



The behaviour of incompatible elements during hydrous melting of metasomatized peridotite at 4–6 GPa and 1000 °C–1200 °C



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ABSTRACT

Trace element behavior during hydrous melting of a metasomatized garnet–peridotite was examined at pressures of 4–6 GPa and temperatures of 1000 °C–1200 °C, conditions appropriate for fluid penetrating the mantle wedge atop the subducting slab. Experiments were performed in a rocking multi-anvil apparatus using a diamond–trap setup. The compositions of the fluid and melt phases were measured using the cryogenic LA-ICP-MS technique. The water-saturated solidus of the K-lherzolite composition is located between 900 °C and 1000 °C at 4 GPa and between 1000 °C and 1100 °C at 5 and 6 GPa. The partition coefficients between fluid or melt and clinopyroxene reveal an asymmetric MREE trough with a minimum at Dy. The clinopyroxene in equilibrium with aqueous fluids is characterized by $D_{\text{U}}^{\text{fluid-cpx}} > D_{\text{Th}}^{\text{fluid-cpx}}$ while $D_{\text{U}}^{\text{melt-cpx}}$ tends to be similar to $D_{\text{Th}}^{\text{melt-cpx}}$. The partition coefficients between fluid or melt and garnet reveal very strong light to heavy REE fractionation, $D_{\text{La}}/D_{\text{Lu}}$ from 95 (hydrous melt) to 1600 (aqueous fluid). The LILE are highly incompatible with partition coefficients > 50 . The behavior of HFSE are decoupled, with $D_{\text{Zr,Hf}}$ close to 1 while $D_{\text{Nb,Ta}} > 10$. Garnet is characterized by $D_{\text{U}}^{\text{melt/fluid-garnet}} < D_{\text{Th}}^{\text{melt/fluid-garnet}}$. A comparison of our experimental partitioning results for trivalent cations as well as the results from the literature and the calculations carried out using the lattice strain model adapted to the presence of water in the bulk system indicates that H₂O in the fluid or melt phase has a prominent effect on trace element partitioning. Garnet in mantle rocks in equilibrium with an aqueous fluid is characterized by significantly higher $D_{\text{O}}(3+)$ for REE in the X site of the garnet compared with the partitioning values of the optimal cation in garnet in equilibrium with hydrous melts. Our data show for the first time that the change in the nature of the mobile phase (fluid vs. melt) does affect the affinities of trace elements into the garnet crystal at conditions below the second critical endpoint of the system. The same also applies for clinopyroxene, although this is less clear. Consequently, our new data allow for refinements in predictive modeling of element transfer from the slab to the mantle wedge and of possible compositions of metasomatized mantle that sources OIB magmatism.

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1. Introduction

Fluid-mediated metasomatism and melting in subduction zones play a key role in the geochemical evolution of the Earth's mantle and crust. Dehydration of the subducting hydrated and altered oceanic lithosphere releases fluids rich in mobile elements into the overlying mantle wedge, leaving a residual slab rich in immobile elements. Many mantle rocks have been documented for evidence of trace element enrichment accompanying enhanced levels of volatile components, specifically associated with the presence of hydrous phases such as phlogopite, amphibole, and apatite (e.g., Maury et al., 1992; O'Reilly and Griffin, 1988; Wass and Rogers, 1980). Melting of the metasomatized mantle wedge is the source of island arc lavas. Many of the distinctive chemical and isotopic characteristics of such lavas are attributed to melt

generation in mantle source regions metasomatized by aqueous fluids and/or hydrous melts released from the oceanic subducting slab (Kushiro, 1987; Tatsumi et al., 1986).

Trace elements have negligible influence on the composition and stability of major phases, and thus have no direct control on their formation or consumption; however, prominent trace element enrichment may lead to stabilization of low amounts of specific accessory minerals that may provide a sink for specific elements (e.g., Nb-Ta in residual rutile; Stalder et al., 1998; Elliott et al., 1997). Trace elements *sensu stricto* are petrologically passive constituents, the distribution of which among the stable phases provides a window on the history of processes a given rock went through. It is thus not surprising that much of what we know about the chemical evolution of, and the processes having occurred in, the inaccessible Earth comes from trace element and isotope geochemistry. Interpretation of the chemical compositions of volcanic rocks or mantle xenoliths relies on our knowledge of high-temperature/high-pressure behavior of trace elements, especially how they are distributed among

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coexisting phases. The partitioning of elements between a fluid or melt phase and a mineral is given as partition coefficient (noted here as $D^{\text{fluid/melt-min}}$). Such parameters are thermodynamic variables, changing as a function of pressure, temperature, phase stabilities, and bulk compositions (e.g., Beattie, 1994; Blundy and Wood, 1994, 2003a, 2003b; Gaetani, 2004; Van Westrenen et al., 2000).

Previous experiments mainly documented trace element partitioning between mantle minerals and anhydrous melts for a wide range of conditions in the Earth's interior (e.g., Gaetani, 2004; Pertermann et al., 2004). Consequently, partition coefficients for hydrous systems are limited. There are only a few studies focusing on trace element partitioning in hydrous peridotitic systems, each focused on a limited set of trace elements at a narrow range of pressures of 1–3 GPa and temperatures of between 850 °C and 1370 °C (Ayers et al., 1997; Brenan and Watson, 1991; Brenan et al., 1995a, 1995b, 1998; Gaetani et al., 2003; Mysen, 1979; Tatsumi et al., 1986). Spandler et al. (2014) extended the P - T range to 4 GPa and 900 °C and used a natural low CaO/Al₂O₃ hydrated peridotite to constrain the subsolidus fluid trace element composition in equilibrium with garnet harzburgite. The scarcity of information on the behavior of a wide range of trace elements in hydrous peridotites at a wide range of pressures and temperatures is a significant impediment to our understanding of elemental fluxes in subduction zones. Serpentinites are arguably the most important carrier of water in slabs to subarc depths; hence, their dehydration seems central to triggering arc magma genesis (e.g., Spandler and Pirard, 2013). But how and to what extent slab fluids are modified during upward reactive flow in the wedge prior to partial mantle melting has remained obscure. Recently, evidence has been accumulated in support of a serpentinite signature in arc lavas, e.g., the recognition of a broad match in trace element pattern of fluid inclusions produced from antigorite breakdown with that of arc lavas (Scambelluri et al., 2004) or the finding of heavy B isotope signatures in lavas from the South Sandwich Islands (Scambelluri and Tonarini, 2012; Tonarini et al., 2011).

Despite the evidence for the role of aqueous fluids as transport agents and melt catalysts in subduction zones, the quantitative assessment of the composition and nature of these fluids and melts is largely lacking. In all the above studies, except for Spandler et al. (2014), who analyzed synthetic fluid inclusions, only the compositions of the solid phases in the peridotitic rocks were directly determined. The composition of the fluid or melt phase was either determined by examining quench precipitates in veins and in between peridotitic minerals or inferred from mass balance constrains. These approaches led to large uncertainties in the fluid or melt composition and, therefore, to significant uncertainties in the trace element partition coefficients dataset.

Kessel et al. (2015) studied the K-rich peridotite + H₂O system at pressures of 4–6 GPa and temperatures of 850 °C–1200 °C (corresponding to 120–180 km depth), conditions appropriate for fluid penetrating the mantle wedge atop the subducting slab (Schmidt and Poli, 1998; Tatsumi et al., 1986). The K-rich peridotite composition chosen for this study is used to represent metasomatized mantle as evident by the presence of phlogopite–spinel– and phlogopite–garnet–peridotite bodies found in ultrahigh pressure (UHP) terrains of orogenic zones. Such metasomatized mantle bodies were found, for example, in Ulten (Rampone and Morten, 2001), Cima di Gagnone (Scambelluri et al., 2014, 2015), and Sulu (Zhang et al., 2007). These small peridotitic bodies are interpreted to be remnants of mantle rocks within the mantle wedge and at slab–mantle interface in subduction zones exposed to metasomatic processes during subduction.

For K-rich peridotite–H₂O system, Kessel et al. (2015) reported the major element compositions of phases, determined the location of the water-saturated solidus, and the reaction occurring during melting. The current study is a continuation of Kessel et al. (2015) focusing on the trace element behavior during hydrous garnet peridotite melting at temperatures of 1000 °C–1200 °C. The results are used to calculate melt/fluid–mineral as well as melt/fluid–peridotite partition coefficients in order to constrain the sinks of specific trace elements and

thus the recycling rates of key elements from the mantle overlying the subducted slab to the continental arc possibly via ocean island magmatism.

2. Experimental procedure and analytical techniques

2.1. Starting materials

The starting material used in this study, identical to that used in Kessel et al. (2015), is a synthetic K-bearing lherzolite (KLZ) doped with 0.66 wt% Cr₂O₃, 0.08 wt% TiO₂ and 0.64 wt% K₂O similar to that used by Konzett and Ulmer (1999) and Fumagalli et al. (2009). This peridotite composition is based on the M. Briançon spinel lherzolite (French Central Massif) with the addition of 5 wt.% phlogopite and 0.4 wt. % Na₂O and final subtraction of 30 wt.% of olivine (Fo₉₁) to enhance the abundance of minor phases (i.e., phlogopite and pyroxenes). The composition chosen for this study is similar to several metasomatized high- P peridotites. For example, the composition of the Su-Lu peridotite (Zhang et al., 2007) after subtraction of 30% of olivine is shown in Table 1 in order to directly compare its composition to our starting material. The KLZ starting material was doped with 22 trace elements. Standard solutions of 1000 µg/g of each element were used to create 3 internal mixed-standard solutions (maximum 10 elements in each solution), containing approximately 50 µg/g of each element. A total of 2.1 ml from each internal standard solution were added to the starting material powder—equivalent to 150 µg/g of each trace element in the bulk composition. The powder was ground in an agate mortar for a total of 4 h and dried at 110 °C for 10 h. The trace element-doped lherzolite is labeled 'KLZT'. The composition of the starting material was determined by dissolving three aliquots for major element [given in Kessel et al. (2015)] and two aliquots for trace elements in a HF-HNO₃ mixture and by liquid ICP-MS analysis at the Geological Survey of Israel. The measured composition of the starting material is given in Table 1.

Table 1
Composition of K-lherzolite starting material used in this study.

KLZT	wt%	C25-60 (–30 ol) ^c		µg/g
SiO ₂	48.13(71) ^b	45.67	Be	120; 158 ^d
TiO ₂	0.084(002)	0.05	Rb	117; 148
Al ₂ O ₃	4.50(05)	5.48	Sr	122; 145
Cr ₂ O ₃	0.66(01)	0.66	Y	124; 148
FeO ^a	6.26(130)	7.17	Zr ^e	ND ^f
MgO	36.16(17)	35.68	Nb	123; 141
CaO	3.46(02)	3.72	Cs	120; 146
Na ₂ O	0.86(12)	0.37	Ba	120; 143
K ₂ O	0.64(03)	0.71	La	122; 150
Total	100.75	99.50	Ce	125; 153
			Nd	129; 158
			Sm	126; 155
			Eu	126; 157
			Gd	122; 153
			Dy	129; 154
			Er	129; 156
			Yb	122; 147
			Lu	127; 156
			Hf	124; 140
			Ta	120; 140
			Pb	123; 152
			Th	121; 146
			U	119; 154

^a All Fe are expressed as FeO.

^b Major element composition taken from Kessel et al. (2015). The average and one standard deviation of the starting material as measured in three aliquots. Numbers enclosed in parentheses indicate one standard deviation of the last digits quoted, i.e., 48.13(71) should be read as 48.13 ± 0.71 wt%.

^c Analyses taken from Zhang et al. (2007) representing the Su-Lu peridotite after final subtraction of 30 wt.% of olivine (Fo₉₁).

^d The results of trace element concentrations in analyses of two aliquots.

^e ND, no data. Zirconium was not analyzed in the starting material.

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