



Mineralogical control of selenium, tellurium and highly siderophile elements in the Earth's mantle: Evidence from mineral separates of ultra-depleted mantle residues



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ABSTRACT

The budget of Se–Te and other highly siderophile elements (HSEs) in major silicate and oxide mineral separates (olivine, orthopyroxene, clinopyroxene, spinel), and intergranular fine components of an ultra-refractory harzburgite have been determined via isotope dilution hydride generation ICPMS after mineral separation, high-pressure acid digestion and chemical purification. In addition, x-ray computed micro-tomography (micro-CT) has been performed in order to constrain and illustrate the original location, distribution and size of sulfides and/or alloys inside their intact, bulk harzburgite matrix. The results show that the whole-rock Se–Te budget of mantle residues, that are devoid of base metal sulfides (BMS) after high degrees of partial melting ($F \approx 23\%$), is almost completely controlled by olivine-hosted micro-inclusions and intergranular fractions of platinum group minerals (PGMs). The scarcity and heterogeneous distribution of intergranular PGMs generally sized 3–5 μm in the harzburgite are revealed by 3D micro-CT, while PGM inclusions in olivine are smaller than 2.5 μm . The heterogeneous distribution of these microphases as well as the range of Se–Te contents in the bulk harzburgite complicate a quantification of individual contributions of separates to the whole-rock Se–Te budget. However, PGM micro-inclusions in olivine with residual-type CI chondrite-normalized HSE patterns and suprachondritic Se/Te ratios between 30 and 218 ($\text{Se/Te}^{\text{CI chondrite}} = 9$) host close to 100% of the bulk rock Se. Interstitial fine components up to 125 μm grain size may contribute only minor amounts of Se, but up to 50% of Te to the bulk harzburgite and show subchondritic Se/Te ratios of ca. 4, resembling metasomatic PGM signatures. In dependence on increasing proportions of interstitial and metasomatic PGMs vs. olivine-hosted residual PGMs, the bulk rock Te contents may increase and the Se/Te ratios decrease. The systematics seen in mineral separates of one harzburgitic sample resembles the entire spectrum of Se–Te signatures of all suites of peridotites and host phases so far published. The results are consistent with a higher compatibility of Se in covalent monosulfides compared to Te that prefers sulfide melts. It is further consistent with the stabilization of residual PGMs after Mss exhaustion as well as with formation of metasomatic PGMs in a S–Cu–volatile-rich sulfide melt. Altogether this reconciles a higher incompatibility of Te over Se during partial mantle melting, subsequent removal of Te over Se with Cu–Ni-rich sulfide fractionation from high-degree partial mantle melts, and explains the similar Te but higher Se abundances in MORBs compared to peridotites that is globally observed.

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1. Introduction

Highly siderophile elements (HSE: Re and platinum group elements [PGE]) show an excess abundance in the Earth's mantle compared to abundances predicted by core–mantle differentiation (Morgan, 1986; Lorand et al., 2008; Walker, 2009; Rose-Weston et al., 2009). The most common, but not unchallenged, hypothesis to explain this HSE-mantle excess is the addition of these elements via meteoritic bombardments

(e.g., Holzheid et al., 2000) shortly after core–mantle differentiation (Late Veneer, Kimura et al., 1974). The characterization of these hypothetical meteoritic components may also provide clues on the potential source of terrestrial volatiles that altogether allowed for the development of life on our planet. Selenium and tellurium belong to the group of HSE and thus show the same “excess” mantle abundances that are also attributed to the Late Veneer (Morgan, 1986). Hence, provided that the Se–Te–HSE composition of the primitive mantle (PM: bulk silicate Earth reservoir without the metallic core) is firmly constrained, inferences on the nature of the Late Veneer composition may be possible. Constraining the Se–Te signatures of the PM requires reliable information on the behavior of both elements during mantle melting processes

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that are among the most significant processes obscuring pristine abundances of available PUM estimates (i.e., the upper part of the PM). The absolute and relative compatibilities of Se and Te during mantle melting are still not well constrained and remain critically debated issues. Experimentally determined sulfide melt–silicate melt partition coefficients (Rose-Weston et al., 2009), analyses of MORB sulfide globules (Peach et al., 1990; Patten et al., 2013) and analyses of fertile and some residual peridotites (Marchesi et al., 2013; Wang et al., 2013) were previously used to argue for a higher relative compatible behavior of Te compared to Se during mantle melting. This is counterintuitive because, as non-metal, Se can replace S in refractory Fe–Ni monosulfides while Te, as a semi-metal, preferentially partition into metal-rich sulfide melts that have lower melting points in the upper mantle (Helmy et al., 2010). In contrast to the other HSE, combined Se and Te analyses of the same mantle-derived rocks are relatively rare (Morgan, 1986; Lorand and Alard, 2010), albeit Se was determined on several suites of mantle xenoliths worldwide (Lorand et al., 2003, 2004). Nevertheless, combined analyses of Se and Te have recently received increasing attention due to analytical advancements, allowing for the determination of low ppb- and sub-ppb-level Se and Te concentrations in mantle rocks, respectively (König et al., 2012). A number of high-precision isotope dilution Se and Te studies on fertile and depleted peridotites from various tectonic settings followed (König et al., 2012, 2014; Wang and Becker, 2013; Wang et al., 2013). An assessment of all data from these studies by König et al. (2014) in the context of other HSE and their dominant host mineral phases (base metal sulfides–BMS and platinum group minerals–PGM; Peach et al., 1990; Rose-Weston et al., 2009; Lorand and Alard, 2010) demonstrated that Se–Te systematics of fertile mantle peridotites can be explained by secondary magmatic processes involving addition of metasomatic Se–Te-rich phases (BMS and tellurides) that were admixed in different proportions to residual BMS and PGM. This heterogeneous mixing between residual and increasing amounts of metasomatic BMS + PGM phases produces a range of Se/Te around near-chondritic values (Se/Te of CI chondrite ≈ 9 ; Lodders et al., 2009), which may appear like primitive features of the Earth's mantle, while being in fact clearly secondary features. On the other hand, repeated analysis of depleted peridotites showed suprachondritic Se/Te ratios that increased with decreasing Te contents, with a very poor reproducibility compared to more fertile peridotites (König et al., 2012). This was attributed to (1) a preserved feature of melt depletion during which Te is more incompatible than Se and (2) heterogeneously distributed Se and Te-bearing microphases inside the bulk harzburgite matrix.

Hence, reliable PM estimates require firm constraints on the behavior of HSE during mantle melting. The HSE abundances in MORB are typically lower than in fertile mantle and the CI-normalized PGE patterns of MORB show depletions in IPGE (Os, Ir, Ru) relative to PPGE (Pt, Pd) (e.g., Rehkamper et al., 1999; Bézou et al., 2005; Lissner et al., 2014). Yet it remains a matter of debate which parameters control the HSE fractionation during partial mantle melting. One scenario is that during mantle melting, BMS as the dominant HSE host phases melt incongruently (Barnes et al., 1997; Alard et al., 2000; Lugué et al., 2003; Bockrath et al., 2004; Ballhaus et al., 2006) to form a Cu–Ni-rich sulfide melt that is physically removed and scavenges the incompatible PPGEs (Pt, Pd) from a refractory monosulfide solid solution (Mss) that retains the compatible IPGEs (Os, Ir, Ru) in the residue. However, some studies pointed out that the temperatures of partial melting in MOR-type melting regimes are too high to allow Mss stability (Fonseca et al., 2012; Mungall and Brenan, 2014; and references therein). In this case, Mss/sulfide melt partitioning would not play a significant role during partial melting, at least not in MOR-type settings. These authors propose an alternative scenario, in which all HSEs are hosted in extracted sulfide melt (no residual Mss stable) and the HSE fractionation is controlled by residual PGM phases (Os–Ir–Ru- and Pt- alloys) that form in response to the consumption of sulfide liquid after high degrees of partial melting ($F > 15\%$) (e.g., Fonseca et al., 2012;

Mungall and Brenan, 2014). The existence of PGMs was indeed confirmed in natural samples (e.g., Lugué et al., 2007; Lorand et al., 2010). The stabilization of residual PGMs, on the other hand, requires high-degree melt fractions ($F > 15\%$) that fully exhaust residual sulfide. In this case, low degree partial melts ($F < 15\%$) would contain virtually no HSEs because of the high sulfide melt/silicate melt partition coefficients (e.g., Mungall and Brenan, 2014). The latter is however unsupported by the global HSE database for MORB (Hertogen et al., 1980; Rehkamper et al., 1999; Bézou et al., 2005; Lissner et al., 2014; Yang et al., 2014) as well as the elevated HSE abundances in OIB (Yi et al., 2000; Day, 2013) and pyroxenites (Montanini et al., 2013) that are thought to typically originate from lower degree partial melts ($F < 10\%$) compared to MORB. In the scope of these unsolved issues, our study targets the extensively studied high-degree melting residue (Lherz harzburgite 64-3), in order to constrain the mineralogical controls on the chalcogenides Se and Te. Knowledge about the Se–Te systematics in harzburgites will provide additional information on the behavior of HSEs during partial melting since Se and Te will show opposite or similar geochemical behaviors, depending if Mss or sulfide melt is the residual sulfide phase during the melting process ($D_{\text{Se}}^{\text{Mss/sulf.melt}}/D_{\text{Te}}^{\text{Mss/ulf.melt}} > 1$; Helmy et al., 2010).

Recent papers on Se–Te systematics of fertile, PUM-like peridotite compositions provide evidence on that the Se budget is 100% hosted by disseminated BMS, while Te is alloyed with PGE (Pt, Pd) and other semi metals (e.g., Bi), forming discrete microphases (Lugué et al., 2004; Lorand et al., 2008; Lorand and Alard, 2010). By contrast, the location and exact distribution of Se–Te-bearing phases inside highly depleted peridotites remain highly speculative, which hampers quantitative constraints regarding their impact on the whole-rock Se–Te budget. A pioneering microstudy focusing on major mineral separates (olivine, orthopyroxene, clinopyroxene and Cr-spinel) and intergranular material, analyzed by isotope dilution (Lugué et al., 2007) provided important constraints. This study revealed that the major silicates and oxides account for less than 30% of the whole-rock PGE budget, whereas between 50 to 100% of the PGEs are accounted for by the intergranular material that contain residual Os–Ru sulfides and Os–Ir alloys, interpreted to have formed in response to the complete consumption of BMS by the high degree of partial melting (i.e., 23–24%). The aim of our paper is to replicate such a study for Se and Te systematics in one of the original Pyrenean peridotites (64-3) that was analyzed by Lugué et al. (2007). Harzburgite 64-3 is an ultra-depleted orogenic peridotite from the Lherz massif, showing similar whole-rock Se–Te–HSE signatures to other suites of harzburgites previously analyzed, that are considered to be representative of mantle residues after complete consumption of BMS following high degrees of partial melting (Lorand et al., 1999). It is also a key sample for which a large range of Se/Te ratios attributed to whole-rock heterogeneity has previously been documented (König et al., 2012). Our aim is to provide constraints on (1) the contribution of the major minerals and intergranular components to the whole-rock Se and Te budget, (2) the signature of Se–Te inside these individual phases, (3) the relative compatibility of Se and Te from the perspective of the individual abundances of these phases, (4) to provide clues on the origin of the whole-rock heterogeneity and the poor reproducibility of Se and Te concentrations and (5) to search for a closest-to-primary signature of melt depletion inside highly refractory peridotites.

2. Sample material

The Lherz massif contains abundant, deformed, up to 10 m thick harzburgite lenses inside layered lherzolites that were altogether emplaced to crustal levels following collision of the Iberian plate with the Eurasian plate in the middle Cretaceous (e.g., Fabries et al., 1991). Sample 64-3 contains mm-sized olivine and orthopyroxene in addition to minor amounts (generally <2%) clinopyroxene and spinel (Lorand et al., 1999); it is only moderately serpentinized (LOI: 2.9 wt.%,

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