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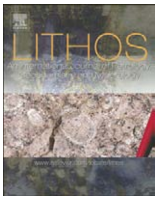

Metasomatic mantle origin for Mbuji-Mayi and Kundelungu garnet and clinopyroxene megacrysts (Democratic Republic of Congo)

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A B S T R A C T

Mbuji-Mayi (east Kasai province) and Kundelungu (Shaba province) are the two kimberlite fields known for a long time in the Democratic Republic of Congo (DRC). Mbuji-Mayi intrudes the Archean basement (Congo–Kasai Craton) and is diamond-rich, whereas Kundelungu cuts across Paleoproterozoic basement (Bangweulu block) and is diamond-poor. The megacryst suites (or discrete nodule suites) of both fields include garnet and clinopyroxene megacrysts. The pyrope-rich megacrysts can be subdivided in three groups on the basis of their Cr contents: low-Cr (0.00–1.79 wt.%Cr_2O_3; $Mg^{\#}$: 72.8–84.0); medium-Cr (1.93–5.16 wt.%Cr_2O_3; $Mg^{\#}$: 76.2–86.3) and high-Cr (5.42–7.10 wt.%Cr_2O_3; $Mg^{\#}$: 79.2–84.6). There are no significant geochemical differences between the garnets from Mbuji-Mayi and from Kundelungu. Polymineral inclusions composed of K-rich hydrated phases (phlogopite and amphibole), fresh glass and Cr-spinels are identified in garnets from all three groups, in both localities, which suggest a common origin. Two groups of diopside megacrysts from Mbuji-Mayi are distinguished on the basis of their Ca content: low-Ca ($Ca^{\#}$: 39.5–42.1; 0.61–0.92 wt.%Cr_2O_3) and medium-Ca ($Ca^{\#}$: 44.1–48.5; 0.41–1.09 wt.%Cr_2O_3); they differ from a third group of high-Cr diopsides ($Ca^{\#}$: 47.1–49.4; 1.31–2.77 wt.%Cr_2O_3). The major element compositions of DRC megacrysts are distinct from those of many other megacryst suites worldwide: the clinopyroxenes are lower in Fe and Ti and higher in Mg and the garnets contain more Cr and significantly less Ti, Fe and Al. These DRC megacryst compositions are intermediate between those of peridotite minerals and those of kimberlite megacrysts from other localities. Most garnets have “normal” REE profiles ($(La/Yb)_N = 0.003–0.027$), whereas clinopyroxenes display relative LREE enrichment ($(La/Yb)_N = 5.1–43.2$). The REE patterns of garnet and clinopyroxene megacrysts are similar to those from metasomatized South African mantle lherzolites. The differences in composition between DRC megacrysts and those from other kimberlites might reflect different modes of formation. Some megacryst suites are related by fractional crystallization processes, the DRC garnet and clinopyroxene megacrysts display geochemical similarities with peridotites and may originate by metasomatic transformation and recrystallization of mantle peridotites.

1. Introduction

The knowledge of the petrologic nature of the deepest parts of the upper mantle is made possible by the study of the rock samples, megacrysts and xenoliths, brought to the surface by kimberlites and alkaline rocks (alnöites, alkali basalts, nephelinites, basanites). Since the 70s, a particular attention has been paid to the megacrysts, also called “discrete nodule suite” (Nixon and Boyd, 1973b) found in kimberlites because of their usefulness in prospecting for potentially diamondiferous kimberlite pipes (Rogers and Grütter, this issue). Many studies have been focused on the possible origins of these megacrysts, particularly on their possible association with their host-rock or with the peridotitic mantle rocks (mostly the sheared peridotites) recovered as xenoliths in the kimberlite breccias (Nixon and Boyd, 1973b; Hops et al., 1992; Burgess and Harte, 1998, 2004). It has been suggested that some megacrysts (the most Mg-rich) could derive from the crumbling of peridotites (e.g. Nixon and Boyd, 1973b). However, this hypothesis is now universally rejected, mainly because: 1) the megacrysts are larger (>1 cm) than the common lherzolite minerals (up to a few mm); 2) some major or minor element binary plots show correlations that are interpreted as evidence for an origin by fractional crystallization and 3) only a part of the wide range of the major element compositions recorded in the megacrysts matches lherzolite mineral compositions (e.g. Nixon and Boyd, 1973b). At present, there is a general agreement that 1) the megacrysts result from fractional crystallization of a silicate melt and 2) the deformation of the sheared peridotites results from the interaction of the megacryst parental melt with the surrounding mantle (Harte and Gurney, 1981; Hops et al., 1992; Burgess and Harte, 1998; Moore and Lock, 2001;

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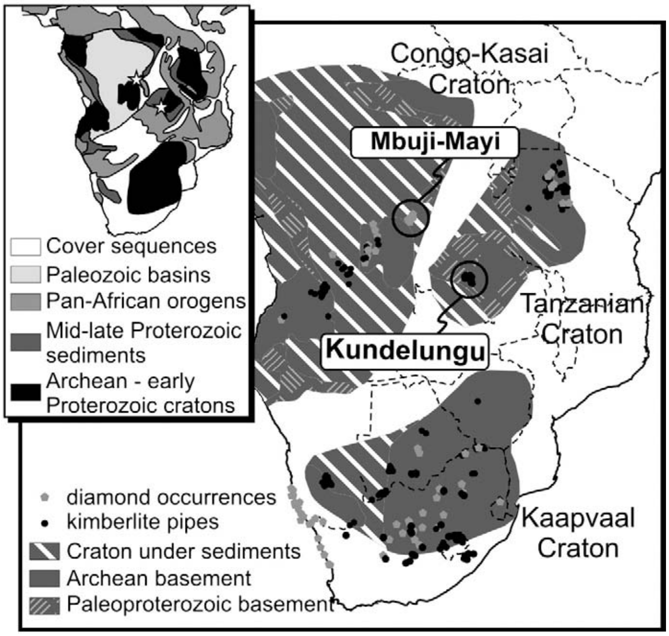

Fig. 1. Location of the Mbuji-Mayi and Kundelungu kimberlite provinces (Democratic Republic of Congo) in the general context of kimberlites of Central and South Africa. Inset from Foster et al. (2001).

Burgess and Harte, 2004; Moore and Belousova, 2005) although sheared peridotites could also result from plastic deformation of the mantle surrounding kimberlite melt pockets at depth due to an excess pressure (Grégoire et al., 2006). However, the link between the megacryst parental magma and the kimberlite magma itself is still debated. Some authors have proposed that there is a genetic link between the two magmas and refer to a kimberlitic or "proto-kimberlitic" melt (Eggler et al., 1979; Gurney et al., 1979; Garrison and Taylor, 1980; Harte and Gurney, 1981; Schulze, 1984; Moore and Lock, 2001; Kostrovitsky et al., 2004; Moore and Belousova, 2005; Kopylova et al., this issue) while others consider the megacryst parental melt to be of foreign origin: crystal mush magmas derived from the asthenosphere (Nixon and Boyd, 1973b), alkali basalts or meimechites (Hops et al., 1992). A more complex process involving the mixing of several sources is also invoked, such as asthenospheric magma interacting (i.e. metasomatic agent) with the subcontinental lithospheric mantle (Davies et al., 2001; Kostrovitsky et al., 2008). There is growing evidence suggesting that the Archean subcontinental lithospheric mantle has been largely metasomatized (Kobussen et al., this issue; Jacob et al., this issue; Viljoen et al., this issue). Many peridotite xenoliths found in kimberlites indeed present evidence of modal metasomatism, characterized by the development of new mineral phases (phlogopite, amphibole, oxides) and/or the replacement of primary minerals. Many xenoliths are also enriched in incompatible trace elements (cryptic metasomatism) (e.g. Hoal et al., 1994; Van Achterbergh et al., 2001; Grégoire et al., 2002, 2003). The proposed fluids/melts responsible for the metasomatic transformations are of various nature and origin and include kimberlitic melts. However, whatever the exact nature of these fluids/melts, they are hydrated and Ti-rich (Harte, 1987).

We have undertaken a detailed investigation of the megacryst suites of two kimberlite provinces in the Democratic Republic of Congo (DRC): the diamond-rich Mbuji-Mayi (formerly Bakwanga) province (Kasai) and the diamond-poor Kundelungu province (Shaba). These megacryst suites themselves have received minimal study (e.g. Mvuemba, 1980; Kampata, 1993) despite their wide diversity. However, the kimberlites of DRC have received more attention (Demaiffe and Fieremans, 1981; Fieremans et al., 1984; Demaiffe et al., 1991), as have the Mbuji-Mayi eclogite nodules (El Fadili and Demaiffe, 1999). In this paper, we focused on the most abundant megacryst phases: garnet and clinopyroxene. Major and trace element compositions are compared with other megacryst suites and with peridotitic minerals.

2. DRC kimberlites

The Mbuji-Mayi kimberlites and their associated diamondiferous deposits have been studied since the 1950s because of the high economical potential of this region (e.g. Meyer de Stadelhofen, 1963; Fieremans, 1966; Mvuemba, 1980). This kimberlite field, exploited since 1920 by the MIBA (Minière de Bakwanga), is located in the east Kasai province and consists of two clusters of pipes that cut across the Archean Congo–Kasai Craton (>2.7 Ga) (Fig. 1). The northern group (Mbuji-Mayi itself) is composed of ten pipes aligned in the east–west direction. Five pipes comprise the southern group (Tshibua-Kalonji) and outcrop about 30 km to the south-west. The diatremes of the northern group cut across three geological units of this region: 1) the Archean Dibaya migmatitic complex (2.8–2.4 Ga); 2) the middle to late Proterozoic sedimentary Mbuji-Mayi Supergroup (1.3–0.95 Ga) and 3) the Cretaceous Lualaba (120 Ma) sedimentary series. The kimberlites of the southern group cut only across the first and third members of this sequence (Demaiffe et al., 1991). A Cretaceous age for the Mbuji-Mayi kimberlites was first suggested by Fieremans (1966) on the basis of stratigraphic relations and was later confirmed by ID-TIMS U–Pb dating on zircon and baddeleyite megacrysts: the average of 45 analyses is 69.8 ± 0.5 Ma (Schärer et al., 1997). The kimberlites contain a wide variety of megacryst mineral species: Mg-ilmenite, diopside, pyrope, zircon, baddeleyite, corundum, Mg-chlorite, kyanite, rutile and rutile–silicate intergrowths. This megacryst suite is obviously heterogeneous and

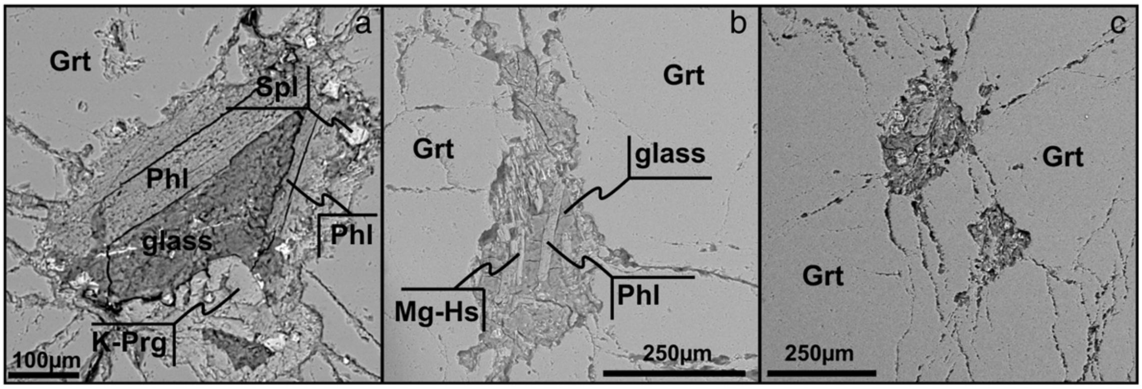

Fig. 2. Backscattered electron microphotographs of inclusions in garnet (Grt) megacrysts. (a) Polygonal shape inclusion made of the association phlogopite (Phl) + Ca-amphibole (K-Pargasite = K-Prg) + fresh glass + Cr-spinels (Spl); (b) non polygonal inclusion made of the association phlogopite (Phl) + Ca-amphibole (Magnesiohastingsite = Mg-Hs) + fresh glass; and (c) K-rich hydrated polycrystalline and glass inclusions are connected to a radial network of fractures.

gathers different mineral species that probably do not belong to a single paragenesis. These megacrysts might be genetically related to the kimberlite (or "proto-kimberlite") magma (e.g. zircon, rutile) or be fragments of xenolith lithologies (e.g. kyanite from kyanite-bearing eclogites; El Fadili and Demaiffe, 1999). The population of xenoliths is largely dominated by eclogite nodules (90%); pyroxenites and peridotites are rare.

The Kundelungu kimberlite field is located in the Katanga (formerly Shaba) province (Fig. 1). Two groups of pipes have been recognized on the eastern and western borders of the Kundelungu Plateau respectively. The eastern group is made of 16 pipes irregularly distributed, while the western group consists of 14 pipes, of which 13 are aligned in N–S direction (Demaiffe et al., 1991; Batumike et al., 2008). The pipes cut across the horizontal sediments of the Kundelungu system (upper part of the Neoproterozoic Katangan Supergroup) and the metasediments of the Kibaran orogenic belt (1.3 ± 0.2 Ga). They certainly also cut across a deeper Paleoproterozoic basement (~1.9 Ga) made of micaschists, metavolcanites and granitoids, belonging to the south-western extension of the Bangweulu block (Ngoyi et al., 1991; Batumike et al., 2007). Because these kimberlites post-date Kundelungu Group deposition and are pre-Miocene, it was thought that they were of Cretaceous age by analogy with the Mbuji-Mayi kimberlites and other African kimberlites (Demaiffe et al., 1991). However, recent in-situ LA-MC-ICP-MS U–Pb radiometric ages obtained on primary groundmass perovskites from two different kimberlite pipes (Kambeli and Msipashi) give a lower Oligocene age of 32.3 ± 2.2 Ma (Batumike et al., 2008). Since the pioneering work of Verhoogen (1938), the Kundelungu kimberlites and their associated megacrysts and xenoliths have not been studied in detail, except in the unpublished PhD Thesis of Kampata (1993). The megacryst suite consists of garnet, olivine, orthopyroxene, clinopyroxene, phlogopite and ilmenite. Unlike Mbuji-Mayi, the mantle xenoliths of the Kundelungu kimberlites are varied and include: abundant peridotites (lherzolites, harzburgites, wehrlites and dunites), a few eclogites and a clinopyroxenite. Both sheared and granular lherzolites (Nixon and Boyd, 1973a) were observed by Kampata (1993).

The Sr and Nd isotopic signatures of the Mbuji-Mayi and Kundelungu kimberlites (Weis and Demaiffe, 1985) are comparable to those of group I kimberlites (Smith, 1983) and suggest a time integrated LIL-depleted upper mantle-source region.

Table 1

Representative major (wt.%) and rare earth element (ppm) analyses of garnet megacrysts from Mbuji-Mayi and Kundelungu kimberlite provinces in the Democratic Republic of Congo.

	Low-Cr pyropes				Medium-Cr pyropes				High-Cr pyropes			
	Mbuji-Mayi		Kundelungu		Mbuji-Mayi		Kundelungu		Mbuji-Mayi		Kundelungu	
	GRA.3	G.18	K.8	Gu.3	MM1.G	G.17	Gu.10	Gu.6	M.2	M.17	Gu.4	K.6
Major element analyses (wt.%) — WDS electron microprobe												
SiO_2	41.51	42.49	42.14	42.05	42.07	42.05	42.01	41.78	42.10	41.71	41.27	41.62
TiO_2	0.55	0.36	0.16	0.10	0.56	0.49	0.28	0.35	0.13	0.24	0.09	0.05
Al_2O_3	21.77	21.45	22.80	22.40	19.97	19.58	21.58	19.68	19.51	18.24	19.06	19.24
Cr_2O_3	0.54	1.12	0.46	1.79	2.95	3.89	2.38	3.84	5.73	6.89	5.52	5.84
FeO	10.80	9.28	10.75	9.64	8.52	7.86	9.23	8.28	7.38	7.22	7.61	8.65
MnO	0.37	0.36	0.47	0.43	0.30	0.34	0.40	0.29	0.37	0.38	0.43	0.53
MgO	19.62	20.97	20.26	20.17	21.49	20.75	20.40	20.59	21.07	20.72	20.73	19.10
CaO	4.10	4.17	3.97	4.34	4.74	5.28	4.59	5.57	4.98	5.27	5.16	6.02
Na_2O	0.14	0.15	0.07	0.04	0.05	0.09	0.04	–	0.04	0.03	0.01	0.01
K_2O	–	0.02	–	–	–	–	–	–	–	0.02	–	–
Total	99.40	100.37	101.08	100.96	100.65	100.33	100.91	100.38	101.31	100.72	99.88	101.06
Number of cations (apfu) calculated on the basis of 12 oxygen atoms and 8 cations												
Si	2.984	3.007	2.971	2.973	2.980	2.998	2.974	2.981	2.980	2.984	2.965	2.984
Ti	0.030	0.019	0.009	0.006	0.030	0.026	0.015	0.019	0.007	0.013	0.005	0.002
Al	1.845	1.789	1.895	1.866	1.667	1.645	1.801	1.655	1.628	1.538	1.614	1.626
Cr	0.031	0.063	0.025	0.100	0.165	0.219	0.133	0.217	0.321	0.390	0.314	0.331
Fe^{2+}	0.534	0.430	0.503	0.486	0.349	0.369	0.454	0.366	0.353	0.347	0.323	0.448
Fe^{3+}	0.116	0.119	0.131	0.083	0.155	0.100	0.093	0.128	0.083	0.085	0.134	0.071
Mn	0.022	0.022	0.028	0.026	0.018	0.021	0.024	0.018	0.022	0.023	0.026	0.032
Mg	2.102	2.212	2.129	2.126	2.269	2.205	2.153	2.190	2.223	2.209	2.220	2.041
Ca	0.316	0.316	0.300	0.329	0.359	0.403	0.348	0.426	0.378	0.404	0.397	0.463
Na	0.020	0.021	0.009	0.006	0.007	0.013	0.005	–	0.005	0.005	0.002	0.001
K	–	0.002	–	–	–	–	–	–	–	0.001	–	–
$Mg^{\#}$	76.4	80.1	77.1	78.9	81.8	82.5	79.8	81.6	83.6	83.7	82.9	79.7
$Ca^{\#}$	13.1	12.5	12.3	13.4	13.7	15.5	13.9	16.3	14.5	15.5	15.2	18.5
Rare earth element analyses (ppm) — LA-ICP-MS												
Average of (*n*) analyses			(6)	(4)		(5)	(3)	(5)			(4)	(5)
La			0.60	0.01		0.02	–	0.08			0.03	0.01
Ce			1.03	0.08		0.24	0.10	0.54			0.38	0.17
Nd			0.90	0.41		1.19	–	1.44			1.56	1.62
Sm			0.71	0.51		1.10	0.10	1.00			1.10	0.82
Eu			0.34	0.14		0.50	0.19	0.50			0.58	0.41
Gd			1.35	1.75		2.12	1.46	2.21			2.41	1.04
Dy			2.89	2.57		3.25	4.22	3.76			2.87	0.99
Er			2.55	1.67		1.91	4.31	2.83			1.75	0.86
Yb			2.79	2.06		1.54	3.21	2.04			1.24	1.28
ΣREE			15.26	10.75		13.55	16.09	16.59			13.49	8.09
$(La/Yb)_N$			0.145	0.003		0.008	–	0.027			0.015	0.006

Fe^{3+} calculated by charge balance.

$Mg^{\#} = Mg/(Mg + \Sigma Fe)$ (apfu).

$Ca^{\#} = Ca/(Ca + Mg)$ (apfu).

$(La/Yb)_N$ calculated using chondritic values of McDonough and Sun (1995).

3. Megacryst descriptions and analytical procedure

A large collection of megacrysts from DRC has been provided by the late Dr. Carlos Fieremans (former executive officer of the MIBA). Additional samples were provided by Dr. Mark Fieremans. Most Mbuji-Mayi megacrysts were recovered after the first stage of crushing at the mine. Only garnets are available from Kundelungu.

The garnet samples from both regions are mostly rather angular monocrystalline specimens of various colors: pale orange, pink, purple red or brown. Their diameter ranges from 2 mm to 2–3 cm. Larger garnets are more often fractured than smaller garnets. Three polycrystalline garnet-aggregates have been identified in Mbuji-Mayi: two of them are composed of an aggregate of small (1–6 mm) isomorphic grains, while the third one consists of a large garnet grain with small isomorphic grains growing in the fractures.

Polished sections of garnet were examined by SEM to identify inclusions. Inclusions have been observed in garnets from both kimberlite fields. They are typically polymineralic, composed of Ca-amphibole + phlogopite + Cr-spinels + fresh glass (Fig. 2a and b). Some have a polygonal shape (Fig. 2a); others are rounded. They are all connected to a radial network of fractures or thin veins (Fig. 2c).

The clinopyroxene megacrysts from Mbuji-Mayi occur only as individual crystals (2 mm to 1.5 cm). The more altered crystals are light green, whereas the freshest samples are characterized by a bright dark "bottle-green" or are sometimes more "apple green". They are mostly angular fragments.

Garnet and clinopyroxene megacrysts were prepared as 150 µm thick sections and/or in epoxy mounts. The major element compositions have been determined by WDS electron microprobes (CAMECA SX100 and SX50) equipped with four wavelength dispersive spectrometers at the CAMPARIS section of the University of Paris 6 (France) and at the CAMST of the University of Louvain-la-Neuve (Belgium) using a combination of natural and synthetic mineral standards.

Trace elements (REE) were analyzed by using in-situ LA-ICP-MS. The data were collected by the coupling of an UV Fisons laser ablation microprobe and a VG Elemental Plasma Quad (PQ2 Turbo Plus) ICP-

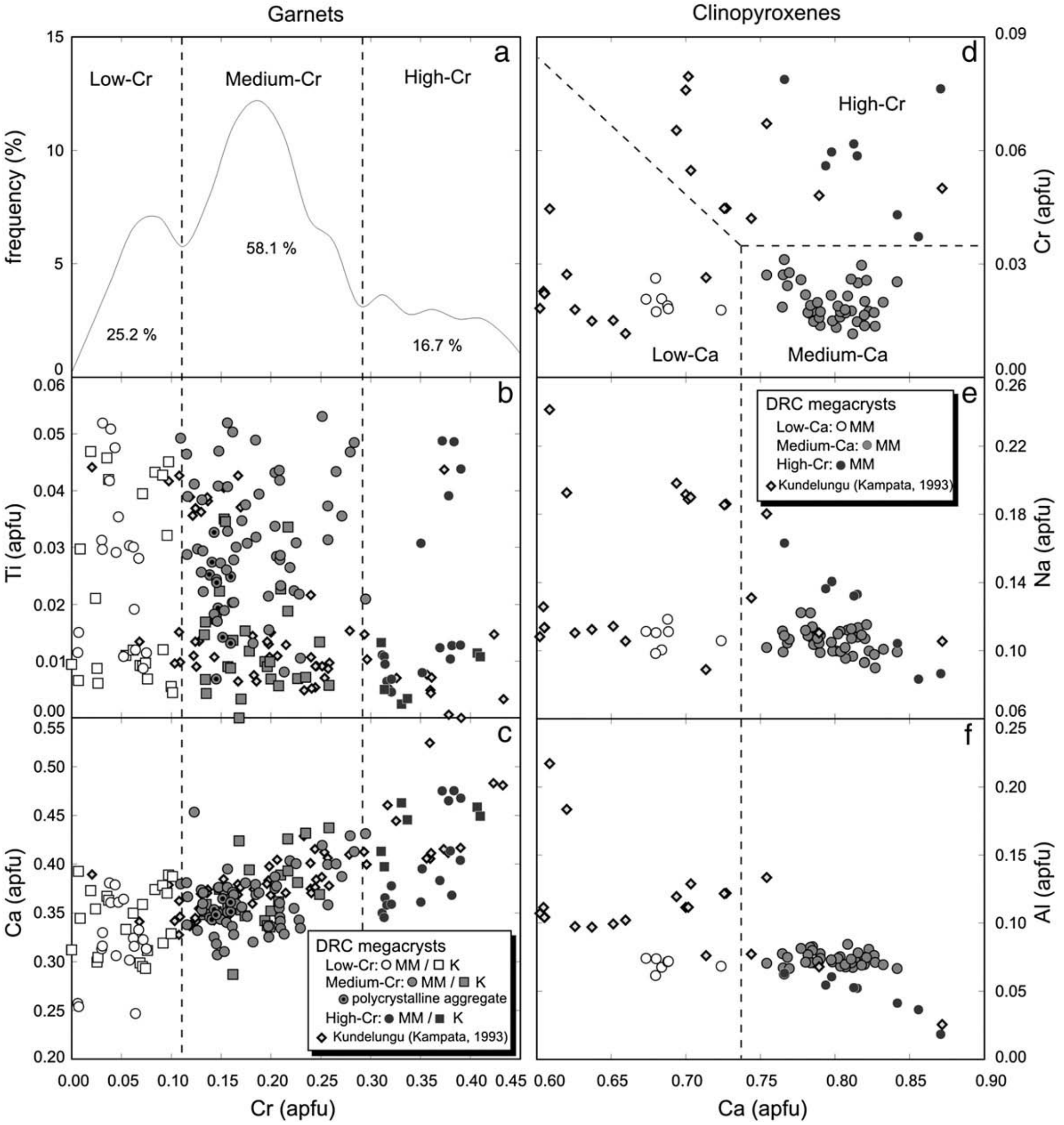

Fig. 3. a–b–c. Binary plots illustrating the three groups of pyropes for Mbuji-Mayi (MM, circles) and Kundelungu (K, squares) megacrysts: (a) frequency distribution of the garnets as a function of their Cr content; (b) Ti (apfu) vs. Cr (apfu); (c) Ca (apfu) vs. Cr (apfu). d–e–f. Binary plots illustrating the three groups of diopside megacrysts in Mbuji-Mayi kimberlites: (d) Cr (apfu) vs. Ca (apfu); (e) Na (apfu) vs. Ca (apfu); (f) Al (apfu) vs. Ca (apfu). Data for Kundelungu megacrysts (Kampata, 1993) are added for comparison.

MS (Musée Royal de l'Afrique Centrale at Tervuren, Belgium). Analytical methods, detection limits, precision and accuracy are the same as those detailed in Féménias et al. (2003).

4. Major element compositions

4.1. Garnet composition

For the two kimberlite fields, representative garnet megacrysts were chosen based on size and color variations: 50 from Mbuji-Mayi and 41 from Kundelungu. Representative analyses are given in Table 1. All analyzed garnets are pyropes (>65 mol% prp) with rather constant grossular (9–15 mol%) and low (<1 mol%) spessartine contents. The ranges of major element compositions for the two kimberlite provinces overlap completely. In numerous kimberlite provinces, garnet megacrysts display a wide range of Cr content which allows subdividing the population into two suites: a "Cr-poor" (0.03–4.8 wt.%Cr_2O_3) and a "Cr-rich" (6.3–13 wt.%Cr_2O_3) suite (e.g. Eggler et al., 1979). The distribution of the DRC garnet population as a function of its Cr content allows the separation of three groups of pyropes (Fig. 3a):

- low-Cr: 0.00–0.10 Cr (apfu); Mg$^{\#}$: 72.8–84.0;
- medium-Cr: 0.11–0.29 Cr (apfu); Mg$^{\#}$: 76.2–86.3; and
- high-Cr: 0.31–0.41 Cr (apfu); Mg$^{\#}$: 79.2–84.6,

which represent respectively 25.2, 58.1 and 16.7% of the population.

These subdivisions are valid for the garnets from both localities (Fig. 3a–b–c; data from Kampata, 1993 are added for comparison). Indeed, the Mbuji-Mayi and Kundelungu garnet megacrysts display similar Cr composition ranges: 0.01–0.39 Cr (apfu) and 0.00–0.41 Cr (apfu) respectively. The ranges of Ti content of the different groups of megacrysts are similar, whatever their origin (Fig. 3b). For the whole garnet population, a positive correlation is observed between the Ca and Cr contents over broad ranges (Fig. 3c), as it is observed for the peridotitic garnets. The small garnet grains of two polycrystalline aggregates from Mbuji-Mayi have exactly the same composition as the monocrystalline megacrysts of the medium-Cr group (Fig. 3b and c). There is no relation at all between the size of the grain and its composition, nor between the presence (or absence) of inclusions and the composition. The chemical compositions of the Kundelungu garnets from this study are similar to those from Kampata (1993) (Fig. 3a–b–c).

4.2. Clinopyroxene composition

Clinopyroxene megacrysts (44 samples) from Mbuji-Mayi only were selected and analyzed (see representative analyses in Table 2). No clinopyroxene megacrysts were available from Kundelungu. All analyzed clinopyroxenes are diopsides: the Na content is low, <0.16 (apfu), with most <0.11 (apfu); the Ti and Al contents are also low: 0.001–0.007 (apfu) and 0.02–0.08 (apfu) respectively.

The diopside population has been divided into two groups, a low-Cr (<0.03 apfu) and a high-Cr (0.04–0.08 apfu) group (Fig. 3d–e–f). The low-Cr diopsides are further subdivided in a low-Ca group and a medium-Ca group. The chemical characteristics of these groups are:

- low-Ca: 0.67–0.72 Ca (apfu); 0.02–0.03 Cr (apfu);
- medium-Ca: 0.75–0.84 Ca (apfu); 0.01–0.03 Cr (apfu);
- high-Cr: 0.77–0.87 Ca (apfu); 0.04–0.08 Cr (apfu).

The Mg$^{\#}$ of the high-Cr diopsides (Mg$^{\#}$ 92.2–93.5) is slightly but significantly higher than in the two other groups (low-Ca: Mg$^{\#}$ 89.3–90.8; medium-Ca: Mg$^{\#}$ 90.6–92.1). The high-Cr group also displays trends of increasing Al and Na contents with decreasing Ca content, contrary to what is observed for the two other groups, for which the Na and Al contents do not vary much with the Ca content (Fig. 3e and f).

The three groups contain crystals of different sizes and colors (fresh and more altered samples) but generally the more "apple-green" grains belong to the Cr-rich group. All the grains of the low-Ca group are rather small (<1 cm).

The Kundelungu diopside megacrysts reported by Kampata (1993) display the same range of Cr content (Fig. 3d). Some samples are comparable to the low-Ca group but extend to lower Ca values and there is no equivalent of the medium-Ca group at Kundelungu (Fig. 3d). Kundelungu diopsides that are similar to the high-Cr group from Mbuji-Mayi in term of Cr content, extend to lower Ca and higher Na and Al contents (Fig. 3d–e–f).

5. Rare earth elements

5.1. Garnet pattern

Garnet megacrysts from DRC have low REE abundances (ΣREE: 7.8–16.6 ppm; Table 1). No trends are observed between the pyrope groups (based on Cr content) and the REE content. Most pyropes display "normal" REE patterns, with relative LREE depletion [$(La/Yb)_N$ = 0.003–0.027)] (Fig. 4a), but some do show a slight LREE enrichment (La_N ~ 2.5–3). Garnets have both positive Eu anomalies (Eu/Eu* ~ 1.35)

Table 2
Representative major (wt.%) and rare earth element (ppm) analyses of clinopyroxene megacrysts from Mbuji-Mayi kimberlite field (Kasai province, DRC).

Mbuji-Mayi									
	Low-Ca diopsides			Medium-Ca diopsides			High-Cr diopsides		
	F2.7	F2.3	F1.5	F2.4	F2.2	F2.5	F1.10	Di.B	Di.C
Major element analyses (wt.%) — WDS electron microprobe									
SiO_2	54.43	54.99	54.01	55.49	55.25	54.35	56.10	55.11	55.32
TiO_2	0.12	0.14	0.20	0.14	0.23	0.16	0.08	0.16	0.23
Al_2O_3	1.73	1.59	1.60	1.81	1.63	1.61	0.86	1.28	1.49
Cr_2O_3	0.61	0.73	0.62	0.60	0.63	1.03	1.31	1.96	2.77
FeO	3.96	3.82	3.85	2.89	2.58	2.76	2.30	2.46	2.20
MnO	0.21	0.10	0.15	–	0.10	0.06	0.06	0.08	0.14
MgO	18.81	18.83	18.40	17.17	16.92	16.71	16.69	16.39	16.05
CaO	17.52	17.65	18.61	20.23	20.79	20.98	22.21	20.48	19.87
Na_2O	1.57	1.43	1.49	1.56	1.45	1.54	1.20	1.94	2.33
K_2O	0.04	0.03	0.04	–	0.04	0.01	0.01	0.02	–
Total	99.00	99.31	98.97	99.89	99.62	99.21	100.82	99.88	100.40
Number of cations (apfu) calculated on the basis of 6 oxygen atoms and 4 cations									
Si	1.972	1.988	1.960	2.001	2.001	1.977	2.018	1.994	1.991
^{IV}Al	0.028	0.012	0.040	0.000	0.000	0.023	0.000	0.006	0.009
^{VI}Al	0.045	0.056	0.029	0.077	0.070	0.047	0.036	0.048	0.054
Ti	0.003	0.004	0.006	0.004	0.006	0.004	0.002	0.004	0.006
Cr	0.017	0.021	0.018	0.017	0.018	0.030	0.037	0.056	0.079
Fe^{3+}	0.073	0.031	0.090	0.004	0.004	0.048	0.000	0.032	0.026
Fe^{2+}	0.047	0.085	0.027	0.083	0.074	0.036	0.069	0.043	0.040
Mn	0.006	0.003	0.004	–	0.003	0.002	0.002	0.003	0.004
Mg	1.016	1.015	0.996	0.923	0.913	0.906	0.895	0.884	0.861
Ca	0.680	0.684	0.724	0.782	0.807	0.818	0.856	0.794	0.766
Na	0.111	0.101	0.106	0.109	0.102	0.109	0.083	0.136	0.163
K	0.002	0.002	0.002	–	0.002	0.001	0.000	0.001	–
Mg$^{\#}$	89.4	89.8	89.5	91.4	92.1	91.5	92.8	92.2	92.8
Rare earth element analyses (ppm) — LA-ICP-MS									
Average of (n) analyses	(5)	(4)		(6)	(4)	(5)		(4)	(5)
La	1.49	1.90		2.19	1.81	3.36		4.56	3.56
Ce	4.50	6.53		7.53	6.52	12.02		14.75	11.32
Nd	4.15	4.29		6.08	5.04	10.32		14.54	10.04
Sm	1.20	1.17		1.54	1.58	2.56		3.47	2.54
Eu	0.43	0.32		0.48	0.43	0.83		0.97	0.74
Gd	1.29	0.55		1.45	1.19	1.76		2.49	1.53
Dy	0.66	0.19		0.77	0.65	0.94		1.07	0.73
Er	0.34	0.03		0.32	0.25	0.23		0.37	0.31
Yb	0.15	0.01		0.08	0.24	0.05		0.17	0.23
ΣREE	15.30	15.99		21.95	19.03	34.40		45.52	33.23
$(La/Yb)_N$	6.55	220.65		18.50	5.10	43.17		18.44	10.53

Fe^{3+} calculated by charge balance.
Mg$^{\#}$ = Mg / (Mg + ΣFe) (apfu).
$(La/Yb)_N$ calculated using chondritic values of McDonough and Sun (1995).

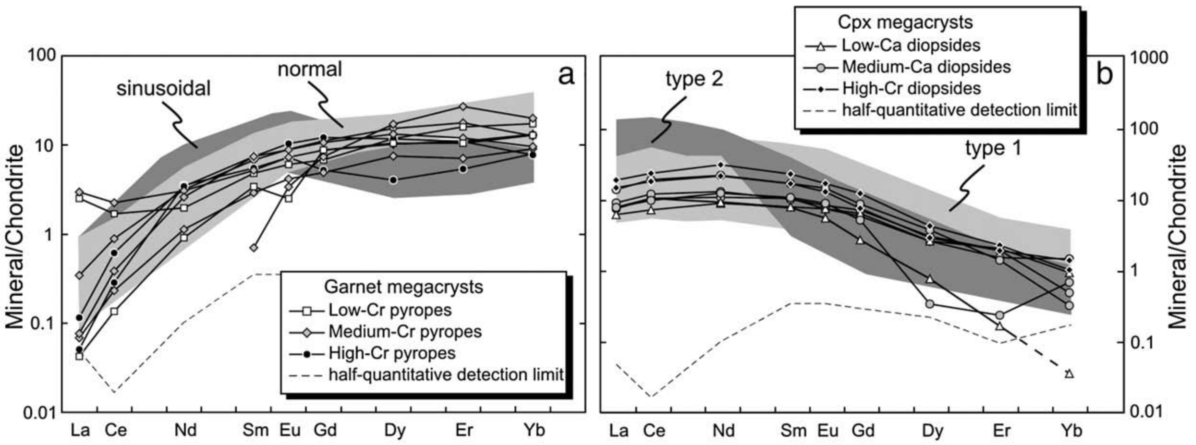

Fig. 4. a. Chondrite-normalized REE patterns for DRC garnet megacrysts compared to ranges of garnet patterns (normal and sinusoidal) in peridotite xenoliths from South African kimberlites (Hoal et al., 1994) and b. Chondrite-normalized REE patterns for DRC clinopyroxene megacrysts. Comparison with type 1 and 2 clinopyroxene profiles in garnet lherzolite from Kaapvaal kimberlites (Grégoire et al., 2003). The REE concentrations are normalized to chondrite values of McDonough and Sun (1995).

and negative (Eu/Eu*~0.46). The presence or absence of K-rich inclusions in the garnet does not correlate with REE patterns: e.g. a low-Cr pyrope that is LREE-enriched contains inclusions, while a similarly LREE-enriched medium-Cr pyrope does not.

The REE concentrations and profiles of DRC garnet megacrysts are similar to those reported by Hoal et al. (1994) for garnets in peridotite xenoliths from South African kimberlites (Fig. 4a) for which normal and sinusoidal REE patterns were identified.

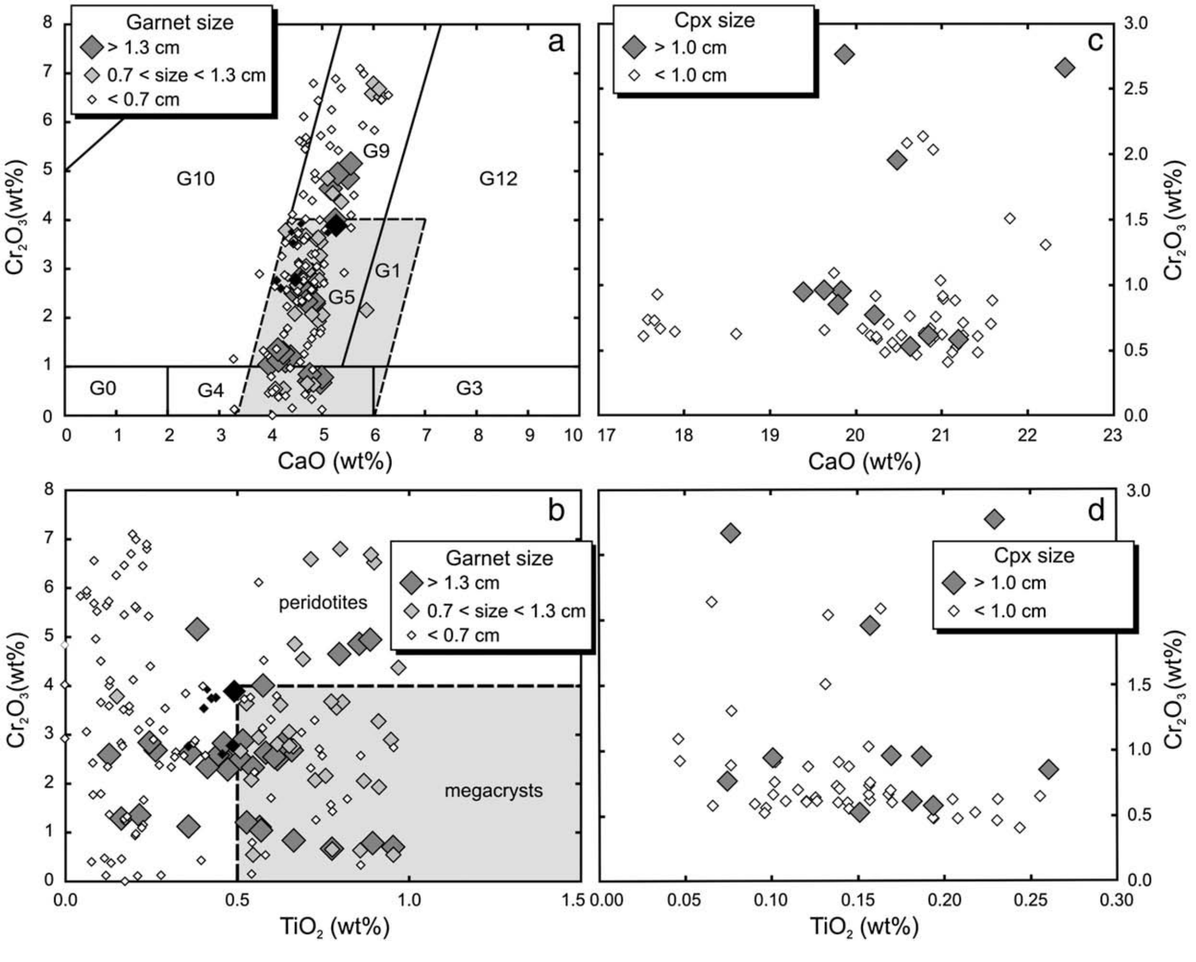

Fig. 5. a–b. Composition of the DRC garnet megacrysts sorted by size in classification diagrams. (a) Cr_2O_3 vs. CaO (wt.%) binary diagram of Grütter et al. (2004): G0 = unclassified; stippled parallelogram G1 = megacrysts; G3 = eclogites; G4 and G5 = pyroxenites; G9 = lherzolites; G10 = harzburgites; G12 = wehrlites and (b) Cr_2O_3 vs. TiO_2 (wt.%) binary diagram of Schulze (2003). Black symbols highlight the samples that fall in the megacryst group G1 (a) and in the peridotite field (b). c–d. By analogy to the garnets, composition of Mbuji-Mayi clinopyroxene megacrysts sorted by size in the Cr_2O_3–CaO (wt.%) (c) and Cr_2O_3–TiO_2 (wt.%) (d) diagrams.

5.2. Clinopyroxene pattern

The DRC clinopyroxene megacrysts are also REE-poor (ΣREE: 15.3–45.5 ppm; Table 2): low-Ca diopsides have 15 ppm, medium-Ca diopsides vary between 18.3 and 34.4 ppm and high-Cr diopsides have the highest abundances (33.2–45.5 ppm). There is certainly a relation between the REE and the major element contents (Fig. 4b), although the restricted number of data requires caution. All the diopsides display relative LREE enrichment, displaying high $(La/Yb)_N$ values (5.1–43.2, Fig. 4b).

The REE concentrations and profiles of Mbuji-Mayi diopside megacrysts are also similar to those reported for two types (1 and 2) of diopsides in garnet lherzolites from some Kaapvaal kimberlites (Grégoire et al., 2003; Fig. 4b).

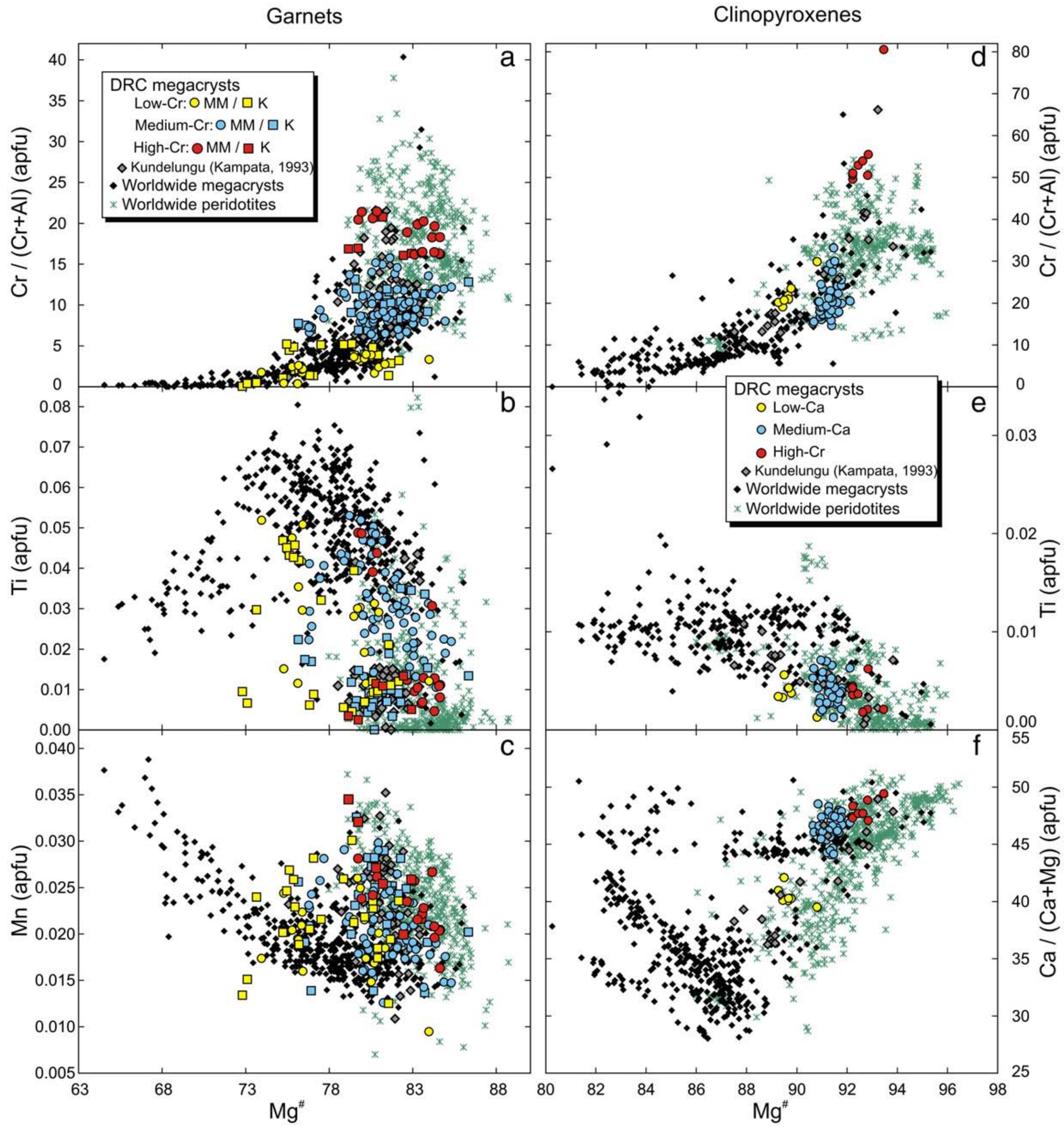

Fig. 6. a–b–c. Binary plots allowing the simultaneous comparison between DRC garnet megacrysts and garnet megacrysts from DRC (Kundelungu) (Kampata, 1993); South Africa (Monastery–Lace–Kaalvallei–Jagersfontein–Premier–Franck Smith–Uintjesberg–Witberg–Schuller) (Gurney et al., 1979; Bell and Rossman, 1992; Hops et al., 1992; De Bruin, 1993; Schulze, 1997; Bell and Moore, 2004; De Bruin, 2005); Namibia (Koherab) (Bell and Rossman, 1992); Lesotho (Kao Mine–Letseng-la-terae–Thaba Putsoa–Solane) (Nixon and Boyd, 1973b; Schulze, 1997); USA (Iron Mountain–State Line district–Winkler–Lone Tree–Fayette County–Elliott County) (Eggler et al., 1979; Garrison and Taylor, 1980; Hunter and Taylor, 1984; Schulze, 1997); Australia (Pine Creek 01) (Schulze, 1997); Russia (Griba pipe) (Kostrovitsky et al., 2003, 2004); Canada (Tunraq); Siberia (Kuonam) and Botswana (Orapa) (Mitchell, 1986) and peridotite garnets from Canada (Ekati diamond Mine Property) (Menzies et al., 2004); Venezuela (Guaniamo kimberlites) (Schulze et al., 2006); USA (Homestead kimberlite) (Hearn, 2004); DRC (Kundelungu) (Kampata, 1993), Lesotho (Letseng-la-terae–Thaba Putsoa–Mothae) (Bloomer and Nixon, 1973; Nixon and Boyd, 1973a; Moore and Lock, 2001; Coussaert, 2005) and South Africa (Bultfontein–Monastery–Premier–Jagersfontein) (Grégoire et al., 2003). d–e–f. Binary plots allowing the simultaneous comparison between Mbuji-Mayi clinopyroxene megacrysts and clinopyroxene megacrysts from DRC (Kundelungu) (Kampata, 1993); South Africa (Monastery–Jagersfontein–Kimberley–Premier–Schuller–Uintjesberg–Witberg) (Bell and Moore, 2004 {for f}; Whitelock, 1973; Gurney et al., 1979; Hops et al., 1992; De Bruin, 1993, 2005); Lesotho (Solane–Thaba Putsoa–Letseng-la-terae) (Bloomer and Nixon, 1973; Nixon, 1973; Nixon and Boyd, 1973b); Botswana (Orapa) (De Bruin, 1993); Russia (Griba pipe) (Kostrovitsky et al., 2003, 2004); USA (Iron Mountain–State Line district–Fayette County–Elliott County) (Eggler et al., 1979; Garrison and Taylor, 1980; Hunter and Taylor, 1984) and peridotite clinopyroxenes from Canada (Ekati diamond Mine Property) (Menzies et al., 2004); USA (Homestead) (Hearn, 2004); DRC (Kundelungu) (Kampata, 1993); Lesotho (Letseng-la-terae–Thaba Putsoa–Mothae) (Bloomer and Nixon, 1973; Nixon and Boyd, 1973a; Moore and Lock, 2001; Coussaert, 2005) and South Africa (Bultfontein–Monastery–Premier–Jagersfontein–Kimberley) (Bell and Moore, 2004 {for f}; Grégoire et al., 2003).

6. Discussion

6.1. Restrictive megacryst definition?

Although researchers attempt to connect the definition of the word "megacrysts" to genetic processes, originally, this term referred to crystals of uncertain origin that are more than 1 cm in size (Dawson, 1980; Mitchell, 1995). For a study focused on the megacrysts from a given locality, only the samples of sufficient size (>1 cm after Dawson, 1980), which are classically regarded as being coarser than mantle peridotite minerals (i.e. <4 mm; after Hunter and Taylor, 1984), should be taken into account. However, many megacrysts are obtained from diamond mining operation centers that process the primary kimberlite material and the grain size of all the crystals, including megacrysts and peridotite-derived fragments, is mechanically reduced. The grain size criterion is then not self-sufficient and several chemical discrimination approaches, which provide support for diamond exploration, have been proposed to pick out "true megacrysts" independently from their original size (e.g. Schulze, 2003; Grütter et al., 2004 for garnet discrimination diagrams). Compositional fields have thus been established after comparison of composition of some megacrysts (e.g. clinopyroxene, orthopyroxene, olivine and garnet) with peridotites.

Nevertheless, regarding natural grain size of mantle peridotite xenoliths in African kimberlites (some of them displaying crystal sizes up to 2.5 cm, e.g. Coussaert, 2005), the definition of "true" megacryst and the assumption of size-chemical discrimination need to be reconsidered and discussed.

In this study, most garnet grains available are in fact smaller than 1 cm in diameter and do not fit the strict definition of megacrysts. In order to determine if size difference is correlated to differences in chemical compositions, representative samples of various sizes and colors were chosen for major element composition analyses. The DRC garnets are plotted in discriminatory diagrams (Fig. 5a and b). Garnets cover a wide range of composition domains, regardless of size (Fig. 5a). Some true megacrysts (>1 cm) fall in the lherzolite group G9 and some small (<1 cm) grains fall in the megacryst group G1 (Fig. 5a). Moreover, garnets of different sizes (highlighted in black in Fig. 5a and b), which are in the megacryst group (G1) in Fig. 5a, plot in the peridotite composition range in Fig. 5b. The Ti content, which is commonly used as a discriminatory element in the classification schemes, is quite low in DRC megacrysts: <0.053 and <0.047 Ti (apfu) for Mbuji-Mayi and Kundelungu garnets respectively (Figs. 3b and 5b). The DRC garnet megacrysts overlap both the mantle and megacryst fields of classification.

On the basis of these data, there is no reason to ignore small garnet grains based on their size, nor large crystals because of their chemical affinity to mantle rocks. As there is no relation between the size of the garnet crystals and their composition, all the samples have to be considered together. Crystals of small size most probably result either from the mechanical abrasion of larger crystals during the ascent in the kimberlite magmas or from the crushing during the processing at the mine.

Similarly, clinopyroxene crystals of different sizes and colors have been chosen for major element composition analyses. They are plotted in Cr_2O_3–CaO (wt.%) and Cr_2O_3–TiO_2 (wt.%) binary plots (Fig. 5c and d) to assess if the smaller samples could come from the fragmentation of the larger. There is no "true" megacryst (>1 cm) that is as Ca-poor (Fig. 5c), but there is no difference on the basis of the Ti and Cr contents between the small and large crystals (Fig. 5d), thus the population of clinopyroxene megacrysts is considered together.

6.2. DRC megacrysts: mantle or megacryst compositional fields?

The major element compositions of DRC garnet and clinopyroxene megacrysts are compared with those of megacrysts from other kimberlite occurrences worldwide and with those of minerals from kimberlite-related mantle peridotite xenoliths. Garnet and clinopyroxene megacryst compositions from kimberlites of DRC, South Africa, Lesotho, Botswana, USA and Russia, and also from Canada, Australia, Siberia and Namibia for garnets only are shown in Fig. 6. The compositions of garnet and clinopyroxene from peridotite xenoliths from various localities (Lesotho, Canada, USA, DRC and South Africa) have also been plotted (Fig. 6).

The DRC garnets do not show the usual trends of correlated Cr/(Cr + Al) and $Mg^{\#}$ [Mg/(Mg + ΣFe) (apfu)] or of inversely correlated Ti and $Mg^{\#}$ (Fig. 6a and b), which are commonly interpreted as typical fractional crystallization trends (Gurney et al., 1979; Garrison and Taylor, 1980; Hops et al., 1992). The DRC garnets have lower Ti (<0.05 apfu), Fe (<0.76 apfu) and Al (<1.97 apfu) but higher Cr (up to 0.41 apfu) contents than megacryst suites worldwide (Fig. 6a and b). The DRC garnet megacrysts have compositions intermediate between those of megacryst suites and those of mantle peridotites: they are richer in Fe and Ti and poorer in Cr than the corresponding minerals in peridotite xenoliths (Fig. 6a and b). Moreover, even if the ranges of Mn content of megacrysts and of garnet peridotites are similar (0.01 to 0.04 apfu), the trend of inversely correlated Mn and $Mg^{\#}$ defined by megacrysts worldwide has a flatter slope than that defined by peridotite garnets (Fig. 6c). It is difficult to reconcile the DRC garnet megacrysts to either trend.

The Mbuji-Mayi diopside megacrysts are richer in Mg (0.86–1.04 apfu) and Cr (up to 0.8 apfu) and depleted in Ti (<0.007 apfu) and Fe (<0.12 apfu) (Fig. 6d–e–f) when compared to other megacryst suites. Besides, they do not display the simultaneously decreasing Cr/(Cr + Al)–$Mg^{\#}$ trend (Fig. 6d) commonly observed in clinopyroxene megacrysts (Eggler et al., 1979; Garrison and Taylor, 1980; Hops et al., 1992) and interpreted as an evidence of fractional crystallization. The extreme compositions (Fig. 6d) and the trends observed for the high-Cr clinopyroxene group (Fig. 3e and f), which might result from a charge equilibrium involving Na–Ca–Al to balance excess Cr and/or from more complex exchange or reequilibration processes, remains comparable to compositional fields and trends observable for megacrysts from Grib kimberlite pipe (Russia; Kostrovitsky et al., 2004) and metasomatic clinopyroxenes in peridotite xenoliths from the Homestead kimberlite (Wyoming; Hearn, 2004) and Thaba Putsoa pipe (Lesotho; Nixon and Boyd, 1973a). Furthermore, the negative correlation between Ca/(Ca + Mg) and $Mg^{\#}$ (Fig. 6f), observed in many other suites (Gurney et al., 1979; Hops et al., 1992; Bell and Moore, 2004), is not present for the Mbuji-Mayi clinopyroxenes. It is interesting to note that, despite the differences discussed in Section 4.2, the Kundelungu diopside megacrysts are compositionally closer to those from Mbuji-Mayi, than to those from other occurrences (Fig. 6d–e–f). The rough Cr/(Cr + Al)–$Mg^{\#}$ correlation trend extends to higher values than for the megacrysts worldwide (Fig. 6d). A Ca/(Ca + Mg)–$Mg^{\#}$ trend is also apparent but clearly differs from that of the megacrysts from other localities (Fig. 6f). By comparison to peridotitic clinopyroxenes from kimberlite-related xenoliths, the Mbuji-Mayi megacrysts are also Fe- and Ti-rich and quite low in Cr (Fig. 6d–e–f). These observations are also valid for the Kundelungu diopsides. So, it is clear that the Kundelungu diopside megacrysts share more characteristics with those from Mbuji-Mayi than with those from other megacryst suites worldwide.

6.3. Magmatic and/or metasomatic origin for DRC garnet and clinopyroxene megacrysts?

The DRC garnet and clinopyroxene megacrysts have compositions intermediate between the megacrysts of assumed magmatic origin from other kimberlite occurrences and the minerals in mantle peridotites (Fig. 6):

- they display neither classical magmatic-related megacryst compositions nor trends interpretable in terms of fractional crystallization, contrary to what is observed in other megacryst suites;

- they are compositionally similar to mantle peridotite minerals; their significant enrichment in Fe and Ti might be related to what is commonly observed in metasomatically modified mantle peridotites.

Such a *Fe–Ti metasomatism* has been recorded for the deep coarse garnet harzburgites and sheared garnet lherzolites derived from the Archean mantle (Burgess and Harte, 1998). Because the typical megacrysts worldwide are strongly richer in Fe and Ti than the peridotite minerals (Fig. 6b and e), it has been proposed that the "megacryst parental melt" is responsible for this enrichment (e.g. Burgess and Harte, 1998). Moreover, the peridotites from the Archean subcontinental lithospheric mantle are often characterized by enrichment in incompatible trace elements, as the LREE (Shimizu, 1975; Hoal et al., 1994; Grégoire et al., 2003; Burgess and Harte, 2004). This enrichment is sometimes also accompanied by the development of new mineral phases (modal metasomatism) rich in K, like phlogopite and K-richterite (e.g. Hawkesworth et al., 1990), characterizing a *K-LREE metasomatism*.

K-rich hydrated polymineral inclusions were observed in many garnet megacrysts from DRC. These inclusions also contain glassy portions. The composition of the glasses is too silica-rich (43–54 wt.% SiO_2) and alkaline-poor (Na_2O+K_2O+CaO: 0.75–3.00 wt.%) to correspond to a typical kimberlite composition. Moreover, the host magma of the megacrysts is not thought to be responsible for the development of these inclusions as veinlets of host magma have never been observed infiltrating the megacrysts. These inclusions are tentatively interpreted as resulting from the destabilization of an unknown original phase (K-amphibole?) that has been equilibrated with its host garnet during a K_2O- and H_2O-rich fluid and/or liquid modal metasomatic event. The similarities of REE profiles and contents observed for the DRC garnet and clinopyroxene megacrysts and the corresponding minerals in metasomatically modified mantle peridotites is striking (Fig. 4). The normal and sinusoidal profiles observed in garnets from peridotite xenoliths from South African kimberlites are respectively interpreted as mantle metasomatized garnets that were, or were not, reequilibrated with a LREE-enriched metasomatic agent (mantle fluid or melt?) (Hoal et al., 1994; Fig. 4a). Most DRC garnets have normal profiles (Fig. 4a). Moreover, various Eu anomalies (both positive and negative) similar to those observed in some DRC garnets (Fig. 4a) have been reported from garnets in peridotite xenoliths and have been interpreted as resulting from variations in redox conditions related to metasomatic events (Griffin and O'Reilly, 2007). Two different REE profiles (the types 1 and 2) have been identified in diopsides from garnet lherzolites from different Kaapvaal kimberlites (Grégoire et al., 2003; Fig. 4b). Both are LREE enriched, but type 2 is more enriched than type 1. The Mbuji-Mayi clinopyroxenes have profiles similar to those of type 1 clinopyroxenes (Fig. 4b). The liquids which are proposed to be responsible for the metasomatic enrichments in both types of clinopyroxene are highly alkaline mafic silicate melts (Grégoire et al., 2003). Besides, similarities have been shown between type 1 clinopyroxene profiles and those in PIC rocks (Phlogopite–Ilmenite–Clinopyroxene) found as xenoliths in kimberlite breccias, while type 2 clinopyroxene profiles are closer to those of MARID (Micas–Amphibole–Rutile–Ilmenite–Diopside) suite xenoliths (Grégoire et al., 2002, 2003). PIC rocks have Sr and Nd isotopic signatures similar to group 1 kimberlites (Mbuji-Mayi and Kundelungu kimberlites belong to group 1 kimberlites, after Weis and Demaiffe, 1985) and the MARID rocks are isotopically similar to group 2 kimberlites. Type 1 mantle clinopyroxenes might have crystallized from, or been completely reequilibrated with, a metasomatic liquid that is genetically related to group 1 kimberlites; the same holds for type 2 clinopyroxenes and group 2 kimberlites (Grégoire et al., 2003).

The similarities between the REE concentrations and profiles of minerals from metasomatically modified mantle peridotites and the DRC garnet and clinopyroxene megacrysts suggest that the latter have been affected by, or are the products of, a metasomatic process.

7. Conclusions

Although the Mbuji-Mayi and Kundelungu kimberlites are of different ages (Cretaceous and lower Oligocene), cut across basements of various ages (Archean and Paleoproterozoic) and have different diamond grades, their garnet and clinopyroxene megacrysts have comparable ranges of major element compositions.

- The DRC garnet megacrysts display lower Ti, Fe and Al and higher Cr contents than other garnet megacrysts worldwide, while clinopyroxenes display higher Mg and Cr and lower Ti and Fe contents.
- These megacrysts have compositions intermediate between those of minerals from mantle peridotites and those of magmatic-related megacrysts.
- The DRC megacrysts do not display the classical bi-element correlations observed for many other megacryst suites that are commonly interpreted as resulting from fractional crystallization processes.
- Garnets and clinopyroxenes display low REE contents and patterns, indistinguishable from metasomatically modified mantle compositions.

Garnet and clinopyroxene megacrysts from DRC kimberlites are thus interpreted as resulting from a metasomatic event that affected the peridotites from the cratonic mantle. This metasomatism enriched the mantle minerals in K, Fe, Ti, and LREE (similarly to what was observed in the Archean mantle xenoliths from South Africa; e.g. Burgess and Harte, 1998) and induced their recrystallization and overgrowth, along with the formation of K-rich hydrated phases. It is related to a K-rich fluid and/or liquid (proto-kimberlite?) that predates the kimberlite emplacement (~70 Ma for Mbuji-Mayi and ~35 Ma for Kundelungu). These inequigranular mantle rocks might be more sensitive to exhumation processes and are probably weakened during the kimberlite eruption, resulting in the isolation of large (>1 cm) round crystals inside the kimberlite magma or small (<1 cm) more angular fragments after the mining separation stage.

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