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Amorphous Materials: Properties, Structure, and Durability†

Arsenic enrichment in hydrous peraluminous melts: Insights from femtosecond laser ablation-inductively coupled plasma-quadrupole mass spectrometry, and in situ X-ray absorption fine structure spectroscopy

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Abstract

Despite the ubiquity of arsenic in hydrothermal-magmatic environments, its abundance, distribution, and chemical and structural status in natural silicate melts and glasses remain poorly known. Here we report the first in situ measurements of the redox state and molecular structure of As, using X-ray absorption fine structure (XAFS) spectroscopy, in a rhyolitic peraluminous glass from Macusani (SE Peru) that is representative of anatectic melts derived from metasedimentary crustal protoliths. Arsenic abundances as well as the concentrations of other trace elements were measured in the glass using a femtosecond laser ablation-inductively coupled plasma-quadrupole mass spectrometry (LA-ICP-QMS). The glass shows enrichments, by factors of 10 to 100, in comparison with the mean continental crust values, for As and other incompatible trace elements (e.g., Be, B, Rb, Sn, Sb, and Ta), and by factors of 100 to 200 for Li, Cd, and Cs. Arsenic is present in the peraluminous glass in trivalent state in the form of oxy-hydroxide complexes like $AsO(OH)_2^-$ and/or $As(OH)_3$, similar to those dominant in the aqueous fluid vapor phases at hydrothermal-magmatic conditions. The similar As chemical speciation between the fluid and the melt is consistent with As fluid/melt partitioning coefficients close to one, as observed in experiments on rhyolite-water systems. The depolymerized melt structure caused by elevated H_2O, F, and P contents is likely to allow accomodation of high concentrations of metalloid hydroxide/hydrated complexes. Consequently, hydrous silicate melts may be important transporting media in shallow magmatic-hydrothermal settings for As and similar elements like B and Sb due to their high affinity to water and hydroxide ligands.

Keywords: Arsenic, macusanite, XAFS spectroscopy, femtosecond LA-ICP-MS, fluid-melt partition coefficient, arsenic hydroxide complex