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Nd - Hf ISOTOPE SYSTEMATICS OF MEGACRYSTS FROM THE MBUJI-MAYI KIMBERLITES, D. R. CONGO: IMPLICATIONS FOR THE COMPOSITION OF THE CRATONIC LITHOSPHERIC MANTLE

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INTRODUCTION

While Cr-poor megacryst suites from Group 1 kimberlites worldwide are usually assigned to magmatic fractional crystallization at depth, close to the lithosphere-asthenosphere boundary, direct relationship with the kimberlite host-magma has been challenged by radiogenic isotope studies of the most common Sr, Nd and Pb isotopic systems (e.g. Weis and Demaiffe, 1985; Hops et al., 1992; Davies et al., 2001): megacryst minerals are usually characterized by a more depleted mantle source signature than their host.

Recently, the genetic relation between megacrysts and their host kimberlite has been reemphasized by combined Nd and Hf isotope studies. They have shown that Group 1 kimberlites and associated megacrysts from South African localities have negative ΔεHf values (Nowell et al., 1999; 2004) and plot below the "mantle array" (MA) defined by almost all terrestrial magmas (Vervoort et al., 1999), suggesting a decoupling of the Lu-Hf and Sm-Nd isotope systems, which is unusual during mantle melting. This characteristic has also been recognized in kimberlite megacrysts from Africa, Siberia and Australia through Hf isotope analyses of zircon megacrysts combined with isotopic composition of Nd on associated low-Cr silicate megacrysts (Griffin et al., 2000), and in kimberlite whole-rocks from the Dharwar craton, South India (Paton et al., 2009).

This typical signature, encountered in megacrysts and kimberlites of various ages, located on different continents (on- and off-craton localities), has led to several interpretations about the origin of the decoupling of the Lu-Hf and Sm-Nd isotope systems, but certainly fueled the debate regarding the potential sources of kimberlite magmas and their associated megacrysts (e.g. Pearson et al., 2008).

While Sr, Nd and Pb isotope compositions of Group 1 kimberlites and megacrysts favor a mantle source similar to that of MORB or OIB, the negative ΔεHf values are only found in some HIMU-type OIB. A common idea has been that the negative ΔεHf signature develops in old subducted oceanic crust stored for a long time (> 1Ga) in a deep layer (transition zone or core-mantle boundary) and isolated from convection, and was later entrained in a rising "kimberlite" diapir that underwent restricted interaction with the subcontinental lithospheric mantle (SCLM) (Nowell et al., 1999; 2004; Paton et al., 2009). Others considered a more extended interaction with low-degree melts from the metasomatized SCLM (Janney et al., 2002). Griffin et al. (2000) also proposed that megacryst parental melts are generated through the interaction between "normal" MORB or OIB-type asthenospheric magmas and high Hf/Nd depleted and metasomatized harzburgitic SCLM with negative ΔεHf values.

However, Group 1 kimberlites and megacrysts from the North Atlantic craton (Greenland) (Tappe et al., 2011) and some kimberlites and megacrysts from the Slave craton (Schmidberger et al., 2002; Pearson et al., 2008; Kopylova et al., 2009) fall within the mantle array. A recent model therefore proposes that there is no negative ΔεHf component in the source of these magmas and that the apparent displacement below the MA observed in some localities results from the mixing of sublithospheric and lithospheric components, both lying within the MA (coupled Lu-Hf and Sm-Nd fractionation), but having contrasted Nd/Hf ratios, i.e. carbonated silicate asthenospheric melts and old metasomes in the SCLM (Tappe et al., 2011).

The present study concerns mainly the megacrysts from the Mbuji-Mayi kimberlite province (with one megacryst from Kundelungu), in Democratic Republic of

Congo (DRC); they have not been investigated yet for coupled Hf and Nd isotope compositions. Moreover, contrary to garnet and clinopyroxene (cpx) megacrysts from most localities worldwide, those from DRC kimberlites do not show any evidence of an origin by fractional crystallization from a parental magma. They are rather interpreted as fragments of mantle peridotites having suffered a "proto-kimberlitic" metasomatic event accompanied by recrystallization in the deep cratonic lithospheric mantle (Pivin et al., 2009).

Six garnet and 6 cpx megacrysts have been chosen for Hf and Nd isotope measurements, while 3 zircon and 3 baddeleyite megacrysts have been selected for Hf isotope analyses.

All analyses were performed on a Nu Plasma multicollector ICP-MS (MC-ICP-MS) at Université Libre de Bruxelles (ULB), Belgium using the purification and analytical procedures presented in Debaille et al. (2008) for garnets and cpx, and of Goolaerts et al. (2004) for zircons and baddeleyites.

GEOLOGICAL SETTING AND SAMPLES

All the megacrysts analyzed in this study are from the Mbuji-Mayi kimberlite except one garnet megacryst that comes from the Kundelungu province.

The Cretaceous (70 Ma; Schärer et al., 1997) Mbuji-Mayi kimberlites (East Kasai) are located on the Archean Congo-Kasai craton (> 2.9 Ga). These diamond-rich Group 1 kimberlites comprise a large suite of megacrysts including garnet, cpx, zircon, baddeleyite, Mg-ilmenite, Nb- or Cr-rich rutile, corundum, kyanite, chlorite and rutile-silicate intergrowths. Eclogite nodules dominate the xenolith population (El Fadili and Demaiffe, 1999).

The Kundelungu kimberlite province is located ~ 600 km to the southeast of Mbuji-Mayi, in the Katanga region. These lower Oligocene (32 Ma; Batumike et al., 2008) Group 1 kimberlites most probably intrude the Paleoproterozoic Bangweulu block (~ 1.9 Ga) at depth. The megacryst suite notably includes garnet and cpx; xenoliths are mostly peridotites (Kampata et al., 1995).

Garnet (pyrope variety) megacrysts from Mbuji-Mayi and Kundelungu show many similarities and were subdivided in three groups based on Cr-content: low-Cr, medium-Cr and high-Cr (0.00-1.79; 1.93-5.16; 5.42-7.10 wt% Cr_2O_3, respectively). Mbuji-Mayi diopside megacrysts were classified into a low-Ca ($Ca^{\#}$: 39.5-42.6; 0.61-0.92 wt% Cr_2O_3) and a medium-Ca groups ($Ca^{\#}$: 43.9-49.5; 0.30-1.94 wt% Cr_2O_3), which are both poorer in Cr than a third group of high-Ca-Cr diopsides ($Ca^{\#}$: 47.0-50.2; 1.13-2.77 wt% Cr_2O_3) (Pivin et al., 2009). Mbuji-Mayi zircons have low trace element (e.g. REE, U) contents that are typical of zircon megacrysts in kimberlites. Baddeleyite megacrysts are usually not found in kimberlites but are common in carbonatites. Those from the Mbuji-Mayi kimberlites have however quite lower Hf contents compared to carbonatite-derived baddeleyite megacrysts (Pivin et al., unpub. data).

Sm-Nd AND Lu-Hf SYSTEMATICS

Results

Data are shown on an initial $\varepsilon Hf_{(t)}$ vs. $\varepsilon Nd_{(t)}$ plot in Figure 1. Initial $\varepsilon Hf_{(t)}$ and $\varepsilon Nd_{(t)}$ values are calculated at the kimberlite eruption age (t) of 70 Ma for Mbuji-Mayi and of 32 Ma for Kundelungu using λ^{176}Lu of Scherer et al. (2001) and λ^{147}Sm of Begemann et al. (2001). Lu-Hf and Sm-Nd CHUR values are from Bouvier et al. (2008). The $\Delta\varepsilon$Hf values discussed below is calculated relative to the "mantle array" of Vervoort et al. (1999). When literature data are used for comparison, they have been recalculated with the same parameters.

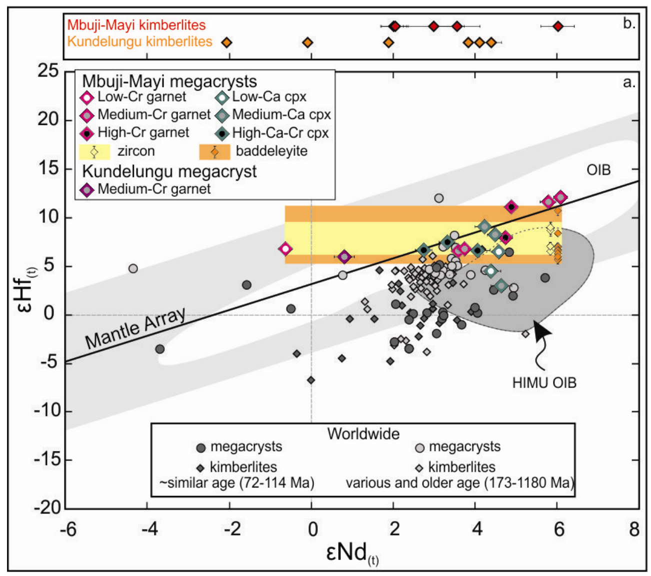

Fig.1. a. Initial $\varepsilon Nd_{(t)}$ vs. $\varepsilon Hf_{(t)}$ values of DRC megacrysts (2σ error bars range from ±0.2 to ±0.3 for $\varepsilon Nd_{(t)}$ and from ±0.4 to ±0.8 for $\varepsilon Hf_{(t)}$, wich are smaller than symbols) compared to available data for on- and off-craton Group 1 kimberlites and megacrysts worldwide for which different colors make the distinction between those of roughly similar age to DRC kimberlites (dark grey; South Africa, Lesotho, Botswana and Somerset Island) and those of older age (light grey) that extend over a wider range of age and geographic distribution (South Africa, India, Canada and Greenland) (Nowell et al., 1999; 2004; Schmidberger et al., 2002; Kopylova et al., 1999; Paton et al., 2009; Tappe et al., 2011). Present day OIB and HIMU-type OIB fields are also plotted (Ballentine et al., 1997; Salters and White, 1998). Fields for Mbuji-Mayi baddeleyite and zircon megacrysts comprise $\varepsilon Hf_{(t)}$ data from this study (2σ error bars from ±0.2 to ±0.3) and those from Schärer et al. (1997) and range over $Nd_{(t)}$ data of garnet and cpx megacrysts. b. Ranges of $\varepsilon Nd_{(t)}$ values for the 70 Ma old Mbuji-Mayi and 32 Ma old Kundelungu kimberlite whole rocks (Weis and Demaiffe, 1985; Kampata et al., 1995).

Mbuji-Mayi garnets display a range of initial $\varepsilon Hf_{(t)}$ values from +6.6 to +12.1 with corresponding initial $\varepsilon Nd_{(t)}$ values that mostly range from +3.6 to +6.1, except for one low-Cr pyrope with an unradiogenic $\varepsilon Nd_{(t)}$ value of -0.6.

Mbuji-Mayi diopside megacrysts also show a wide range of $\varepsilon Hf_{(t)}$: +3.0 - +9.1, which is quite less radiogenic than garnets. The $\varepsilon Nd_{(t)}$ range of +2.7 - +4.6 overlaps that of garnets (except for the outlier), especially if previously published $\varepsilon Nd_{(t)}$ data for two Mbuji-Mayi cpx megacrysts are accounted for (+6.5 and +7.0; Weis and Demaiffe, 1985).

There is no striking correlation between the group of pyrope or diopside megacrysts and their Nd or Hf isotope composition (Fig.1).

Three Mbuji-Mayi zircon megacrysts have radiogenic initial $\varepsilon Hf_{(t)}$ values ranging from +6.5 to +7.1 that are relatively similar to those of baddeleyite megacrysts from the same locality: +6.0 to +8.4. Schärer et al. (1997) analyzed similar megacrysts and found matching values that however extend the respective ranges to more radiogenic $\varepsilon Hf_{(t)}$ values: up to +9.0 for zircons and to +10.7 for baddeleyites.

The Kundelungu garnet megacryst has a low $\varepsilon Nd_{(t)}$ value of +0.8 and a radiogenic $\varepsilon Hf_{(t)}$ of +6.0 that is similar to those of Mbuji-Mayi garnets. Previously published $\varepsilon Nd_{(t)}$ data on one Kundelungu garnet is slightly more radiogenic (+2.5) and corresponds well to $\varepsilon Nd_{(t)}$ values of diopside megacrysts from the same locality (+2.2 - +2.5) (Kampata et al., 1995).

DRC pyrope megacrysts mostly plot within the mantle array (Fig.1), with either slightly negative or positive $\Delta\varepsilon Hf_{(t)}$ values: -1.5 to +4.4 for Mbuji-Mayi and +1.7 for Kundelungu. Most Mbuji-Mayi diopside megacrysts have slightly negative $\Delta\varepsilon Hf_{(t)}$: -6.4 - +0.3 but still plot within the quite large range that defines the mantle array ($-7 < \Delta\varepsilon Hf_{(t)} < +7$; e.g. Janney et al., 2002).

The ranges of $\varepsilon Hf_{(t)}$ data obtained on zircon and baddeleyite megacrysts from Mbuji-Mayi are comparable to those of garnet and cpx megacrysts from the same locality and would also plot in the mantle array if similar Nd isotope compositions are assumed (Fig.1).

Timing Constraints

Lu-Hf and Sm-Nd isochrons should reflect the time since closure of the respective isotopic systems if no later disturbance has occurred. As such, megacrysts are known to be potentially good indicators of the kimberlite eruption age (e.g. Nowell et al., 2004; Kopylova et al., 2009). Unfortunately, high precision Lu-Hf and Sm-Nd isotopic analyses of individual megacryst phases reported either individually or combined has not given any reliable geochronological constraint for the Mbuji-Mayi megacrysts.

Hf model ages relative to a depleted mantle source (T_{DM}) for Mbuji-Mayi clinopyroxene, zircon and baddeleyite megacrysts give quite similar ages (362-694 Ma, 355-451 Ma and 286-484 Ma, respectively) but with no apparent geological meaning. T_{DM} calculated for garnet megacrysts are much more variable (from highly negative to > 3.9 Ga) because measured $^{176}Lu/^{177}Hf$ ratios in garnets (0.0304-0.0397) are too close to that of model DM (0.0384; Griffin et al., 2000).

Depleted mantle Nd model ages of clinopyroxene megacrysts (460-837 Ma) compare relatively well to those of Hf. Mbuji-Mayi garnets show a narrow range but negative Nd model ages (-202 Ma to -85 Ma), except for the unradiogenic low-Cr megacryst that has a lower $^{147}Sm/^{144}Nd$ (0.1374) compared to the others (0.3563-0.5308), resulting in a T_{DM} of 1234 Ma.

Both Hf and Nd model ages on the Kundelungu garnet give negative values.

DISCUSSION

Comparison to Worldwide Megacrysts and Kimberlites

Before comparing DRC megacrysts to kimberlites and megacrysts from other localities worldwide on a $\varepsilon Hf_{(t)}$-$\varepsilon Nd_{(t)}$ diagram, it is important to make the distinction between isotopic data of kimberlites and megacrysts according to their age (Fig.1). In addition, while initial isotopic compositions of megacrysts are traditionally calculated at the time of kimberlite eruption, either because they are considered as phenocrysts in the host magma, or because their isotopic system should have been reset at the time of kimberlite eruption, this inference is not certain for DRC megacrysts as a metasomatic origin is favored (Pivin et al., 2009), which possibly implies an older protolith.

It is interesting to note however that most kimberlites, whenever they erupted, display a rather restricted field in this plot, with mean values of +2.6 ± 2.2 (2σ dispersion) and of +1.9 ± 6.0 for $\varepsilon Nd_{(t)}$ and $\varepsilon Hf_{(t)}$ respectively, suggesting identical source components that evolve with similar $^{147}Sm/^{144}Nd$ and $^{176}Lu/^{177}Hf$ ratios. Worldwide megacrysts show similar averages to kimberlites: $\varepsilon Nd_{(t)}$ of +3.3 ± 3.6 and $\varepsilon Hf_{(t)}$ of +1.6 ± 6.1 (references in Fig.1).

Mbuji-Mayi megacrysts have $\varepsilon Nd_{(t)}$ values that are relatively well comparable to those of DRC kimberlite host-rocks (Fig.1b) though the low-Cr pyrope data extends the range to more unradiogenic Nd composition. Identically, the Kundelungu garnet and previously published values for garnet and cpx megacrysts (Kampata et al., 1995) also overlap the Nd isotopic field of the host kimberlites. Hf isotope ratios of DRC kimberlites are not known but we may suppose that they are similar to other kimberlites worldwide, and should therefore be less radiogenic than DRC megacryst minerals, which average to $\varepsilon Hf_{(t)}$ +7.5 ± 2.9 for Mbuji-Mayi and is of +6.0 for the Kundelungu garnet.

By comparison to the worldwide megacryst database, data from the Mbuji-Mayi megacryst population significantly extend the isotopic field, especially to more radiogenic $\varepsilon Hf_{(t)}$ and $\varepsilon Nd_{(t)}$ values.

The variability in $\varepsilon Hf_{(t)}$ for Mbuji-Mayi zircon megacrysts is quite common in kimberlite zircons from a single locality and similar values have been reported for zircon megacrysts from on- and off-craton localities of various ages, like Jwaneng (young population of 240 Ma, Kaapvaal craton) and Dianga (156 Ma, off-craton, Siberia). The closest values relative to the DRC data are reported for Siberia on- and off-craton kimberlites (Griffin et al., 2000; Nowell et al., 2004).

What About The SCLM?

The proposed metasomatic origin for DRC garnet and cpx megacrysts has arisen from several observations: 1) they do not display the classical bi-element correlations observed for many other megacryst suites, 2) they have major element compositions that are intermediate between megacrysts from other localities and garnets and cpx from Archean mantle peridotites, and 3) they have rare earth element patterns similar to those of mantle minerals having undergone kimberlite-related metasomatic events (Pivin et al., 2009).

The cratonic SCLM is well-known to be highly heterogeneous in isotopic composition, because of its long lived history and various magma interactions (e.g. Simon et al., 2007). Nowadays, combined Nd-Hf isotope compositions (on whole rocks or mineral separates) have been investigated in the SCLM for only few localities.

Literature compilation brings that whole rock peridotites display a subvertical trend with highly variable and radiogenic $\varepsilon Hf_{(t)}$ (up to +52) compositions and more restricted $\varepsilon Nd_{(t)}$ values (-5.1 to +3.9) (Fig.2). This trend might reflect the more contamination-sensitive character of Nd isotopes relative to Hf; the latest reflecting time integrated long term depletion of the cratonic SCLM (e.g. Schmidberger et al., 2002). When garnet and cpx separated minerals are considered, a much wider range of isotope compositions is observable, with mostly radiogenic to highly radiogenic $\varepsilon Hf_{(t)}$ compositions (up to few hundreds for garnets; not shown on Fig.2) and mostly unradiogenic εNd values that mirror old LREE enrichment.

Surprisingly, high-T peridotite whole-rock xenoliths, which are usually inferred to represent the base of the SCLM and to have suffered extensive kimberlite-related metasomatic interaction, have more restricted isotope compositions that sometimes plot within the kimberlite field ($\varepsilon Hf_{(t)}$: -2 to +21; $\varepsilon Nd_{(t)}$: 0 to +5), as do their constitutive garnet minerals (Fig.2).

In addition, increasing evidences argue for a recent crystallization of clinopyroxene in low-T peridotite xenoliths from the cratonic SCLM, through interaction with the erupting kimberlite magma or a precursor (e.g. Simon et al., 2007); Nd-Hf isotope systematics seem to favor such an origin since some peridotitic cpx effectively plot in, or close to, the kimberlite field (Fig.2).

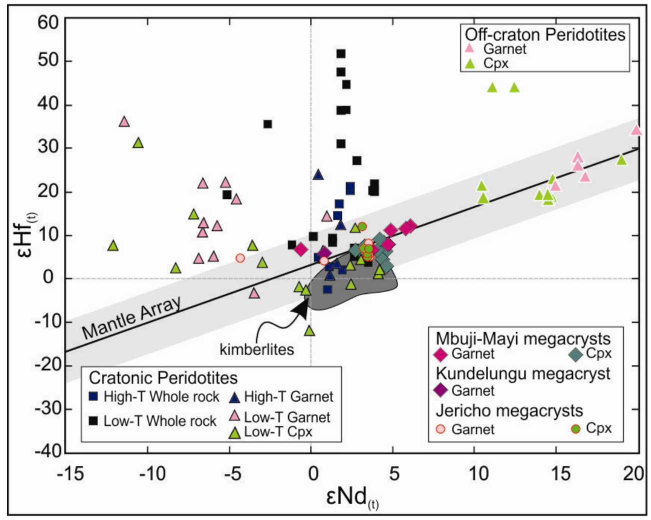

Fig.2. Initial $\varepsilon Nd_{(t)}$ vs. $\varepsilon Hf_{(t)}$ values of DRC garnet and clinopyroxene megacrysts compared to those of 1) peridotite xenoliths and constituting minerals from cratonic SCLM from the Kaapvaal and Slave cratons (Schmidberger et al., 2001; 2002; Bedini et al., 2004; Simon et al., 2007) and 2) fertile off-craton peridotite xenoliths from Vitim volcanic field, Siberia (Ionov et al., 2005). High-Cr megacrysts from the Jericho kimberlite field are also plotted for comparison (Kopylova et al., 2009).

The narrow range of Nd-Hf isotopic compositions for cpx of inferred metasomatic origin in peridotites and Mbuji-Mayi clinopyroxene megacrysts (Fig.2) might reflect similar petrogenetic processes; i.e. recent (re)crystallization in metasomatized mantle peridotites by kimberlite melts.

A protokimberlitic metasomatic origin has also been proposed for bimineralic megacrysts from the Jericho kimberlites (Slave craton; Kopylova et al., 2009) and Nd-Hf isotope compositions of the latter compare well to those from Mbuji-Mayi (Fig.2).

CONCLUDING REMARKS

Garnet, cpx (and zircon and baddeleyite) megacrysts from DRC kimberlites do not show important downward displacement relative to the mantle array in the $\varepsilon Hf_{(t)}$ vs. $\varepsilon Nd_{(t)}$ plot. They all are characterized by positive $\varepsilon Hf_{(t)}$, as observed in some other localities.

DRC megacrysts display a range of Nd isotope composition similar to their kimberlite host-rock. However, because they did not form through magmatic fractional crystallization from the kimberlite magma (Pivin et al., 2009), they most probably result from metasomatic interaction between an old SCLM, as represented by various peridotite whole-rock xenoliths, and the kimberlite magma (Fig.2).

Although depleted mantle model ages do not have any geological meaning, they nevertheless suggest a common origin for Mbuji-Mayi cpx, zircon and baddeleyite megacrysts compared to garnets. We therefore suggest that while garnet megacrysts may effectively be formed by recrystallization of a garnet-bearing peridotite protolith; clinopyroxenes (and zircons and baddeleyites) might directly crystallize from the inferred "proto-kimberlitic" magma via AFC-type reactions in the SCLM.

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