



Complexation of Sr in aqueous fluids equilibrated with silicate melts: effect of melt and fluid composition

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At crustal conditions, the fluid-melt partitioning of Sr is mainly controlled by the salinity of the fluid and the composition of the melt (Borchert et al., 2010). The data show a sharp increase in the Sr partition coefficient with the alumina saturation index (ASI) to a maximum of 0.3 at an ASI of 1.05. Because fluid-melt partitioning of a given element depends on its complexation in the fluid and its incorporation in the melt, these data imply a change in the Sr speciation at least one of the two phases. For silicate melts, Kohn et al. (1990) found only small changes in the first coordination shell of Sr in a suite of melts with various degrees of polymerization, and argued that incorporation of Sr in the melt should not play a major role in controlling Sr partitioning. For the aqueous fluid, Bai and Koster van Groos (1999) and Webster et al. (1989) suggested a control of the Sr partition coefficient by SrCl_2 complexes based on the correlation between partition coefficient and Cl concentration in the fluid after quenching. Both hypotheses cannot explain our partitioning data. Thus, new information on Sr complexation is required. Here, we studied the complexation of Sr in peraluminous or peralkaline melt dissolved in aqueous fluids in-situ at elevated PT conditions using hydrothermal diamond-anvil cells (HDAC) and X-ray absorption near edge structure (XANES) spectroscopy. The starting materials were peraluminous or peralkaline glass and H_2O or a chloridic solution. The glass was doped with high concentrations of 5000 or 10000 ppm Sr. We used bulk compositions with 10 to 15 wt.% glass to ensure that the melt was completely dissolved in the fluid at high PT conditions. For qualitative evaluation, we analyzed the starting glasses and various crystalline compounds and standard solutions. The experiments were performed at beamline ID26 at ESRF (Grenoble, France) using a high resolution emission spectrometer and Si(311) monochromator crystals for high resolution and Si(111) for low resolution measurements. An overall resolution of 1.98 eV (Si(311)) and 3 eV (Si(111)) was achieved using a beamsize of 120x400 μm , Si(777) analyzer crystals and a Rowland circle diameter of 1 m. Strontium XANES spectra of peraluminous and peralkaline starting glasses show distinct differences in pre-edge, main edge and position of the first EXAFS maximum which is directly correlated to the distance of Sr and its nearest neighbor. Therefore, the spectra indicate an effect of the ASI on the local structure around Sr. Spectra of standard solution at various PT conditions indicate detectable temperature-dependent changes in the intensity and peak width of the white line. The XANES spectra of chloridic solutions with peraluminous or peralkaline melt dissolved differ significantly from each other. The latter are similar to the one of the peralkaline starting glass, while this is not the case for solutions with peraluminous melt. The spectra of water with dissolved peralkaline melt distinctly differ from those using chloridic fluids. In conclusion, the first spectroscopic evidence on Sr complexation at elevated PT condition indicates significant speciation changes in both fluid and melt, and suggest ASI-dependent formation of Sr-Si complexes in the fluids.

Literature

Bai and Koster van Groos (1999), GCA 63, 1117-1131.
Borchert et al. (2010), GCA 74, 1057-1076.
Kohn et al. (1990), CMP 105, 359-368.
Webster et al. (1989), Econ. Geol. 84, 116-134.