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THE ORIGIN AND EVOLUTION OF THE LITHOSPHERIC MANTLE BENEATH THE MAKONDI FOLD BELT IN BOTSWANA; A COMPREHENSIVE GEOCHEMICAL STUDY OF PERIDOTITE XENOLITHS FROM THE LETLHAKANE DIAMOND MINE

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INTRODUCTION

The Letlhakane kimberlite pipes were emplaced in the early Proterozoic Makondi Fold Belt, Botswana. They belong to a cluster of approximately 82 kimberlite diatremes that also includes the Orapa diamond mine, approximately 40 km to the northwest. The Makondi Belt is a SE-vergent thrust and fold belt consisting of volcanics, orthoquartzites and carbonate sequences (Stiefenhofer et al., 1994) considered to be the passive margin of the Zimbabwe Craton (Griffin et al., 2003). Previous geochemical analyses on small chips of xenoliths (<5cm) from Letlhakane led Stiefenhofer et al. (1994) to conclude that the Makondi Fold Belt is underlain by Archean mantle belonging to the Zimbabwe Craton. This conclusion is supported by a magnetotellurics study, showing that the Letlhakane kimberlite is located at the Zimbabwe Craton- Makondi Belt contact (Jones et al., 2009). This implies that the kimberlites were sourced from the edge of Zimbabwean cratonic mantle and ascended through Proterozoic Makondi crust, explaining their rather anomalous tectonic setting. Similar to the Kaapvaal Craton and the Limpopo Mobile Belt, the Zimbabwe sub-cratonic lithospheric mantle (SCLM) is characterized by high amounts of melt depletion (>30%) and

subsequent silica re-enrichment. The latter process, reflected by the re-introduction of pyroxenes, is however reported to be less pervasive than in the Kaapvaal sample suite (Smith et al., 2009).

The Letlhakane kimberlites carry xenoliths of Karoo age and are overlain by a 4 to 10 m thick cover of Kalahari sand and calcrete (Stiefenhofer et al., 1994). The timing of the kimberlite emplacement has not been determined, but is inferred to be post-Karoo and pre-Kalahari. They are probably of the same age as the Orapa kimberlite (93 Ma; Davis, 1977). Therefore, Letlhakane mantle rocks are likely affected by the Karoo and Bushveld magmatic provinces. Extensive modal and cryptic metasomatism of Letlhakane xenoliths has been reported by Van Achteberg et al (2001), expressed by the presence of phlogopite, clinopyroxene and Cr-spinel, which grow at the expense of garnet and orthopyroxene with increasing degrees of metasomatism.

METHODS

In this study we report geochemical analyses on large peridotitic xenoliths in order to characterize the Zimbabwe SCLM. We aim to constrain the spatial variations in melt depletion as well as metasomatic processes by comparing



our data with geochemical data from peridotite xenoliths from the Orapa and Venetia mines. 132 peridotite xenoliths with diameters over 2 cm were identified from the D/K1 pit and associated drill cores. 37 fresh samples were selected for detailed study based on the degree of weathering and a preferred absence of major metasomatic features, such as large amounts of phlogopite. Thick sections were prepared for in-situ mineral major and trace element analyses with an electron microprobe (EMP) and laser ablation ICP-MS. The largest samples (13) were analysed for whole rock major and trace element compositions using XRF. The selected specimens range in size from 4 to 30 cm in diameter and consist of garnet lherzolites (23), garnet harzburgites (4), spinel lherzolites (6), spinel harzburgites (1), a wherlite, a garnet orthopyroxenite and a dunite. Approximately 40% of the selected samples contain small amounts (<2%) of phlogopite, often related to the breakdown of garnet. Clinopyroxene is present in ~76 % of the samples. Four samples contain a dark green Cr-rich (2.1%) amphibole phase, interpreted as pargasite. A similar phase is also reported in peridotite samples from Orapa (Meulemans et al., 2012, this issue).

PETROGRAPHY

Samples define four groups based on mineral assemblages and compositions. All garnet-free samples make up group I, including harzburgites, lherzolites, a dunite and a wherlite. The remaining samples were classified into three groups based on garnet major element composition. Samples with a harzburgitic garnet composition or harzburgitic mineral assemblage are placed in group II. Some group II samples, however contain trace amounts of (poikilitic) cpx as well as phlogopite. Lherzolitic samples make up group III and amphibole-bearing lherzolites group IV. The presence of pargasitic amphibole considerably affects the garnet major element composition that become enriched in Al_2O_3 and

FeO and depleted in Cr_2O_3 and MgO. Amphibole also affects the REE content of garnet and clinopyroxene, see below. Some clinopyroxenes in group IV show exsolution of spinel, possibly indicating low pressure re-equilibration. Textures are mostly equant coarse, with the exception of some porphyroblastic samples and one sheared sample (AT1405).

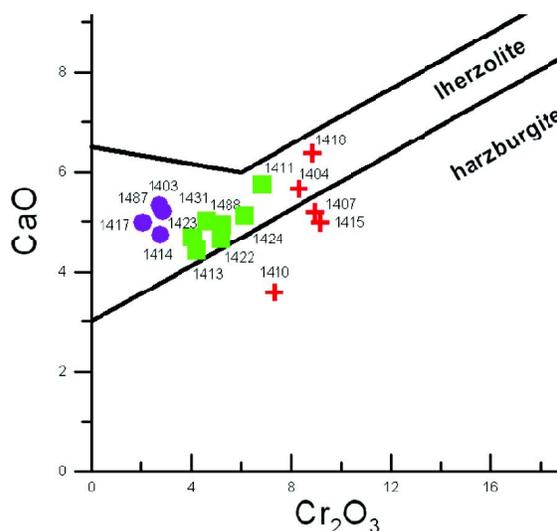


Fig. 1 Cr_2O_3 vs. CaO composition of analyzed garnets. Red crosses: group II, green squares: group III, purple circles: group IV. Lherzolite field is equivalent to G9 garnets and the harzburgite field to G10 (Grutter et al., 2004).

DISCUSSION

Whole rock major and trace elements indicate that most samples are highly melt depleted, being enriched in compatible elements and depleted in magmaphile elements relative to fertile mantle. Re-enrichment of the most incompatible elements such as K_2O , NaO and TiO_2 seems to have occurred, most likely due to contamination from small amounts of kimberlitic melt. Magnesium numbers of 90.4 to 93.5 (average 92.4) for the majority of olivines indicate ~20 to 50% of melt extraction (Pearson and Wittig, 2008). These data are comparable with data reported by Stiefenhofer et al. (1994) and Van Achteberg et al. (2001). Such high degrees of



melt depletion should have exhausted pyroxenes and garnet, leaving residual lithologies of garnet-free dunites and harzburgites (Bernstein et al., 2007). The fact that most samples have a much more fertile assemblage suggests that the Zimbabwe sub-cratonic lithosphere has been subjected to significant re-enrichment after initial depletion.

Calculated whole rock major element compositions compared with melt depletion models from Walter et al., 2004 also suggest 20 to 50% melt extraction between ~3 and 5 GPa. Similarly, we can infer melting in the garnet stability field followed by garnet absent melting from HREE patterns of reconstructed whole rock trace elements. These whole rock HREE patterns compared with melting models from Simon et al. (2007) indicate approximately 20% melting in the garnet stability field and subsequently 5 to 20% melting in the absence of garnet (Fig. 2). It should be noted that these are minimum melting estimates due to possible metasomatic re-enrichment in HREE. The garnet-bearing samples show a progressive melt depletion trend, with the most extensive melting seen in group II samples. This is consistent with both the garnet (high Cr) and olivine major element compositions (high Mg#’s) of group II samples.

Pressure and temperature calculations from co-existing garnet, opx and olivine range between 2.6 GPa at 752 °C and 6.9 GPa at 1355 °C using the barometer and thermometer pair of Brey and Kohler, 1990 ($P_{bkn} - T_{bkn}$). Calculations using P_{bkn} combined with other thermometers (T_{Harley} ; Harley, 1984 and $T_{ca-in-opx}$; Brey and Kohler, 1990) give a narrower range of equilibration pressures (2.9 GPa to 6.3 GPa; Fig. 2). Calculated P/T values fit best with a geotherm of 40 mW/m² (Pollack & Chapman, 1977).

The modal orthopyroxene contents vary between 0 and 40%, with one exception of 90% (AT1411, garnet websterite). The average opx content is 17%, which is significantly less than the average Kaapvaal orthopyroxene content

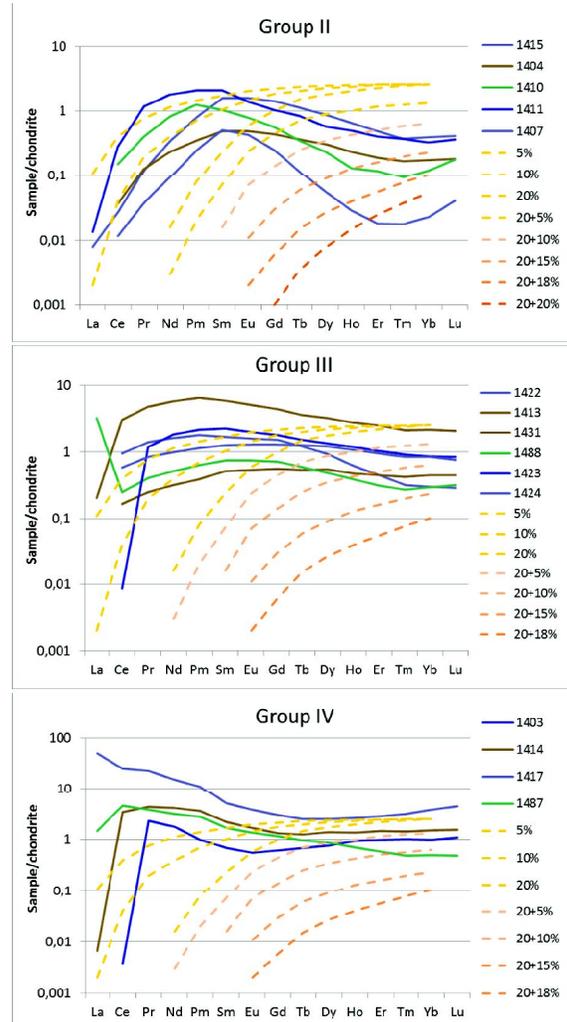


Fig. 2 Reconstructed whole rock trace element patterns for group II, III and IV samples compared with melting models from Simon et al. (2007). The first three (yellow) melting lines are in the garnet stability field, the orange melting lines indicate initial melting in the garnet field with additional melting in the spinel field. HREE are least affected by metasomatic events and therefore best for melt extraction estimates

(~30%, Simon et al., 2007). Magnesium numbers in olivine range between 90.4 and 93.5, with one outlier of 87.9 from the wherlitic sample. These values compared with modal olivine contents suggest significant SiO₂ re-enrichment, to a similar extent as the Kaapvaal xenoliths (Fig. 4). These results are in contrast with the findings of

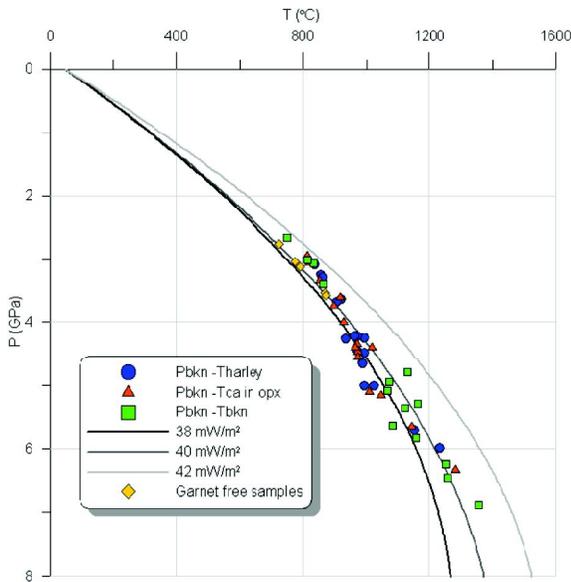


Fig. 3 Pressure temperature calculations with three different thermometer pairs. Geotherms of 38, 40 and 42 mW/m² plotted for comparison. Garnet free samples are calculated with a preset pressure of 2 GPa and projected onto the 40 mW/m² geotherm.

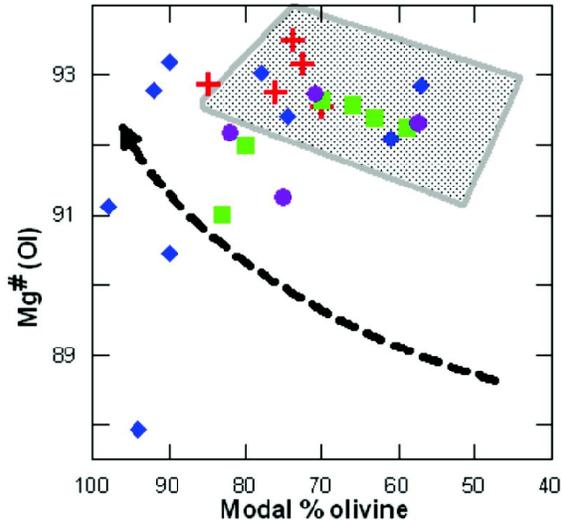


Fig. 4 Olivine Mg# versus modal olivine content for Letlhakane samples. Blue diamonds: group I. For other colour symbols see Fig. 1. Black arrow is oceanic melting trend; grey field represents typical Kaapvaal xenoliths (Boyd, 1989). SiO₂ enrichment reduces modal olivine, i.e. points move to the right of the oceanic melting trend.

Smith et al (2009), based on samples from the Murowa and Sese kimberlites in Zimbabwe, which did not show such pronounced silica re-

enrichment. Apart from the addition of silica and associated re-introduction of pyroxenes, many samples contain >5% garnet, which also requires the addition of Al₂O₃ and CaO. One sample (AT1413) with 20% garnet and 15% clinopyroxene is particularly enriched in these elements: 4.52% Al₂O₃ and 3.36% CaO.

LA-ICPMS trace element analyses on 20 samples reveal great variety in garnet REE patterns. Garnet [La]_N vary between 0.07 and 1.6 and [Lu]_N between 0.85 and 34.9. We can distinguish three different patterns for garnets: sinusoidal, flat HREE and rising HREE. All garnets are LREE depleted. The shape of the REE patterns largely correlate to the earlier grouping based on major element compositions. The rising HREE patterns coincide with group IV (amphibole bearing) samples. Flat HREE patterns are all from group III (lherzolitic) garnets. Sinusoidal patterns are measured in garnets from both group II (harzburgites) and III. The sinusoidal patterns are variable with maxima from Nd to Gd.

By comparing trace element patterns of co-existing minerals, equilibrium conditions in the rock can be inferred. Equilibrium conditions between minerals may indicate if phases originate from late stage changes due to transportation or fluid infiltration. As the major phase in most samples, all mantle-derived minerals should have reached equilibrium with olivine. In the absence of trace element data for olivine (below detection limits), magnesium numbers of different phases can also be used, based on the partitioning of Mg and Fe. In equilibrium Mg#’s of cpx and opx should be similar or slightly greater than that of co-existing olivine (Pearson et al., 2003). Letlhakane opx have Mg#’s between 0.7 and 1 higher than co-existing olivines, suggesting that olivine and opx have reached equilibrium in all samples. Magnesium numbers of clinopyroxene, however, vary from +2.8 to -5 compared to co-existing olivine Mg#’s. Clinopyroxenes in group IV have generally higher Mg#’s than olivine (+2.8 to +1.7), while Mg#’s of clinopyroxenes in group



II (harzburgites) range from -4.7 to -1.7 with olivine.

These group II clinopyroxenes make up less than 1% of the rock and all appear to have exsolved from garnet. The low Mg#’s suggest that exsolution occurred in a late stage such that equilibrium has not been attained. In group III (lherzolites) most cpx Mg#’s are within 0.3 of olivine and thus seem to be in equilibrium. REE patterns of group III samples also suggest equilibrium conditions between garnet, cpx and opx. Again, sample AT1413 of group III is an outlier with a Mg# for cpx that differs from olivine by -5. Major elements thus suggest disequilibrium between cpx and olivine, while trace element patterns of AT1413 suggest cpx is in equilibrium with garnet and opx. This discrepancy may be explained by a late-stage addition of phlogopite, which requires Mg and K and consequently changes the major element composition of clinopyroxene, bringing it out of equilibrium with the other phases. Since phlogopite is not a reservoir for REE, the trace elements still show equilibrium while the major elements do not.

Equilibrium conditions in group IV are complicated by the presence of pargasitic amphibole. Amphiboles measured in two samples (AT1417 and AT1487) are more enriched in REE than co-existing clinopyroxenes (x 1.3). In both samples amphibole seems to be in equilibrium with clinopyroxene with respect to LREE and MREE. However, a considerable spread in HREE in both amphibole and cpx suggest that full equilibrium has not yet been achieved in HREE. This indicates late metasomatic introduction of amphibole.

The amphiboles have a Y/Zr ratio of 0.14 compared to clinopyroxene with a Y/Zr ratio of 0.11. The much higher observed concentrations of Y and Zr makes the amphibole a large reservoir for these elements. Garnet in samples with pargasitic amphibole (Group IV) have higher Y/Zr ratios (Fig. 5). The changes in garnet chemistry are likely due to partitioning between these two

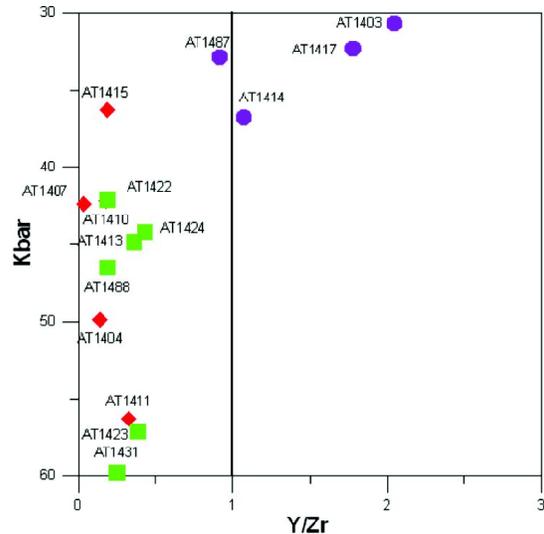


Fig. 5 Y/Zr ratio of garnets vs. depth, showing a large difference between those from amphibole-bearing group IV and other samples.

phases. Using Y/Zr ratios in garnet as a proxy for amphibole, the already large amounts of garnet concentrate data can be used to infer the existence of shallow metasomatic amphibole and additionally, shed light on why some xenoliths suites have shallow excursions of garnet Y/Zr ratios while others do not.

Gaul et al. (2000) argued that Fe percolated from the asthenosphere changed the composition of olivine near the base of the lithosphere. This conclusion was based on garnet concentrate data to calculate the depth and co-existing olivine chemistry. Our sample suite shows good correlation with the Gaul et al data (Fig. 6), but is assessed using a more laborious but straightforward approach, giving credence to their approach. Gaul et al. (2000) also note that TiO₂ concentrations in garnet increase with depth. This is generally true for our sample suite. However, at a given depth lherzolitic samples (more metasomatised) have higher garnet TiO₂ concentrations than garnet harzburgites at similar depth.

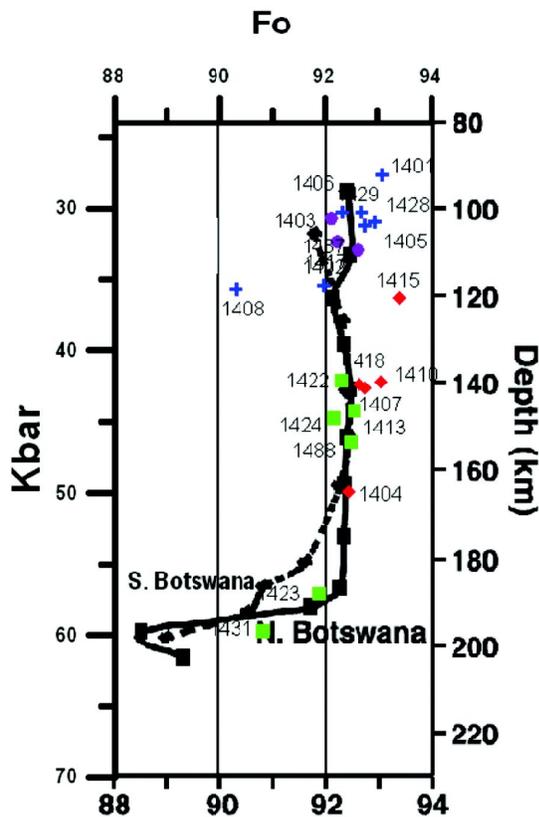


Fig. 6 Graph of depth vs. Fo content in olivine. The black lines are from Griffin et al. (2003) and show calculated averages of olivine Fo compositions for depth intervals. Other data points are from olivine Fo compositions measured by EMP. Griffin et al. (2003) depths are calculated using the garnet Ni thermometer (Ryan et al., 1996).

CONCLUSIONS

The lithospheric mantle beneath the Letlhakane kimberlite mine is characterized by high degrees of partial melting, consistent with Archean craton genesis models. The residual lithologies were subsequently enriched in Si, with a component of Al and Ca, leading to the re-introduction of pyroxenes and garnet. Sufficient amount of time has passed for all major phases to have reached equilibrium in the mantle. Minor clinopyroxenes in group II samples are likely due to late decompression exsolution. Phlogopite and amphibole metasomatism occurred relatively late,

leading to a change in clinopyroxene and garnet major and trace element chemistry. The introductions of clinopyroxene, phlogopite and amphibole at different stages indicate the Zimbabwe SCLM underwent a complex history of metasomatic events.

ONGOING WORK

Sm-Nd and Lu-Hf isotope analysis on garnet, clinopyroxene and amphibole are underway to provide age constraints on the timing of melting and subsequent metasomatic events. Additionally, an attempt is being made to determine the Letlhakane eruption age using Rb-Sr dating of primary phlogopites from the kimberlite.

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