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## Letters

## Experimental study of the partitioning of Cu during partial melting of Earth's mantle

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## ABSTRACT

Primitive basaltic glasses from mid-ocean ridges (MORB), ocean islands (OIB) and arcs contain three to five times the Cu as the currently accepted primitive upper mantle (PUM) value, suggesting a bulk partition coefficient  $D_{\text{Cu}}^{\text{mantle/melt}}$  of  $\sim 0.20$ . Sulfide, with a  $D_{\text{Cu}}^{\text{sulfide/melt}}$  of 250–960 is presumed to be ubiquitous in the mantle. The Cu abundances in basalts are unable to be achieved if Cu behaves compatibly, or if a high enough modal abundance of sulfide exists during the onset of melting to sequester significant Cu. To address this conundrum we determined  $D_{\text{Cu}}$  in olivine and orthopyroxene at 1250–1525 °C and 1.0 GPa in a hydrous basalt and KLB1 peridotite, at  $fO_2$  of FMQ-1, near to melting conditions of the upper mantle. The measured  $D_{\text{Cu}}^{\text{ol/liq}}$  of 0.06–0.21 and  $D_{\text{Cu}}^{\text{px/liq}}$  0.15–0.82 do not vary with melt fraction, or significantly with  $fO_2$ , and can be combined with estimates for  $D_{\text{Cu}}$  for clinopyroxene into melting models to examine the Cu contents of mantle-derived melts. The Cu abundances for MORB, OIB, and arc glasses are all explicable by up to 15% melting of the silicate—only portion of the mantle in which Cu behaves as a mildly incompatible element ( $D_{\text{Cu}}^{\text{mantle/melt}} \sim 0.26$ ). For Cu to be enriched in basaltic melts in the presence of sulfide, the melt/sulfide ratio, or the oxidation state of the mantle during melting, must be significantly high to diminish the potential of sulfide to sequester any Cu. Our results suggest the role of sulfide during mantle melting may be insignificant with regards to controlling the concentration of moderately chalcophile elements like Cu, and confirm that the previously estimated Cu content for PUM of 20 ppm is correct.

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## 1. Introduction

Over 60% of total global Cu production is attributed to porphyry Cu deposits (Sinclair, 2007). Porphyry Cu deposits are large, low-grade occurrences of Cu that form as mantle-derived magmas ascend into the upper crust above subduction zones, fractionate, and release metal-rich fluids into the surrounding rocks. The quantity and grade of porphyry Cu deposits depend on many factors and processes operating in the arc system from source to sink. Most studies on the origin of Cu porphyry deposits have focused on the behavior of Cu at shallow depths ( $< 3$  km), whereas processes at deeper levels in the mantle, where parental arc magmas form, are understudied (Mungall, 2002; Mungall et al., 2006).

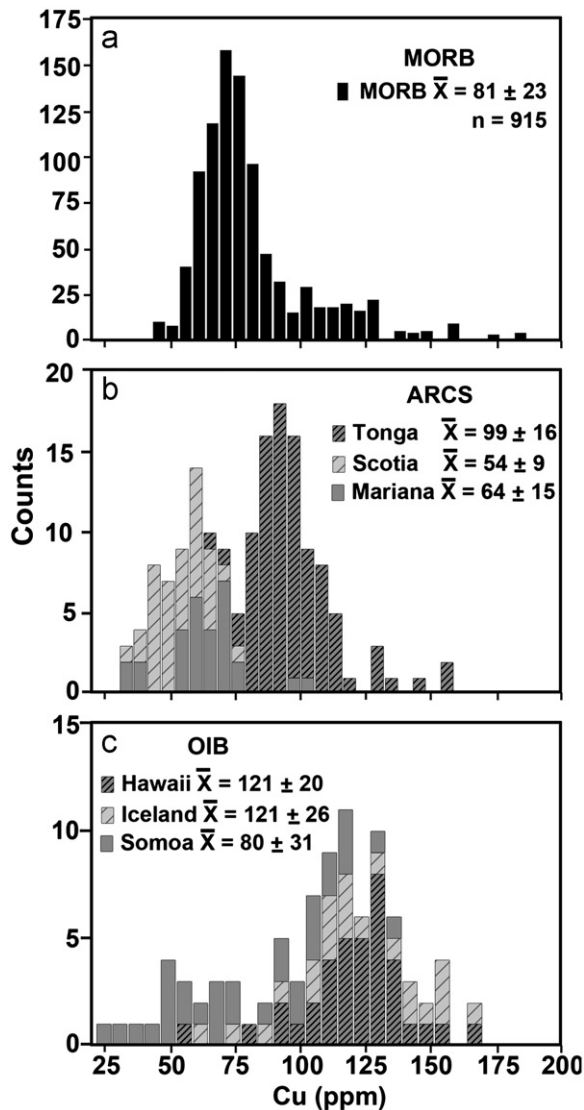
Copper has an estimated concentration of between 20 and 30 ppm in the primitive upper mantle (PUM) (O'Neill, 1991; Sun, 1982). The mean concentrations of Cu in primitive basaltic glasses (filtered to have MgO  $> 6\%$  and SiO<sub>2</sub> 48–52%) vary with tectonic setting (Fig. 1). MORB has Cu concentrations averaging

$81 \pm 23$  ppm, whereas arcs show a wide range both within and among individual arcs ( $99 \pm 16$ ,  $64 \pm 15$ , and  $54 \pm 9$  ppm in the Tonga, Marianas, and Scotia arcs, respectively). Amongst OIB samples, the Cu in Hawaiian and Icelandic basaltic glasses are highest of all settings ( $121 \pm 20$  and  $121 \pm 16$  ppm respectively) whereas Samoan glasses have a mean of  $80 \pm 31$  ppm and a wider range. The variation in Cu concentrations in these different settings may reflect differences in mantle source endowment, depth of formation,  $fO_2$ , or sulfide content. In MORB, arc, and OIB settings the highest Cu abundances occur in the most primitive basaltic glasses with MgO  $> 8\%$  (Fig. 2). The mean of Cu for the most primitive basalts is different for each setting, with the highest Cu mean in Hawaiian OIB and the lowest in MORB. As the melt decreases in MgO along the liquid line of descent, Cu concentrations decrease slightly in each of the three settings.

The data shown in Fig. 1 imply that Cu is a mildly incompatible element during mantle melting, with a  $D_{\text{Cu}}^{\text{mantle/melt}}$  of  $\sim 0.20$ . This simple observation raises a conundrum for the budget of Cu during mantle melting when sulfide, a ubiquitous accessory phase in the mantle, is considered. For example, PUM contains  $\sim 250$  ppm S (0.0065%) as a trace liquid sulfide (Fe–S–O) phase in the mantle (Rehkamper et al., 1999), and sulfide/silicate melt  $D_{\text{Cu}}$  values range from 250 to 960 (Gaetani and Grove, 1997).

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**Fig. 1.** Compilation of Cu abundances in basaltic glasses from various geologic settings: (a) MORB ( $n=915$ ); (b) OIB: Hawaii ( $n=37$ ), Iceland ( $n=22$ ), and Somoa ( $n=31$ ); (c) Arcs: Tonga ( $n=92$ ), Mariana ( $n=38$ ), and Scotia ( $n=29$ ). MORB data from Jenner and O'Neill (2012), Batiza et al. (1992), Castillo et al. (2000), Ferguson and Klein (1993), Hekinian and Walker (1987), Karsten et al. (1996), Klein and Karsten (1995), Langmuir (1999), Perfit et al. (1996), Pyle (1994), Reynolds (1995), Reynolds and Langmuir (1997) and Thompson et al. (1985); Hawaiian data from Dzurisin et al. (1995), Norman et al. (2004), Pitcher et al. (2009), Poustovetov and Roeder (2000), Wright and Helz (1996); Icelandic data from Breddam (2002), Condomines et al. (1983), Hansen and Gronvold (2000), MacLennan et al. (2001), Sigmarsson et al. (2000) and Steinthorsson et al. (2000); Somoan data from Workman et al. (2004); Tongan Arc references: Bezos et al. (2009), Dril et al. (1997), Falloon et al. (1992), Pearce et al. (1994); Mariana data from Pearce et al. (2005); Scotia data from Barry et al. (2006) and Fretzdorff et al. (2002).

If sulfide is a residual phase until  $\sim 25\%$  melting (Hamlyn et al., 1985) and the modal abundance of sulfide is greater than 0.001–0.004%, Cu would be sequestered during melting, in stark contrast to the elevated concentrations in basaltic glasses, most of which form by 5–20% melting (Gaetani et al., 1993; Hamlyn et al., 1985; Kinzler and Grove, 1992; Klein and Langmuir, 1987; McKenzie and Bickle, 1988; McKenzie and O'Nions, 1995; Sisson and Grove, 1993). This conundrum can be explained by two scenarios. In the first scenario Cu could behave as a mildly incompatible element (low Bulk  $D_{Cu}$ ) due to small  $D_{Cu}$  for silicate phases and a modal abundance of sulfide that is insufficient to sequester significant Cu. In this way, sulfide does not play any role in moderately

chalcophile element abundances (Mungall et al., 2006). In the second scenario Cu behaves compatibly, but the currently accepted value of 20 ppm Cu in PUM is far too low, as has been proposed by Kamenetsky and Eggins (2011) to explain the negative correlation of Cu with La/Sm in primitive ophiolite basaltic glasses. Currently, there is a dearth of experimental measurements on the partitioning of Cu during mantle melting to test either scenario. The few extant  $D_{Cu}$  values for the key mantle minerals olivine, orthopyroxene, clinopyroxene, and spinel are internally inconsistent and vary by several orders of magnitude (Fig. 3). Better experimental data are required to understand the behavior of Cu during mantle melting. To this end, we determined the mineral/melt partitioning of Cu in olivine and orthopyroxene at high temperatures (1250–1525 °C), at a fixed pressure of 1.0 GPa, at  $fO_2$  near to melting conditions of the upper mantle. The new experimental results are incorporated into a mantle melting model and compared with Cu concentrations in basaltic glass from MORB, OIB, and arcs.

## 2. Methods

A synthetic hydrous basalt (GSB) and natural peridotite (KLB-1) were used as starting compositions (Table 1) because they saturate with orthopyroxene and/or olivine over a large span of pressures, temperatures and degrees of melting (Grove et al., 2003; Hirose and Kushiro, 1993; Takahashi, 1986; Takahashi et al., 1993). The GSB composition was synthesized using reagent grade  $SiO_2$ ,  $TiO_2$ ,  $Fe_2O_3$ ,  $Na_2CO_3$ , and  $K_2CO_3$ . Gibbsite ( $Al(OH)_3$ ), portlandite ( $Ca(OH)_2$ ), and brucite ( $Mg(OH)_2$ ) were added as sources of  $H_2O$  to a total of 14.25% of which 4.4% is estimated to be dissolved in the glass based on the missing percentage from the EMP total in Table 1. The oxide mixture was shaken in a plastic canister for 15 min and ground under ethanol in an agate mortar for 15 min. This process was repeated three times. The GSB composition was doped to  $> 1000$  ppm with V, Cu, Mo and Re (Table 1) as reagent grade  $V_2O_5$ , CuO, and  $MoO_3$ . Rhenium was added by micropipette as a NIST certified solution, and dried down. The powder was again mixed in a plastic canister for 15 min and ground in an agate mortar for 15 min, and this process repeated three times to homogenize the trace elements into the composition. Natural powder of KLB-1 peridotite was ground under ethanol for 15 min, then doped with reagent grade CuO,  $V_2O_5$ ,  $MoO_3$ , and  $Sc_2O_3$  (Table 1) and homogenized using the same methods described above for GSB. Starting materials were doped with high enough concentrations of Cu to be analytically detectable in all phases, and to obviate any Cu loss from the starting material during the experiment (described in detail below).

Four capsule configurations were used for the experiments: Pt, graphite-lined Pt, Re and Re-lined Pt. For the Pt capsule experiments, the starting powder was packed into a 3 mm OD  $\times$  0.127 mm wall thickness Pt capsule, then sealed using an arc welder. Starting powder for the graphite-lined experiments was first packed into 3.8 mm  $\times$  1.8 mm graphite capsules, then placed into a 4 mm OD  $\times$  0.127 mm wall thickness Pt capsule and sealed by arc welding. Capsules of Re were constructed from Re foil wrapped around a 2 mm rod and then folded and crimped on one end. Starting powder was then packed into the capsule and the remaining end was folded, crimped tight and sealed in a die. For Re-lined experiments a Re capsule prepared as above was then inserted into a 3 mm OD  $\times$  0.127 mm wall thickness Pt capsule, and then sealed using an arc welder. In some experiments, a 1.5 mm length of 30  $\mu$ m diameter Pt wire was packed with the starting material to monitor  $fO_2$  during the experiment after the method of Rubie et al. (1993).

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