Rhenium-osmium isotope systematics and platinum group element concentrations in oceanic crust

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ABSTRACT

Knowledge of the ^{187}Os/^{188}Os ratio as well as the inventories of rhenium and platinum group elements (PGE) in oceanic crust allows quantification of the proportion of recycled oceanic crust in oceanic basalt sources. Our knowledge is limited by the availability of well-characterized sections of oceanic crust, specifically of the plutonic, lower portion that has not been drilled in situ to the Moho. Here we report new data for plutonic rocks that compose the bottom 4680 m of an ocean crust section from the Oman ophiolite. Major and trace element data as well as mineral analyses indicate that Oman gabbros are primitive cumulates from melts similar to typical mid-oceanic ridge basalt. The mean weighted composition of this section (Re: 427 pg/g; Os: 55 pg/g; Ir: 182 pg/g; Pd: 2846 pg/g; Pt: 4151 pg/g; initial ^{187}Os/^{188}Os: 0.142) indicates significantly higher Os and lower Re concentrations than previously analyzed partial sections of ocean crust that lack cumulate lower crust [Deep Sea Drilling Project–Ocean Drilling Program (DSDP-ODP) Hole 504B, ODP Hole 735B], emphasizing that the lower, cumulate oceanic crust dominates the Os budget of oceanic crust. Analyses of mineral grain size fractions indicate that rhenium, PGE, and lead are enriched in the sulfur-rich, fine fraction. This corroborates the notion that small accessory phases, and the melt migration processes affecting them, control these elements' budgets, distributions, and susceptibilities to alteration. The Re-Os-PGE inventories of a hypothetical 6.5-km-thick composite section that consists of 1825 m of DSDP Hole 504B–like upper oceanic crust and 4680 m of Oman-like lower ocean crust (Re: 736 pg/g; Os: 45 pg/g; Ir: 133 pg/g; Pd: 2122 pg/g; Pt: 2072 pg/g; initial ^{187}Os/^{188}Os: 0.146) provide a new comprehensive assessment of oceanic crust composition. Upon recycling and mixing with reasonable proportions of mantle peridotite, this composite requires at least 2 G.y. to develop sufficiently radiogenic ^{187}Os/^{188}Os to generate high μ (HIMU: $\mu = {}^{238}$U/^{204}Pb) basalts.

INTRODUCTION

During partial melting of the Earth's mantle, it is commonly inferred that Os behaves as a compatible element, partitioned preferentially into the solid residue, while Re is considered to be a moderately incompatible element. This is supported by the observation that most primitive basalt and picrite lavas, closely approximating partial melts of mantle peridotite, have lower Os and higher Re concentrations than residual mantle peridotites (e.g., Shirey and Walker, 1998). Similarly, high and nearly flat primitive mantle normalized platinum group element (PGE: Ru, Rh, Pd, Ir, Os, Pt) abundance patterns in slightly to moderately melt-depleted lherzolite suggest that all or most of these elements are compatible during melting (Rehkämper et al., 1997; Lorand et al., 1999).

We use the term cumulate to refer to plutonic rocks formed by partial crystallization from a primitive melt, after which the remaining melt was removed; we do not mean to imply any specific process for separation of crystals from melt. We use the term primitive to refer to lavas with molar Mg/(Mg + Fe), or Mg#, >0.65, and to plutonic rocks with Mg# >0.8. We use the term evolved to refer to lavas with Mg# <0.65 and plutonic rocks with Mg# <0.8. Most mid-oceanic ridge basalt (MORB) lavas are evolved, and thus underwent crystal fractionation below the seafloor, in the lower crust, forming primitive, cumulate plutonic rocks (usually gabbros) from primitive magmas.

Given that Os is compatible while Re is moderately incompatible, one might expect that cumulate gabbros would have higher Os and lower Re concentrations than evolved lavas, assuming that the phases that control solid/liquid partitioning of Re and Os during crystallization are similar to those that control partitioning during melting. Primitive cumulates in the lower oceanic crust should therefore dominate the PGE budget of the oceanic crust and bias Re/Os toward values lower than those observed in MORB. The primitive cumulate component of the oceanic crust has not been drilled in situ. The only in situ plutonic sections are from Ocean Drilling Program (ODP) Hole 735B and Site 894. These have average compositions that are evolved, not primitive. Surprisingly, perhaps due to their evolved compositions, gabbros from the top 500 m of ODP Hole 735B (Blusztajn et al., 2000) have PGE (e.g., Os: 9 pg/g) and Re concentrations (median Re: 487 pg/g; average Re: 2.5 ng/g) similar to those in MORB (Roy-Barman and Allègre, 1994; Peucker-Ehrenbrink et al., 2003; Gannoun et al., 2007). However, interpretation of the data is complicated by the ill-constrained abundance of PGE-rich troctolite, a trace lithology in this section through oceanic crustal gabbros.

Average MORBs record ~50% crystal fractionation (e.g., Kelemen et al., 1997a) with low molar Mg# ~0.5 and elevated incompatible element concentrations (e.g., Zr: ~100 μg/g) compared to primitive MORBs with Mg# ~0.7 and, e.g., Zr: ~50 μg/g. There must be complementary crystal products of this fractionation with Mg# >0.8 and, e.g., Zr: <50 μg/g. Dick et al. (2000) and Hart et al. (1999) emphasized that the average composition of gabbros from Hole 735B corresponds closely to that of a primitive, mantle-derived melt with Mg# ~0.7, which would be an evolved composition for a cumulate gabbro. Surprisingly, incompatible element concentrations in the average of Hole 735B gabbro samples are high (e.g., Zr: 78 μg/g), similar to a lava composition such as average MORB. Thus, the average 735B gabbro composition cannot be the primitive complement to typical evolved MORB. Similarly, Deep Sea Drilling Project (DSDP) Site 504B, drilled into lavas and sheeted dikes of the Nazca plate, is sometimes considered as an analogue for plutonic oceanic crust, but has a composition similar to that of the overlying lavas, not that of the cumulate lower crust (Bach et al., 2003).

The evolved, MORB-like nature of the Hole 735B plutonic section, and of the sheeted dikes at ODP Site 894, emphasizes the need to analyze more refractory, primitive oceanic cumulates, the complementary crustal reservoir to MORB lavas. Prior geochemical work on in situ upper crust such as DSDP-ODP Sites 417, 418, 504 (Bach et al., 2003; Peucker-Ehrenbrink et al., 2003), and 801 (Reisberg et al., 2008), and evolved gabbros at ODP Hole 735B (Hart et al., 1999; Blusztajn et al., 2000) and Site 894 (Lecuyer and Reynard, 1996) therefore needs to be complemented with geochemical and petrologic studies of primitive gabbroic cumulates from oceanic lower crust. Here we use well-characterized samples from the lower crustal section of the Wadi Tayin and Samail massifs in the Oman ophiolite to augment data from submarine sampling and from metamorphosed sections of orogenic ophiolites (Becker, 2000; Dale et al., 2007) in order to more accurately assess the global Re-PGE chemistry of the oceanic crust.

If the lower oceanic crust dominates the PGE budget and dilutes Re enrichments observed in

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Geology, March 2012; v. 40; no. 3; p. 199–202; doi:10.1130/G32431.1; 4 figures; Data Repository item 2012054.

altered upper oceanic crust, the long-term evolution of subducted oceanic crust is controlled by the chemical composition of the lower oceanic crust, which would have the effect of allowing for more recycled crust in global mass balance models. In addition, a PGE-rich lower oceanic crust would be much less susceptible to hydrothermal alteration of PGE inventories and initial ^{187}Os/^{188}Os. Consequently, the long-term evolution of the Re-Os isotope system and PGE concentrations in oceanic crust might not be noticeably affected by hydrothermal alteration (cf. Reisberg et al., 2008), thereby better recording primary igneous processes occurring during the creation and recycling of oceanic lithosphere.

OMAN OPHIOLITE AND OCEANIC CRUST

The Oman ophiolite has one of the best exposed and intensely studied sections of plutonic oceanic crust in the world. Because the crustal section of the Oman ophiolite includes a continuous layer of sheeted dikes overlain by pillow basalts, it is clear that most of the igneous rocks of the ophiolite formed at a submarine spreading center. On the basis of radiometric age data, subdued crustal thickness variations, a general lack of paleo-fracture zones, a nearly continuous layer of gabbro between volcanics and mantle, and other geological observations, it is probable that the ophiolite formed ca. 96 Ma (Tilton et al., 1981; Hacker et al., 1996; Warren et al., 2003) at a medium- to fast-spreading ridge (Nicolas, 1989).

In contrast to the well-exposed and well-studied northern Oman massifs that show a polygenetic history with the importance of a subduction component increasing over time (Alabaster et al., 1982; Lippard et al., 1986), the few data on dikes and lavas from the southern Oman massifs (Samail, Wadi Tayin) are comparatively similar to MORB, and there is no evidence for a second, depleted lava series (Pallister and Knight, 1981; Pearce, Kelemen and Braun, unpublished data).

Orthopyroxene is essentially absent from the lower crustal section in the southern massifs (Pallister and Hopson, 1981), though orthopyroxene-bearing cumulates form small, isolated intrusions and dikes in the mantle section (Amri et al., 1996; Benoit et al., 1996; Kelemen et al., 1997a). Most mantle spinels plot in the compositional field of spinel from abyssal peridotite samples. We therefore hypothesize that there is a gradient in igneous processes and composition of the Oman ophiolite, with the northern massifs recording a polygenetic history involving an increasingly important subduction component, while the southern massifs were formed primarily via a MORB-like, single-stage process.

Data for the southern massifs suggest that >95% of the igneous rocks form a single liquid line of descent (Pallister and Hopson, 1981; Pallister and Knight, 1981; Browning, 1984; Benoit et al., 1996; Kelemen et al., 1997b; Korenaga and Kelemen, 1997; Pearce, Kelemen and Braun, unpublished data), similar to average MORB differentiation. Oman samples tend to plot toward the depleted end of the MORB spectrum (e.g., Meyer et al., 1989). However, provided this is kept in mind, the southern massifs are perhaps the world's best exposure of MORB-like oceanic crust that can be studied to understand its geochemical composition.

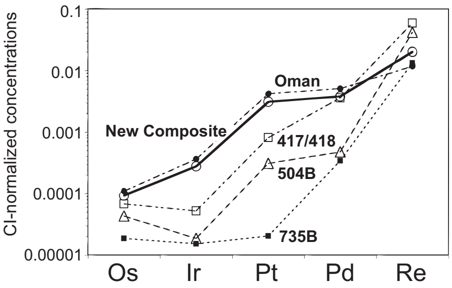

Figure 1. CI-chondrite (Anders and Grevesse, 1989) normalized platinum group element abundance pattern. Open squares—average Deep Sea Drilling Project (DSDP) Sites 417/418; triangles—DSDP Hole 504B (Peucker-Ehrenbrink et al., 2003); solid squares—Ocean Drilling Program Hole 735B (Blusztajn et al., 2000); filled circles—Oman crustal section (this paper); open circles—composite ocean crust (this paper). Note that logarithmic scale diminishes significant concentration differences between some of these crustal averages, e.g., Re concentrations of 2153 pg/g (DSDP Site 417/418) and 427 pg/g (Oman).

RESULTS AND DISCUSSION

Gabbroic rocks from the Wadi Tayn and Samail crustal sections have fractionated primitive mantle normalized PGE abundance patterns with low Os and Ir, but high Pt and Pd concentrations (see the methods description, Tables DR1 and DR2, and Fig. DR1 in the GSA Data Repository[1]). Average weighted concentrations of this section are Re: 427 pg/g; Os: 55 pg/g; Ir: 182 pg/g; Pd: 2846 pg/g; and Pt: 4151 pg/g (Fig. 1). Average present-day and age-corrected (96 Ma) ^{187}Os/^{188}Os are 0.203 and 0.142, respectively. This demonstrates that lower oceanic crust is the main PGE reservoir in the oceanic crust, and that the average Re concentration in these cumulates is much lower than in lavas at, for example, DSDP Hole 504B (Peucker-Ehrenbrink et al., 2003). The average initial ratio of this crustal section coincides with the upper bound of normal MORB (0.133 ± 0.009, 2σ; Gannoun et al., 2007), making such crust a potential contaminant of MORB lavas.

Whereas bulk-rock data do not show any correlations between sulfur (S) and chalcophile element (Re, PGE, Pb) concentrations, analyses of grain size fractions that were separated by electric pulse disaggregation (Andres et al., 2001) followed by wet sieving and gravitational settling reveal that Re and the PGE are mainly carried by the smallest crystal size fraction, which also has high S (Fig. 2) and Pb concentrations (Fig. 2B), most likely in the form of sulfide minerals. Sulfide phases control the PGE geochemistry of the mantle through the formation of sulfide liquids that are immiscible with silicate melts, inclusions of sulfides within silicate phases, and formation of PGE-rich residual phases after consumption of base-metal sulfides at high degrees of melting (Luguet et al., 2007).

An interesting feature of the Oman gabbros is their subchondritic average Os/Ir of ~0.3. The Os/Ir ratio does not correlate with absolute abundance of these elements (Fig. 3A) or with ^{187}Os/^{188}Os (Fig. 3B). Significant deviations from the chondritic ratio of ~1 are surprising, because Ir is generally viewed as a geochemical analog to Os during magmatic processes (Brügmann and Naldrett, 1988; Puchtel and Humayun, 2000). The Os/Ir fractionation observed in Oman gabbros is opposite to that observed in upper crustal lavas from DSDP Hole 504B (average Os/Ir of ~2.4; Peucker-Ehrenbrink et al., 2003). Harzbur-

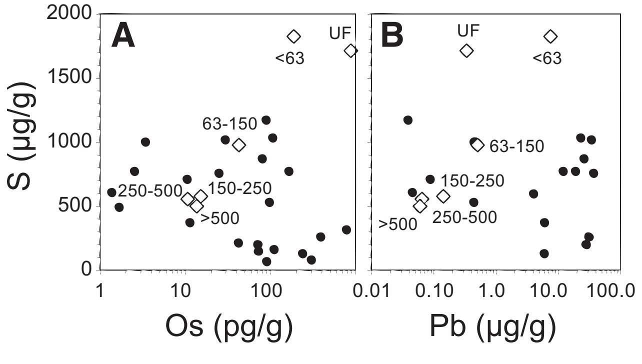

Figure 2. A: Sulfur concentration versus Os concentrations. B: Sulfur concentration versus Pb concentrations. Bulk samples are shown as black circles, grain size separates are shown as diamonds. Numbers on grain size fractions in inset refer to sieve sizes (in µm) and ultrafine (UF) fraction of grains that were separated by electric pulse disaggregation.

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[1] GSA Data Repository item 2012054, method description, Tables DR1 and DR2, and Figure DR1, is available online at www.geosociety.org/pubs/ft2012.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

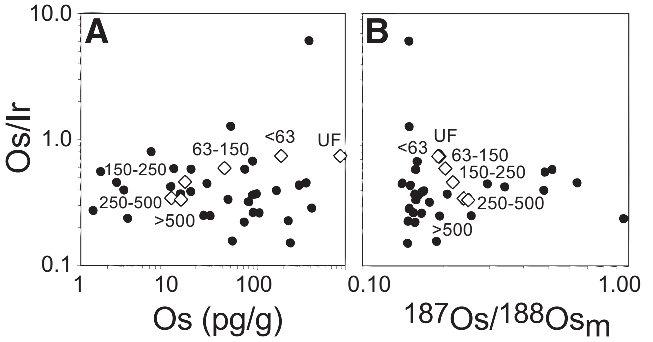

Figure 3. A: Os/Ir versus Os concentration. B: Os/Ir versus measured (m) ^{187}Os/^{188}Os. Bulk samples are shown as black circles, grain size separates are shown as diamonds. Numbers on grain size fractions in inset refer to sieve sizes (in μm) and ultrafine (UF) fraction of grains that were separated by electric pulse disaggregation.

gites from the mantle section of the Wadi Tayin massif and abyssal peridotites have chondritic Os/Ir, typical of upper mantle rocks (Hanghoj et al., 2010). If the Wadi Tayin harzburgites are representative of the mantle source, then the subchondritic Os/Ir in Oman gabbros cannot represent a source signature. However, Oman dunites have superchondritic as well as subchondritic Os/Ir (0.5–8.3; Hanghoj et al., 2010). As laurite included in chromite has been observed in Oman dunites (Ahmed and Arai, 2002; Ahmed et al., 2006), we hypothesize that igneous laurite and/or other sulfides with high Os/Ir control fractionation of Os from Ir during melting, melt extraction, or crystal fractionation.

The observation that present-day ^{187}Os/^{188}Os ratios correlate inversely with Os concentrations likely reflects the sensitivity of Os-poor lithologies to uptake of hydrogenous Os and Re during interaction with hydrothermal fluids at or near the ocean ridge (Peucker-Ehrenbrink et al., 2003; Reisberg et al., 2008). Preferential uptake of Re, possibly combined with some exchange of Os between crust and seawater, leads to ^{187}Re/^{188}Os values as high as ~1400 in some gabbros and some scatter in the Re-Os isochron diagram (Fig. 4). However, some of this scatter may also result from performing Re analyses on different sample splits from the Os analyses, and from significant blank corrections of Os-poor samples (see the Data Repository). As the extent of Re enrichment determines the long-term evolution of ^{187}Os/^{188}Os in recycled oceanic crust, it is important to note that the Re concentration of seawater coupled with estimates of hydrothermal fluxes through oceanic crust limit the globally averaged Re uptake in altered oceanic crust to a few percent of the magmatic inventory (Peucker-Ehrenbrink et al., 2003).

To assess the role of altered oceanic crust as a contaminant of mantle-derived melts, we calculate Re-Os-PGE inventories of a hypothetical 6.5-km-thick composite section of intermediate- to fast-spreading oceanic crust that consists of 1825 m of DSDP Hole 504B–like upper oceanic crust (Bach et al., 2003; Peucker-Ehrenbrink et al., 2003) and 4680 m of Oman-like lower ocean crust (see the Data Repository). The weighted chemical and isotope characteristics of this hypothetical crust section, age corrected to resemble newly formed crust altered at the mid-ocean ridge, are Re: 736 pg/g; Os: 45 pg/g with an initial ^{187}Os/^{188}Os of 0.146; Ir: 133 pg/g; Pd: 2122 pg/g; and Pt: 3072 pg/g (Fig. 1). The two main uncertainties with the PGE budget of the 4680 m Oman section are related to the weighting procedure and the variable thickness (~10–100 m) of the PGE-rich mantle transition zone. Average PGE concentrations are therefore uncertain by ~±30%. This uncertainty includes replicate analyses that in some cases differ in concentrations by more than an order of magnitude, presumably caused by the nugget effect (Peucker-Ehrenbrink et al., 2003).

Our new composite, intermediate- to fast-spreading oceanic crust is more enriched in Re (736 pg/g vs. 480 pg/g in ODP Hole 735B), less depleted in PGE (e.g., Os: 45 pg/g vs. 9 pg/g; see Fig. 1), and has a lower ^{187}Re/^{188}Os (~79 vs. 257) than observed in average gabbros at ODP Site 735B. The higher Re concentrations compared to the Hole 735B section imply more rapid ingrowth of a radiogenic ^{187}Os/^{188}Os signature when the effects of mixing of mantle peridotite with subducted oceanic crust is modeled in an attempt to understand the formation of radiogenic ^{187}Os/^{188}Os (~0.15) in high μ (HIMU: $\mu = {}^{238}$U/^{204}Pb) type ocean island basalts. However, even this more Re-rich composite crust requires an unreasonably large crustal proportion (35%–40%) in order to generate HIMU-like ^{187}Os/^{188}Os in less than 2 G.y. Only Re-rich altered upper oceanic crust (DSDP Site 417/418, DSDP-ODP Hole 504B) will develop HIMU-like ^{187}Os/^{188}Os signatures in less than 2 G.y. with more reasonable proportions of recycled crust (~20%; see Fig. DR2). However, none of the ocean crust sections studied thus far will develop complementary HIMU characteristics (unradiogenic ^{87}Sr/^{86}Sr, Pb isotopes, characteristic trace element pattern) without further chemical modifications during subduction (Hart and Staudigel, 1989; Bach et al., 2003; Kelley et al., 2005). While such processing in subduction zones potentially meets the Sr and Pb isotope constraints, the projected ingrowth of radiogenic ^{187}Os/^{188}Os may be retarded by the preferential loss of Re from the Re-rich basaltic (Becker, 2000; Dale et al., 2007), but apparently not the gabbroic (Dale et al., 2007), portion of the subducting slab.

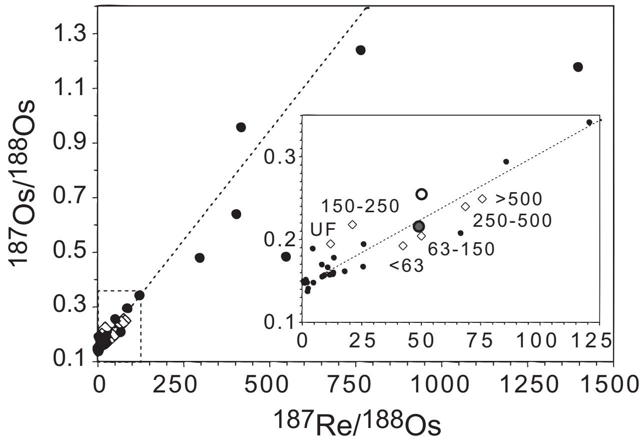

Figure 4. ^{187}Os/^{188}Os versus ^{187}Re/^{188}Os (with 96 m.y. reference line). Large open black circle marks split of gabbro that was analyzed in bulk. Large gray circle with black rim denotes reconstituted mass balance of all grain size fractions from another split of same gabbro, taking bulk weights and concentrations of each fraction into account. Bulk samples are shown as black circles, grain size separates are shown as diamonds. Numbers on grain size fractions in inset refer to sieve sizes (in μm) and ultrafine (UF) fraction of grains that were separated by electric pulse disaggregation.

ACKNOWLEDGMENTS

The late Brian Schroeder prepared sample powders. Lary Ball and Dave Schneider helped with Re and platinum group element analyses in the National Science Foundation (NSF) supported (grants EAR/IF-0318137, EAR/IF-0651366) Woods Hole Oceanographic Institution (WHOI) Plasma Mass Spectrometry Facility. We thank Ed Ripley (Indiana University) for sulfur concentration and isotope analyses, and Jochen Hoefs (Göttingen University, Germany) for O isotope analyses. Marguerite Godard, Carlos Garrido, and Charles Knack helped with the major and trace element analyses that were carried out in the X-ray fluorescence laboratory at Washington State University. Anna Cipriani helped with the Pb isotope analyses at Lamont-Doherty Earth Observatory. Oman samples were collected with NSF support (grants OCE-9416616, OCE-9711170, OCE-9819666, OCE-0118572) to Kelemen. Kelemen and Peucker-Ehrenbrink acknowledge support from NSF grant EAR-0337677. WHOI and the NSF (grant EAR/IF-9905530) funded the electric pulse disaggregator. We thank Laurie Reisberg, Steven Shirey, and an anonymous reviewer for insightful comments.

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Manuscript received 10 May 2011
Revised manuscript received 20 September 2011
Manuscript accepted 28 September 2011

Printed in USA