

Complexation of Sr in aqueous solutions equilibrated with silicate melts: Implications for fluid-melt partitioning

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Fluid-melt partitioning of Sr, Ba, La, and Y strongly depends on fluid and melt composition [1-2]. That is, partition coefficients of these elements increase with i) salinity of the fluid and ii) increasing alumina saturation index (ASI: $Al/(Na+K)$ in moles) of the silicate melt (Fig.1).

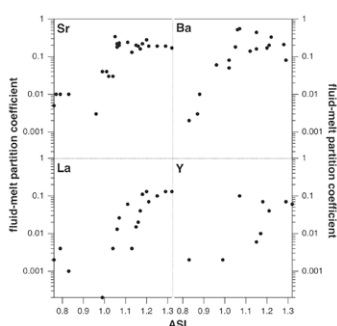


Fig. 1. Fluid-melt partition coefficients of Sr, Ba, La and Y as function of melt composition at 750°C, 200 MPa, and using a chloridic fluid.

On one hand, the distinct affinity for chloridic fluids points to a preferential complexation of these elements with chlorine. On the other hand, the strong dependence on melt composition suggests that a different release of melt components into the aqueous fluid in peralkaline systems changes the speciation in the aqueous fluid. Up to now, the exact nature of the complexation in the fluid is not clear.

Here, we present experimental and theoretical data on the complexation of Sr in three fluid-melt systems. X-ray absorption spectroscopy was applied to probe the local environment around Sr in silicate-bearing fluids, i.e., peralkaline or peraluminous melts completely dissolved in the aqueous fluid, and in various model systems, e.g., crystalline compounds, silicate glasses, and aqueous solutions. In order to decode the complexation of Sr in model systems and silicate-bearing fluids, theoretical spectra are calculated either using model structures, by testing several small cluster geometries or based on snapshots of trajectories produced with MD simulation to account for atom movements in aqueous solutions. Experimental data clearly show differences in the local environment around Sr in studied silicate-bearing fluids. Theoretical spectra point to a predominant formation of i) SrO_6 cluster in chlorine-free peralkaline fluids, ii) distorted SrO_6 clusters in chloridic-peralkaline fluids, and iii) complexes that very likely incorporate Cl in chloridic-peraluminous fluids.

[1] Borchert *et al.* (2010a), *GCA* **74**, 1057-1076. [2] Borchert *et al.* (2010b), *Chem. Geol.* **276**, 225-240.

Formation of arsenic bearing apatite from calcite. Chemistry and microstructures

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Apatite, a calcium phosphate mineral, can form naturally as solid solutions with between 0-100% of the phosphate anions substituted with compatible molecules in the crystal lattice [1]. Arsenate, $AsVO_4^{3-}$ is one such molecule, and there are many examples in nature of high As bearing apatite [2].

In order to track the chemistry of ancient fluids that formed the observed composition variation in apatite and related mineral deposits, a series of hydrothermal synthesis experiments were performed, aiming to establish a relationship between apatite composition and fluid chemistry. Utilising a general method from a previous study [3], we have generated a range of synthesised apatite samples starting from calcite, primarily by reaction with phosphate (PO_4^{3-}), substituting with similar molecules (e.g. AsO_4^{3-} , SO_4^{2-}). Chemical analysis of thus formed apatite-like compounds show differing level of incorporation depending on the molecule being substituted, while SEM and CL mapping of grains show very interesting and complex textures and element distribution patterns. Two stage experiments, where one solution composition is reacted with pure calcite, then a different solution is reacted with the product, show that phosphate is an important driver for As-bearing apatite growth, and that arsenic incorporated into the apatite structure is not readily remobilised. This level of understanding is important when considering natural apatite samples as indicators to track ore fluids.

[1] Pan and Fleet (2002) *Rev. Mineralogy* **48**, 13-49. [2] Mailloux *et al.* (2009) *Appl. Environ. Microbiol* **75**, **8**, 2558-2565. [3] Kasiopas *et al.* (2011) *Geochimica et Cosmochimica Acta* **75**, **12**, 3486-3500.