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Diamonds reveal subducted slab harzburgite in the lower mantle

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ABSTRACT

Characterizing compositional heterogeneity in Earth's lower mantle is critical to understanding its dynamics. Three low-nitrogen diamonds from Koffiefontein (South Africa), containing inclusion assemblages of ferropericlase ± orthopyroxene ± magnesite, constrain diamond formation in an Mg-rich lower-mantle environment. Ferropericlase inclusions have Mg# 82.7–88.5 and orthopyroxene inclusions (retrogressed bridgmanite) have Mg# 95.0–95.1 and mantle-like δ^{18}O of +5.6‰ ± 0.2‰. Magnesite included in one diamond implicates carbonated fluids in diamond formation. High Mg# and low Ca, Al, and Na of the assemblage indicate a melt-depleted meta-harzburgitic environment, in contrast to more fertile compositions expected for primitive lower mantle. Extremely low Ca in orthopyroxene inclusions may reflect a combination of melt depletion and low equilibration temperatures at the time of trapping. Inclusion compositions implicate subducted oceanic slab meta-harzburgite as the host for diamond growth. Mantle-like δ^{18}O of the orthopyroxene inclusions indicates unaltered oceanic lithosphere. Similar melt-depleted characteristics in lower-mantle inclusion assemblages worldwide support that meta-harzburgite is the dominant host of lower-mantle diamonds.

INTRODUCTION

Seismic observations of lower-mantle heterogeneities (Dziewonski et al., 1977) and tomography of subducting slabs penetrating the lower mantle (van der Hilst et al., 1997) show that not all of this vast reservoir is composed of primitive, undifferentiated mantle. Sublithospheric diamonds proposed to originate from lower-mantle depths (e.g., Harte et al., 1999) sample material that in some way is related to the diamond-forming event. The question of whether the peridotitic mineral associations sampled by lower-mantle diamonds represent primitive mantle or lithologies subducted to these depths in slabs has, however, not been resolved. Our study addresses this question.

Studies of sublithospheric diamonds indicate an intimate relationship with the subduction cycle, making them excellent probes of deep mantle compositional variability (Stachel et al., 2000; Shirey et al., 2021). Calcium-rich silicate and majorite garnet inclusions in sublithospheric diamonds provide evidence for carbonated melts derived from deeply subducted basaltic oceanic crust (Stachel et al., 2000; Walter et al., 2008). Metallic inclusions in sublithospheric diamonds have heavy Fe isotope compositions consistent with deep subduction of meta-serpentinites (Smith et al., 2021). The source of lower-mantle diamonds carrying Mg-rich silicate and oxide inclusions is enigmatic. Kaminsky (2017) suggested that these assemblages represent primitive mantle based on their Mg#, while Kesson and Fitz Gerald (1991) argued for derivation from depleted mantle based on reconstructed bulk-rock chemistry.

We report on three nitrogen-poor diamonds from the Koffiefontein mine (South Africa) containing inclusions of ferropericlase (fper) ± orthopyroxene (opx) ± magnesite, representing a phase assemblage that can only coexist in the lower mantle where the opx inclusions originally were bridgmanite (brd). Bridgmanite trapped as a single-phase inclusion in diamond in the lower mantle undergoes a retrograde phase change to opx upon decompression; all previously reported bridgmanite inclusions occur as opx (Walter et al., 2022).

The presence of both silicate and oxide inclusions within the same diamond allows assessment of their parent lithology. We used the compositions of our new lower-mantle inclusion assemblage together with lower-mantle inclusion compositions from global localities (Walter et al., 2022) to place key constraints on the question of whether they derive from primitive or recycled compositions.

METHODS

We recovered three diamonds containing fper only, fper + opx, and fper + opx + magnesite from a suite of ~200 diamonds from Koffiefontein mine (29°25′07″S 24°59′36″E). Raman spectroscopy and electron probe microanalysis enabled structural and chemical characterization of the liberated inclusions. Oxygen-isotope compositions (^{18}O/^{16}O) of opx inclusions and carbon-isotope compositions (^{13}C/^{12}C) and nitrogen contents of diamond hosts were conducted via ion microprobe. Diamond photographs and descriptions, analytical methods, and full results are provided in the Supplemental Material[1].

RESULTS AND DISCUSSION

Major Element Evidence for an Origin in a Depleted Lower-Mantle Reservoir

Twenty-five (25) non-touching inclusions (Table S1 in the Supplemental Material) were released from the three Koffiefontein diamonds including 17 fper, 4 opx, 1 magnesite, 1 magnesite rim around fper, and 2 epigenetic Mg-Fe silicates. Two diamonds had co-occurring fper and opx, and one only fper.

Ferropericlase occurring alone does not establish a lower-mantle origin because it can be stable throughout the upper mantle, possibly in reactions related to diamond formation (Brey et al., 2004; Thomson et al., 2016). However, when fper co-occurs with opx, this defines an inclusion assemblage that is stable only at lower-mantle conditions (Liu, 1976). Fper inclusions

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[1] Supplemental Material. Supplemental sample descriptions, full methods, and results. Please visit https://doi.org/10.1130/GEOL.S.21743381 to access the supplemental material, and contact editing@geosociety.org with any questions.

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in diamond #21/01 are chemically similar to those in diamonds 23/01 and 25/01 that co-occur with opx, which together with the very low N contents of these diamonds (5–46 atomic ppm [atppm]; Table S2) and intense plastic deformation strongly indicate a lower-mantle origin.

Orthopyroxene (opx) inclusions are interpreted as retrogressed bridgmanite based on their low Ni content, a presumed consequence of preferential partitioning of Ni into coexisting fper (Stachel et al., 2000). The NiO contents of the Koffiefontein opx inclusions range from ≤0.01 wt% to 0.03 wt% (Fig. 1A; Table S3), values at the low end of sublithospheric opx inclusions globally (Walter et al., 2022) and much lower than in opx inclusions from cratonic lithosphere (0.13 ± 0.03 wt%, 2σ) (Stachel and Harris, 2008). This observation suggests that Koffiefontein opx inclusions were originally bridgmanite equilibrated with fper. We note, however, that the partition coefficient (D) for Ni calculated for our co-occurring fper and opx inclusions ($D^{\mathrm{Ni}}_{\mathrm{fper/brd}}$ of ~50 for diamond 23/01 and 130–170 for diamond 25/01) is much higher than observed in experiments ($D^{\mathrm{Ni}}_{\mathrm{fper/brd}}$ of ~6; Irifune et al., 2010; Ishii et al., 2011); that is, the NiO contents of the Koffiefontein and most other sublithospheric opx inclusions are unusually low (Walter et al., 2022).

Koffiefontein opx inclusions are much higher in Mg# and lower in Al_2O_3 and CaO (Fig. 1) than bridgmanite coexisting with fper or fper + Ca-silicate perovskite (Ca-pvk) in primitive mantle based on experiments at ~25–47 GPa (Irifune, 1994; Wood, 2000; Hirose, 2002; Irifune et al., 2010; Ishii et al., 2011, 2018). At ~23–25 GPa, experiments produce assemblages where bridgmanite also coexists with majorite garnet ± ringwoodite. When majorite garnet coexists with bridgmanite, Al_2O_3 contents as low as ~1.2 wt% are observed (Wood, 2000; Ishii et al., 2011).

However, no experiments on primitive mantle have produced bridgmanite compositions matching those of the Koffiefontein opx inclusions, or most other inclusions in the global data set (Fig. 1). Neither majorite garnet nor ringwoodite inclusions have been reported to co-occur with opx in sublithospheric diamonds (Walter et al., 2022), and of more than 200 reported sublithospheric garnet inclusions, only one co-occurs with an iron-rich fper. Further, all majorite garnet inclusions that yield pressures >~15 GPa are low-Cr "eclogitic" rather than peridotitic garnets, and only one yields a pressure >20 GPa (Walter et al., 2022). Thus, we conclude that the low Ni-Al-Ca composition of the Koffiefontein and other sublithospheric opx inclusions is not caused by equilibration with majorite garnet or other transition-zone phases near the 660 km discontinuity.

Alternatively, the opx inclusions may have been former akimotoite (Walter et al., 2022), which is stable over a limited pressure-temperature range at the base of the transition zone in primitive mantle (Ishii et al., 2011). Experimental akimotoites coexisting with majorite garnet ± ringwoodite ± Ca-pvk have high Mg# and low Al and Ca (Figs. 1B and 1C) like Koffiefontein opx inclusions. However, akimotoite is not stable with fper in these experiments and there is no evidence for co-occurring majorite garnet in any sublithospheric diamond hosting opx and fper.

If the low-Al and low-Ca Koffiefontein opx inclusions and most others in the global bridgmanite inclusion data set are not due to equilibration in primitive mantle, then a bulk compositional control is suggested. Although there are few experiments on depleted mantle compositions at lower-mantle conditions, the Al contents of the Koffiefontein opx inclusions are akin to those of experimental bridgmanites produced in meta-harzburgite at ~23 GPa (Ishii et al., 2019). In this case, bridgmanite would be the primary host of Al, and due to its high solubility in this phase (25 mol%; Andrault, 2003), Al content cannot constrain depth of origin. There are too few experiments to gauge whether the low Ca can also be accounted for by a depleted composition in the lower mantle, but harzburgite derived through melt extraction in the shallow upper mantle is highly depleted in both Al and Ca (Godard et al., 2008).

Very low Ca in opx may also reflect low temperatures of equilibration due to the widening of the silicate perovskite solvus with decreasing temperature if Ca-pvk is part of the coexisting assemblage (Irifune et al., 2000). The very low CaO contents (0.04–0.06 wt%) of the Koffiefontein opx inclusions and most others in the global data set are indicative of temperatures of <1200 °C, consistent with a low-temperature slab geotherm in the lower mantle when the inclusions were trapped (Shirey et al., 2021; Walter et al., 2022). In the absence of experimental data at low temperatures, we speculate that the high $D^{\mathrm{Ni}}_{\mathrm{fper/brd}}$ and low Ni contents in former bridgmanite inclusions from Koffiefontein and worldwide may also relate to formation in a cool slab. Indeed, the calculated bulk rock NiO contents of ~0.27 wt% based on our Koffiefontein inclusions and a modal composition of 80% bridgmanite and 20% fper in meta-harzburgite (Ishii et al., 2019) fall within the typical range of abyssal harzburgites (Godard et al., 2008).

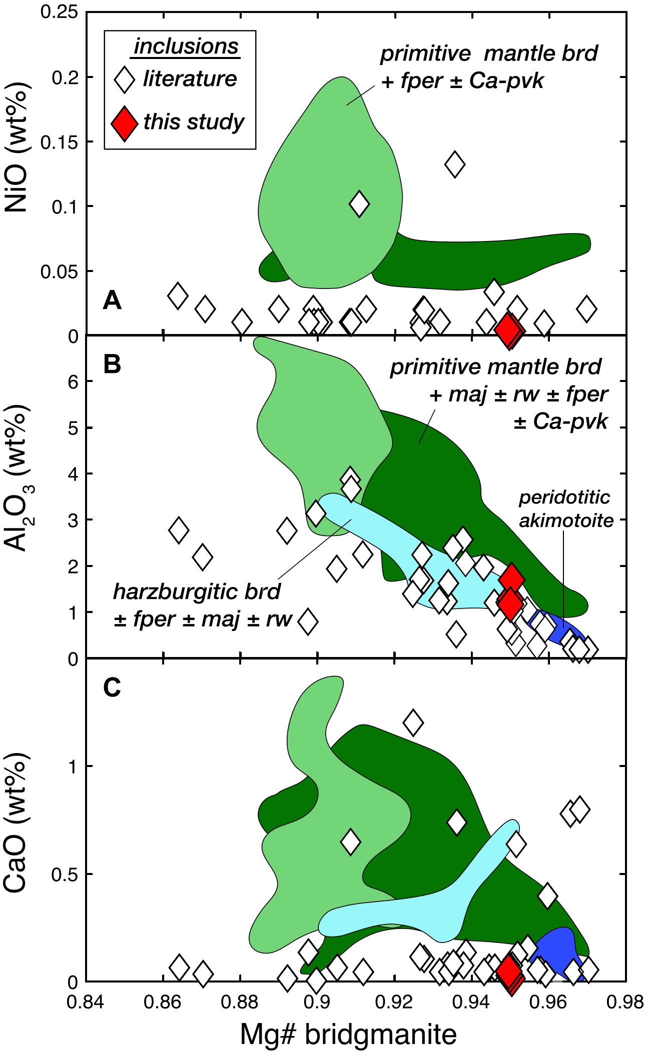

Figure 1. Mg# versus NiO (A), Al_2O_3 (B), and CaO (C) of former bridgmanite (brd) inclusions from Koffiefontein mine, South Africa (red diamonds), and lower-mantle diamonds globally (white diamonds; Walter et al., 2022). The field of experimental bridgmanite in primitive mantle coexisting with ferropericlase (fper) and Ca-silicate perovskite (Ca-pvk) (~25–47 GPa) is shown in light green, and coexisting with majoritic garnet (maj) ± fper ± Ca-pvk ± ringwoodite (rw) (~23–25 GPa) in dark green. Cyan field is experimental bridgmanite in meta-harzburgite. Dark blue field is experimental akimotoite coexisting with maj ± Ca-pvk ± rw (see Walter et al., 2022, for references).

Melt-Depleted Lithospheric Peridotite in Earth's Lower Mantle

Further insights into the role of depleted lithologies in the origin of sublithospheric opx inclusions can be made by projecting opx and co-occurring fper inclusions from Koffiefontein and global data sets (Walter et al., 2022) into the system Mg-Fe-Si (Fig. 2) together with bridgmanites produced in experiments that coexist with fper ± Ca-pvk. The Mg# of most sublithospheric opx inclusions is distinctly higher than in experiments on primitive mantle but is consistent with the higher Mg# produced in meta-harzburgite (Ishii et al., 2019). The Mg# of co-occurring fper shows a wide range that overlaps most of the experimental range. Koffiefontein and other sublithospheric opx inclusions define a unique field relative to experimental bridgmanite-fper pairs (Fig. 2 inset), with apparent regional differences among the global population. The stronger fractionation of Mg-Fe between fper and bridgmanite compared to experiments conducted at temperatures expected at the top of the lower mantle (∼1700 °C) may possibly relate to formation within a cool slab (<1200 °C), as discussed above. The low Al in bridgmanite inclusions (Fig. 1B) strongly limits the Fe^{3+} they can accommodate (Walter et al., 2022), further increasing their Mg# and the Mg-Fe fractionation between fper and bridgmanite.

Tie lines in Mg-Fe-Si space connect co-occurring opx and fper inclusions from Koffiefontein and the global data set (Fig. 2). In the absence of other co-occurring phases, these tie lines should intersect the projected system bulk composition. We note that some of the diamonds in the global data set also contain former Ca-pvk inclusions, but these inclusions have exceptionally low Mg and Fe (typically <0.2 wt%) and do not significantly bias this projection.

Tie lines between fper and opx inclusions from Koffiefontein pass through neither pyrolite nor moderately melt-depleted compositions such as average abyssal peridotite (Fig. 2) but instead are consistent with Mg-rich, strongly melt-depleted bulk compositions such as harzburgite and dunite. This observation holds for the entire global data set where nearly every tie line is consistent with a moderately to strongly melt-depleted bulk composition. Such depleted compositions are also consistent with the low Na and Al contents of high-Mg# fper inclusions relative to fper produced in experiments on fertile compositions (Walter et al., 2022).

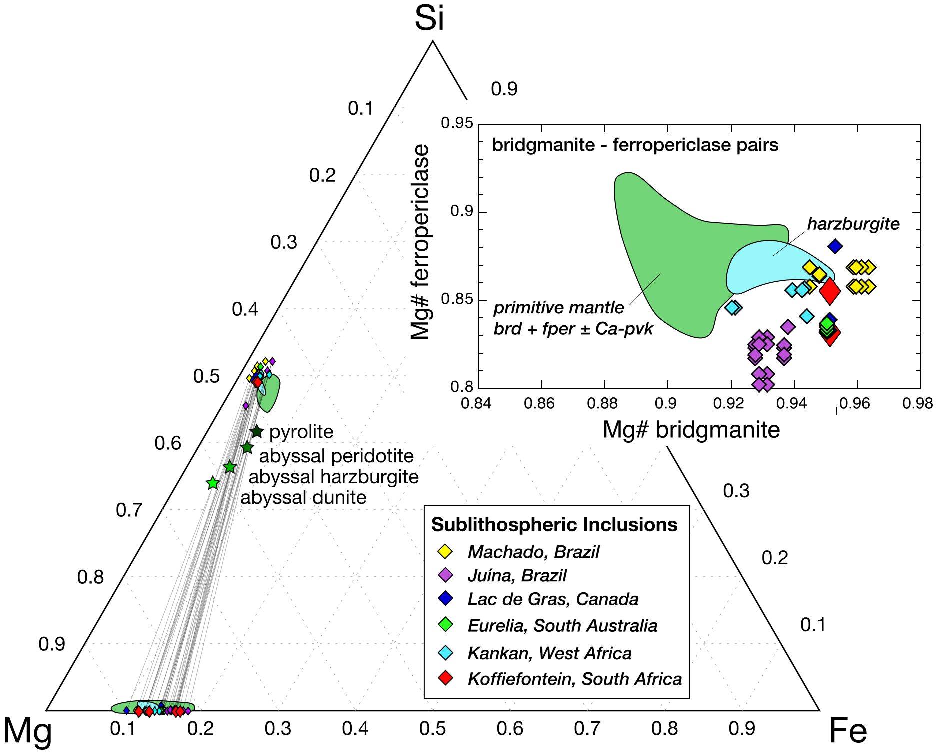

Figure 2. Composition of orthopyroxene (opx) (retrogressed bridgmanite [brd]) and ferropericlase (fper) inclusions co-occurring in sublithospheric diamonds projected onto the Mg-Fe-Si ternary diagram (molar). Inclusions are shown as diamonds and color coded by locality. Compilation of sublithospheric inclusions in diamonds from global localities is from Walter et al. (2022). Inclusions from the same diamond are connected by tie lines. Fields for coexisting brd + fper ± Ca-silicate perovskite (Ca-pvk) in experiments (see Walter et al., 2022, for references) are shown for primitive mantle (green) and meta-harzburgite (cyan). Stars are model mantle bulk compositions: pyrolite (McDonough and Sun, 1995), average abyssal peridotite (Niu, 2004), and abyssal harzburgite and dunite (Godard et al., 2008). Inset shows Mg# of co-occurring opx (brd) and fper inclusions relative to those produced in experiments.

δ^{18}O in Former Bridgmanite and the Role of Carbonated Fluids

Oxygen isotopes trace surficial recycling into the mantle. δ^{18}O values of mantle peridotite unaffected by surface processes are +5.5‰ ± 0.2‰ (Mattey et al., 1994). Metabasaltic inclusions in sublithospheric diamonds preserve the δ^{18}O signature of altered oceanic crust with δ^{18}O ranging from +7.5‰ to +12.9 ‰ (e.g., Burnham et al., 2015).

The four opx inclusions from Koffiefontein have an average δ^{18}O value of +5.6‰ ± 0.2‰ (Table S4), indistinguishable from olivine and opx from lithospheric mantle peridotites and from retrogressed bridgmanite inclusions from Kankan in Guinea (Regier et al., 2018, 2020). Peridotites in oceanic lithosphere that experienced hydrothermal alteration display a wider range in δ^{18}O from about +1‰ to +8‰ (Regier et al., 2018), but shifts from the mantle value decrease with depth in oceanic lithosphere (Eiler, 2001). The mantle-like δ^{18}O range of retrogressed bridgmanite inclusions implies an origin in a mantle environment unaffected by hydrothermal alteration processes near Earth's surface, such as the deeper parts of oceanic lithosphere.

The carbon isotope compositions (δ^{13}C range of −6.3‰ to −3.4‰; Table S2) of the three Koffiefontein diamonds fall within the δ^{13}C mantle array of −5‰ ± 2‰ (Cartigny et al., 2014). The presence of magnesite in diamond 25/01 suggests diamond formation from a carbonate-bearing melt or fluid, such as implied for Ca-rich inclusions in transition-zone diamonds (Thomson et al., 2016). The mantle-like δ^{13}C and δ^{18}O of the Koffiefontein diamonds and inclusions do not suggest involvement of recycled crust but instead point to diamond-forming fluids derived from a carbonated peridotite source. Thermal models indicate that in cold subducting slabs, water is retained in hydrous peridotite and transported into the lower mantle in dense hydrous Mg-silicates (Iwamori, 2004; Shirey et al., 2021). Breakdown of these minerals during slab stagnation and heating near the 660 km discontinuity would cause the generation of hydrous, carbon-bearing supercritical fluids (Shirey et al., 2021). Pristine (carbonate-free, anhydrous) residual harzburgite becomes increasingly reducing during the phase changes associated with subduction into the lower mantle to form an assemblage of bridgmanite plus fper (Ishii et al., 2019; Frost and McCammon, 2008).

In oceanic lithospheric mantle unaffected by hydrothermal alteration, carbon enrichment occurs in the form of carbonate veins derived from magmatic CO_2 that consequently carry a mantle-like δ^{13}C signature (Eickmann et al., 2009). Mobilization of original vein carbonate during passage through the 660 km discontinuity and associated dehydration reactions create carbonated fluids that, upon infiltration into unaltered, reduced harzburgite, crystallize diamond on reduction. Preservation of mantle-like $\delta^{18}O$ values occurs as a consequence of low fluid-rock ratios, aided by buffering of the O isotope composition of the migrating fluid to peridotite values.

CONCLUSIONS

Coexisting lower-mantle inclusions of retrogressed bridgmanite and fper in sublithospheric Koffiefontein diamonds and the global database indicate derivation from parent lithologies chemically akin to harzburgite. This, together with the low Ca content of bridgmanite, indicate an origin within a thermally unequilibrated subducting slab in the lower mantle, possibly at temperatures <1200 °C. Further indications for an origin in cold slabs come from the stronger fractionations of Mg-Fe and Ni between bridgmanite and fper compared to experiments at ambient lower-mantle temperatures. Similar "depleted" compositions of bridgmanite and fper from other sublithospheric diamond locations suggest that subducted oceanic harzburgite is the principal source of peridotitic lower-mantle diamonds, providing a lithological identity to the revelation made by seismology of slab penetration into the lower mantle.

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