



Lutetium incorporation in magmas at depth: Changes in melt local environment and the influence on partitioning behaviour



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ARTICLE INFO

Article history:

Received 26 June 2016

Received in revised form 16 December 2016

Accepted 9 February 2017

Available online 1 March 2017

Editor: J. Brodholt

Keywords:

x-ray diffraction

high pressure

rare earth elements

silicate melts

ABSTRACT

The structure of two Lu doped (4000 ppm) model end member silicate liquids, a highly polymerised haplogranite (Si–Al–Na–K–O) and a less polymerised anorthite–diopside (Si–Al–Mg–Ca–O), have been studied up to 8 GPa using *in situ* x-ray diffraction techniques. The results are the first to identify trace rare Earth element incorporation in silicate melts at high pressure. At pressures below 5 GPa, the bonding environment of Lu–O was found to be dependent on composition with coordination number $CN_{Lu-O} = 8$ and bond distance $r_{Lu-O} = 2.36 \text{ \AA}$ in the haplogranite melt, decreasing to $CN_{Lu-O} = 6$ and $r_{Lu-O} = 2.29 \text{ \AA}$ in the anorthite–diopside melt. This compositional variance in coordination number at low pressure is consistent with observations made for Y–O in glasses at ambient conditions and is coincident with a dramatic increase in the partition coefficients previously observed for rare Earth elements with increasing melt polymerisation. With increasing pressure we find that CN_{Lu-O} and r_{Lu-O} remain constant in the haplogranite melt. However, an abrupt change in both Lu–O coordination and bond distance is observed at 5 GPa in the anorthite–diopside melt, with CN_{Lu-O} increasing from 6 to 8-fold and r_{Lu-O} from 2.29 to 2.39 Å. This occurs over a similar pressure range where a change in the *P*-dependence in the reported rare Earth element partition coefficients is observed for garnet–, clinopyroxene–, and olivine–melt systems. This work shows that standard models for predicting trace elements at depth must incorporate the effect of pressure-induced structural transformations in the melt in order to realistically predict partitioning behaviour.

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1. Introduction

Trace elements are highly fractionated during planetary differentiation events, because of differential partitioning between minerals, melts and metals at high pressures (*P*) and temperatures (*T*). The resulting partition coefficients are used as key indicators of depth, temperature, composition and potential age of melting events since planetary formation (McIntire, 1963). Trace element partitioning behaviour at mantle pressures is of particular interest for dating early crust formation on Earth and the Lu/Hf isotope system is regularly used to constrain early Earth melting processes (Guitreau et al., 2012). Many studies have shown that variables such as composition, *P*, *T* and oxygen fugacity have a control on the mineral/melt partitioning of trace elements (see

Wood and Blundy, 2013 and references within). Although it has long been recognised that crystal chemistry and composition play a major role in controlling how elements partition (Blundy and Wood, 1994), fewer studies have focused on the influence of the melt structure due to the lack of systematic experimental data (Corgne et al., 2012).

Variations in the melt however do influence partitioning significantly, as first reported by several compositional studies, e.g. olivine/melt Ni partitioning (Hart and Davis, 1978), rare earth elements (REEs) partitioning between granitic and gabbroic melts (Ryerson and Hess, 1978; Schmidt et al., 2006), REE clinopyroxene/melt partitioning (Gaetani, 2004), titanite/melt trace element partitioning (Prowatke and Klemme, 2005), REE forsterite/melt partitioning (Evans et al., 2008). As the polymerisation of the melt was increased, a two orders of magnitude increase in partition coefficient for trivalent rare earth elements such as Lu, Y, La, is observed, with high field strength elements (HFSE) such as Hf and Zr unaffected (Prowatke and Klemme, 2005; Schmidt et al., 2006).

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Prowatke and Klemme (2005) concluded that the polymerisation of the melt reduces the number of non-bridging oxygens available to bond with the trivalent cations, resulting in a preference for REEs to enter less polymerised melt structures. This was confirmed by a molecular dynamic study of Y in silicate melts (Haigis et al., 2013), that demonstrated the greater affinity of Y for Ca than for Si or Al in depolymerised melts, due to the weaker nature of the Ca–O bond compared to the Si(Al)–O bonds, resulting in more non-bridging oxygens being effectively available for Y. If pressure also has a significant effect on the melt structure and influences the compatibility of elements, then our current perception of how these element pairs behave at the conditions of melting may be altered.

To predict partitioning ratios, current models are based on experimental data for partition coefficients measured within individual systems and extrapolated for different size and charge of cations. One model that is frequently used to predict partitioning ratios is the ‘lattice strain model’ (Blundy and Wood, 1994). This incorporates the effect of ionic radius, charge and cation size in a model that links the parabolic relation between the partition coefficient and ionic radius with the size of the ion and Young’s modulus or elasticity of the lattice site. No explicit term for the effect of melt structure is included as it is assumed to be negligible and can be summarised within the initial measured partitioning ratio D_0 , which normalises the partition coefficient of a chosen cation with a specific melt composition.

Amongst the few existing studies on REE partitioning with pressure, only the garnet/melt, clinopyroxene/melt and olivine/melt systems have been studied over a P -range large enough to cover most terrestrial magmatic processes i.e. up to 5–6 GPa, other studies being restricted to below 3.5 GPa. The garnet/melt studies show a change in the P -dependence of mineral/melt partitioning, with an initially large decrease in partition coefficient and a compatibility plateau after 5 GPa (Fig. 1). For Hf and Zr this trend was not observed and they were largely unaffected. This would suggest that at $P > 5$ GPa partition coefficients between REE and 4+ cations may become similar. The transition to majorite may be the cause of this change in REE partitioning behaviour (Draper et al., 2003), however, it does so at the cost of reduced site elasticity and cation size which is incompatible with mineral physics rules. To solve inconsistencies in apparent site elasticity trends, van Westrenen and Draper (2007) introduced corrections for T , including an empirical correction for site elasticity by fitting a larger set of garnet/melt partitioning data. Under increased T , $D^{\text{mineral/melt}}$ is expected to get closer to unity due to the induced thermal disorder on crystal lattice sites. This better parameterisation of the lattice strain model clearly improved its robustness, although site elasticity values outside the 500–600 GPa range still cannot be reproduced. Besides, later datasets showed the strongest P -change of $D_{\text{Lu}}^{\text{garnet/melt}}$ between 3 and 8 GPa (Fig. 1), where data were collected over a 100 K T -range only (Suzuki et al., 2012). Datasets collected on larger P -ranges (up to 17 GPa for Corgne et al., 2012 and 20 GPa for Suzuki et al., 2012 vs up to 10 GPa for van Westrenen and Draper, 2007) point out a worsening of the predictive value of the model at higher pressures. Clinopyroxene/melt partitioning studies at P up to 3.5 GPa have been parameterised by Wood and Blundy (1997) and subsequent papers. The exploration of a larger compositional space at 1.5 GPa demonstrated the dependence of $D_{\text{Lu}}^{\text{CPx/melt}}$ on melt polymerisation rather than crystal composition (Gaetani, 2004), an effect restrained to compositions close to the lherzolite solidus. Higher P data collected up to 12 GPa (Suzuki et al., 2012) show an overall similar P -trend to the garnet/melt system (Fig. 1). In the case of olivine/melt data, the REE partition coefficients decrease with increasing silica in the melt at room P (Evans et al., 2008). This is at odds with the main stream of studies showing the opposite behaviour

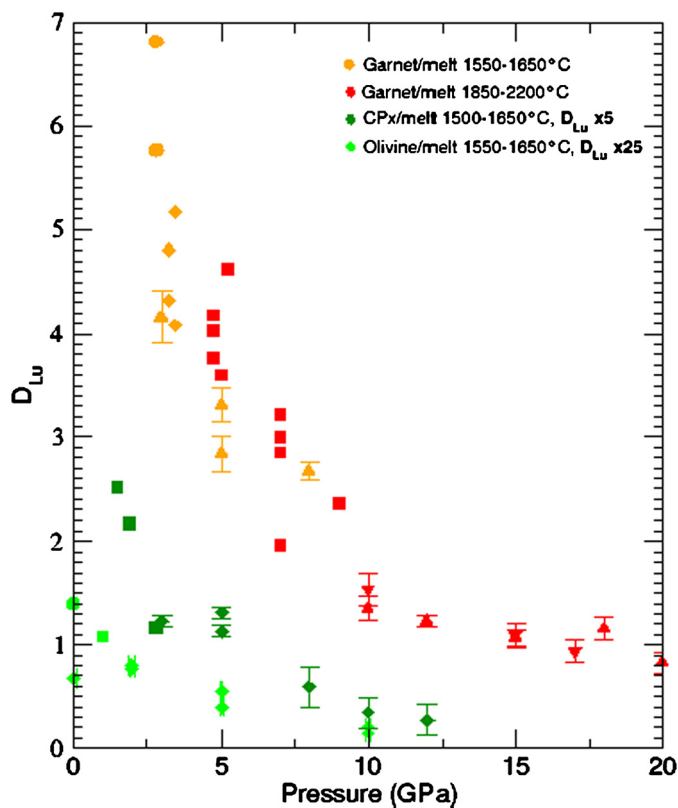


Fig. 1. Mineral–melt partition coefficient data ($D_{\text{Lu}}^{\text{min/melt}}$) for Lu with pressure. For the sake of inter-comparison, $D_{\text{Lu}}^{\text{CPx/melt}}$ have been multiplied by 5, and $D_{\text{Lu}}^{\text{Olivine/melt}}$ multiplied by 25. Data presented are from studies where the composition was consistent over all P - T conditions, i.e. basaltic composition for garnet and clinopyroxene studies, and peridotitic composition for olivine studies. Other studies (i.e. van Westrenen et al., 1999) have varying starting compositions that may mask any P effects; similarly, datasets have been limited to restricted T -range for Gt/melt and CPx/melt studies, which was not possible for olivine/melt due to the small number of studies available at high P . Symbols represent various studies; for Gt/melt (circles: Salters and Longhi, 1999, diamonds: Salters et al., 2002, squares: Draper et al., 2003, up triangles: Suzuki et al., 2012, down triangles: Corgne et al., 2012), for CPx/melt (squares: Salters and Longhi, 1999, diamonds: Suzuki et al., 2012), for Ol/melt (circle: Evans et al., 2008, squares: Salters et al., 2002, diamonds: Imai et al., 2012). Error bars are taken from references where possible and for those not shown are smaller than the symbol size.

(Hart and Davis, 1978; Ryerson and Hess, 1978; Prowatke and Klemme, 2005; Schmidt et al., 2006), and might be taken with caution as the authors pointed out the poor fit statistics (Evans et al., 2008). Under high P , REE partition coefficients between olivine and peridotitic melt for 3+ cations decrease with increasing pressure between 2 and 10 GPa (Imai et al., 2012). As for garnet, the apparent site elasticity behaves anomalously with P , decreasing by 200 GPa of which the T -effect can only explain a maximum of 33 GPa. Instead, Imai et al. (2012) proposed that elasticity of the cation site in the melt affects partitioning as changes on the crystal lattice alone would not produce a large enough effect. It is currently challenging to test this hypothesis without a proper understanding of site elasticity in melts. It is nonetheless true that the large compressibility difference between melts and crystals that exists at ambient pressure considerably diminishes with pressure (see Sanloup, 2016 for a review and references therein) as the melt becomes much harder to compress. Consequently, it is expected that the strain energy resulting from the insertion of REE elements in the melt ceases to be negligible. However, the fine P -mesh available for the Gt/melt system implies that the strong change in P -dependence of $D_{\text{Lu}}^{\text{Gt/melt}}$ excludes a purely gradual transformation, an effect that could be artificially smoothed for CPx/melt, Olivine/melt systems due to the insufficient mapping

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