



Mobility of major and trace elements in the eclogite-fluid system and element fluxes upon slab dehydration

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Abstract

The equilibrium between aqueous fluids and allanite-bearing eclogite has been investigated to constrain the effect of temperature (T) and fluid composition on the stability of allanite and on the mobility of major and trace elements during the dehydration of eclogites. The experiments were performed at 590–800 °C and 2.4–2.6 GPa, and fluids were sampled as synthetic fluid inclusions in quartz using an improved entrapment technique. The concentrations and bulk partition coefficients were determined for a range of major (Mg, Ca, Na, Fe, Al, Ti) and 16 trace elements as a function of T and fluid composition. The results reveal a significant effect of T on element partitioning between the fluids and the solid mineral assemblage. The partition coefficients increase by more than an order of magnitude for most of the major and trace elements, and several orders of magnitude for light rare-earth elements (LREE) from 590 to 800 °C. The addition of various ligand species into the fluid at 700 °C results in distinctive trends on element partitioning. The concentrations and corresponding partition coefficients of most of the elements are enhanced upon addition of NaF to the fluid. In contrast, NaCl displays a nearly opposite effect by suppressing the solubilities of major elements and consequently affecting the mobility of trace elements that form stable complexes with alkali-(alumino)-silicate clusters in the fluid, e.g. high field strength elements (HFSE). The results further suggest that fluids in equilibrium with orthopyroxene and/or diopsidic clinopyroxene are peralkaline (ASI \sim 0.1–0.7), whereas fluids in equilibrium with omphacitic pyroxene are more peraluminous (ASI \sim 1.15). Therefore, natural aqueous fluids in equilibrium with eclogite at about 90 km depth will be slightly peraluminous in composition. Another important finding of this study is the relatively high capacity of aqueous fluids to mobilize LREE, which may be even higher than that of hydrous melts.

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1. INTRODUCTION

Fluids generated upon the dehydration of subducting oceanic lithosphere play an essential role in mass transfer of various elements from the slab to the overlying mantle wedge, ultimately leading to arc magmatism. It is well

established that the characteristic geochemical signatures observed in arc magmas display a significant contribution of slab – derived component (e.g. Morris et al., 1990; McCulloch and Gamble, 1991; Hawkesworth et al., 1993; Elliott et al., 1997). However, it remains unclear whether it is an aqueous fluid, a hydrous melt or a supercritical liquid that has the capacity to extract the elements upon slab dehydration/partial melting and further transport them to the source of arc magma generation. To address this uncertainty, considerable amount of information has been accu-

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mulated based on thermal models (e.g. Kincaid and Sacks, 1997; Syracuse et al., 2010; Bengtson and van Keken, 2012), isotopic characteristics of slab components (e.g. Hawkesworth et al., 1997; Elliott et al., 1997; Sigmarsson et al., 1998) as well as numerous experimental studies on mineral solubilities and trace element partitioning (e.g. Brenan et al., 1995a,b; Keppler, 1996; Stalder et al., 1998; Klemme et al., 2002; Kessel et al., 2005a). Yet, the uncertainties between the relatively low slab surface temperatures predicted by thermal models, which primarily define the nature of slab fluids, and the efficiency of various fluids to transport material from the slab to the mantle wedge require further investigation.

Among existing experimental studies, the partitioning of trace elements has been most intensively investigated at conditions of partial melting of subducting sediments and basaltic crust (e.g. Klemme et al., 2002; Kessel et al., 2005a; Klimm et al., 2008; Hermann and Rubatto, 2009; Skora and Blundy, 2010). It has been shown that temperature (T), pressure (P) and mineral chemistry of modal mineralogy mainly control the element partitioning during partial melting, while the generated partial melts are typically highly siliceous in composition (Peacock et al., 1994; Drummond et al., 1996; Ryabchikov et al., 1996; Hermann and Spandler, 2008). However, not all the characteristics of the typical trace element abundances and patterns of arc magmas can be explained invoking slab-derived partial melts as the primary phase responsible for slab to wedge element transfer. For example, a few experimental studies have demonstrated relatively low solubilities of accessory phases such as allanite and monazite in silicate melts at sub-arc conditions (Klimm et al., 2008; Hermann and Rubatto, 2009; Skora and Blundy, 2012), which in turn limits light rare earth element (LREE) budgets and the degree of light rare-earth elements over heavy rare-earth elements (LREE/HREE) fractionation if one of these phases is present in the solid residue. In addition, the highly silicic melts derived from the slab have high viscosities, compared to aqueous fluids or supercritical liquids (Hack and Thompson, 2011), which would certainly affect their mobility and hence their capacity as transport agents. However, compared to the extensive set of data available for hydrous melts, only few experimental studies are available on trace element partitioning during slab dehydration processes, where complex fluid–rock equilibria have been investigated (Green and Adam, 2003; Kessel et al., 2005a; Spandler et al., 2007). In the study of Green and Adam (2003), the partitioning of trace elements has been investigated in MOR basalt – H₂O system at subsolidus conditions of 650–700 °C and 3 GPa. The results revealed the general patterns of trace element partitioning, yet the element abundances and ratios in the fluid phase remained poorly constrained as the estimates on fluid composition were based on quenched solute analysis. Later on, with new developments in experimental and analytical approaches, the partition coefficients of trace elements have been determined in a similar MOR basalt – H₂O system at 700–800 °C and 4 GPa (Kessel et al., 2005a), using diamond traps to sample the fluid phase at high P and T (Kessel et al., 2004; Aerts et al., 2010). Additional element parti-

tioning experiments in a metasediment – H₂O system at 600–750 °C and 2.2 GPa have been performed using the synthetic fluid inclusion (SFI) technique (Spandler et al., 2007). The three studies revealed common enrichment in large ion lithophile elements (LILE) relative to high field strength elements (HFSE), although the ratios of other elements, e.g. LREE/HREE, were most likely controlled by the mineralogy of the residue. For example, the partitioning of REE in the study of Green and Adam (2003) was controlled by residual garnet and lawsonite; a steep LREE/HREE pattern in the experiments of Kessel et al. (2005a) displayed a characteristic signature of residual garnet whereas in the study of Spandler et al. (2007), garnet and epidote were the main hosts for REE. In the latter study, it was also pointed out that the overall fluid composition in equilibrium with the rock is rather diluted (~5 wt.% of total solute), suggesting that aqueous fluids may be unable to efficiently transport trace elements to the mantle wedge.

Considering the above mentioned studies on aqueous fluid–rock equilibria, it is apparent that the bulk rock composition, the T – P conditions and the corresponding mineral assemblages exert a major control on trace element partitioning, as observed for hydrous melts. However, unlike in the case of predominantly siliceous slab melts, the chemistry of aqueous fluids may induce an additional effect on trace element partitioning. A number of studies performed on relatively simple systems, consisting of a single mineral phase (i.e. quartz, rutile, zircon, apatite, monazite, xenotime, clinopyroxene) and water indicate a prominent effect of salt (e.g. NaCl, NaF) on element solubilities and partitioning (Brenan et al., 1994; Keppler, 1996; Newton and Manning, 2000; Antignano and Manning, 2008a; Rapp et al., 2010; Tropper et al., 2011, 2013; Bernini et al., 2013; Tsay et al., 2014). Moreover, as the concentration of major elements increase in the fluid with temperature, the polymerization of dissolved alkali (alumino)silicate components significantly promotes the solubilities of certain trace elements, e.g. HFSE *via* the formation of stable HFSE-O-Si(Al)/Na complexes (Audetat and Keppler, 2005; Antignano and Manning, 2008b; Hayden and Manning, 2011; Wilke et al., 2012; Louvel et al., 2013). It is, therefore, crucial to investigate the effect of ligands on fluid–rock equilibrium in complex – geologically relevant mineral assemblages.

As mentioned above, different mineral phases, such as garnet, lawsonite and epidote/zoisite, may control REE partitioning and fractionation during slab dehydration processes, depending on P – T conditions and bulk solid composition. Another potentially important host for REE in subducting oceanic crust is allanite, in which LREE represent major constituents. Allanite – bearing eclogites have been found in a number of locations related to previously subducted high – P and ultrahigh – P complexes, e.g. Dora-Maira, Lanzo and Mt. Mucrone massifs in western Alps, Caledonian orogen in western Norway, Dabie orogen, Sulu and Tianshan terranes in China, UHP units in western Himalaya and Mexican subduction zone (Boundy et al., 2002; Rubatto et al., 2008; Fulai et al., 2009; Cenko-Tok et al., 2011; Gomez-Tuena et al., 2011; Zhou et al., 2011; Lu et al., 2012; Gao et al., 2013; St-Onge

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