transvaal Printers, Cape Town, pp. 106–109
- Pasteris JD, Boyd FR, Nixon PH (1979) The ilmenite association at the Frank Smith mine, RSA. In: Boyd FR, Meyer HOA (eds) Proceedings 2nd international kimberlite conference, vol 2 (AGU). pp. 265–278
- Pearson NJ, Griffin WL, Doyle BJ, O'Reilly SY, Van Achterbergh E, Kivi K (1999) Xenoliths from kimberlite pipes of the Lac de Gras area, Slave Craton, Canada. In: Gurney JJ, Gurney JL, Pascoe MD, Richardson SH (eds) Proceedings VIIth international conference, vol 1 (Red Roof Design). pp. 644–658
- Pouchou JL, Pichoir F (1984) A new method for quantitative X-ray microanalysis. Part 1: application to the analysis of homogeneous rock samples. Reserche Aerospatiale 5:13–28
- Richardson SH (1986) Latter-day origin of diamonds and eclogitic paragenesis. Nature 322:623–626
- Richardson SH, Gurney JJ, Erlank AJ, Harris JW (1984) Origin of diamond in old enriched mantle. Nature 310:198–202
- Roeder PL, Emslie RF (1970) Olivine-liquid equilibrium. Contrib Mineral Petrol 29:275–289
- Schulze DJ (1984) Cr-poor megacrysts in the Hamilton Branch kimberlite, Kentucky. In: Kornprobst J (ed) Proceedings 3rd international kimberlite conference, vol 2, Elsevier, Amsterdam, pp. 97–108
- Schulze DJ (1987) Megacrysts from alkalic volcanic rocks. In: Nixon PH (ed) Mantle Xenoliths, Wiley, New York, pp. 433–451
- Schulze DJ, Valley JR, Bell DR, Spicuzza MJ (2001) Oxygen isotopes variations in Cr-poor megacrysts from kimberlite. Geochim et Cosmochim Acta 65:4375–4384
- Shee SR, Gurney JJ (1979) The mineralogy of xenoliths from Orapa, Botswana. In: Boyd FR, Meyer HOA (eds) Proceedings 2nd International kimberlite conference, vol 2 (AGU). pp. 37– 49
- Smith CB (1983) Pb, Sr and Nd isotopic evidence for sources of southern African kimberlites. Nature 304:51–54
- Smith D, Boyd FR (1987) Compositional heterogeneities in a high temperature lherzolite nodule and implications for mantle processes. In: Nixon PH (ed) Mantle xenoliths. Wiley, New York, pp. 551–561
- Smith D, Boyd FR (1989) Compositional heterogeneities in minerals of sheared lherzolite inclusions from African kimberlites. In: Ross (ed) Proceedings 4th International kimberlite conference Geological Society Australian Special Publications 14:709–724
- Smith CB, Gurney JJ, Skinner EMW, Clement CR, Ebrahim N (1985) Geochemical character of southern African kimberlites: an approach based on isotopic constraints. Trans Geol Soc S Afr 88:267–280
- Smith CB, Schulze DJ, Bell D, Viljoen KS (1995) Bearing of the subcalcic, Cr-poor megacryst suite on kimberlite petrogenesis and lithospheric structure. Extended abstracts, 6th international kimberlite conference, Novosirbirsk, Russia, pp 546–548
- Sweeney RJ, Prozesky V, Przybylowicz W (1995) Selected trace and minor element partitioning between peridotitic minerals and carbonate melts at 18–46 kb pressure. Geochim Cosmochim Acta 59:3671–3683
- Tainton KM, McKenzie D (1994) The generation of kimberlites, lamproites and their source rocks. J Petrol 35: 787–817
- Tollo RP (1982) Petrology and mineral chemistry of ultramafic and related inclusions from the Orapa A/K-1 kimberlite pipe, Botswana. MSc Thesis (unpublished), University of Massachusetts, pp. 203

- Van Achterbergh E, Griffin WL, Ryan CG, O'Reilly SY, Pearson NJ, Kivi K, Doyle BJ (2002) A subduction signature for quenched carbonatites from the deep lithosphere. Geology 30:743–746
- Wagner PA (1914) The diamond fields of southern Africa. The Transvaal Leader, Johannesburg, p. 355
- Wyatt BA, Lawless PJ (1984) Ilmenite in polymict xenoliths from the Bultfontein and de Beers mines, South Africa. In: Kornprobst J (ed) Proceedings 3rd international kimberlite conference, vol 2, Elsevier, Amsterdam, pp. 43–56
- Wyllie PJ, Lee W-J (1999) Kimberlites, carbonatites and peridotites and silicate-carbonate immiscibility explained in parts of the system CaO-(Na<sub>2</sub>O+K<sub>2</sub>O)-(MgO+FeO)-(SiO<sub>2</sub>+Al<sub>2</sub>O<sub>3</sub>)-CO<sub>2</sub>. In: Gurney JJ, Gurney JL, Pascoe MD, Richardson SR (eds) Proceedings of the VIIth international kimberlite conference, vol. 2, Red Roof Design, pp. 923–932
- Zartman RE, Richardson SH, Gurnet JJ, Moore RO (1977) U-Th-Pb ages of megacryst zircons from the Monastery kimberlite, Free State, South Africa. Extended abstracts, 7<sup>th</sup> international kimberlite conference, Cape Town, South Africa