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In-situ XANES study on Fe, U & Th in hydrous melts

M. Wilke (1), C. Schmidt (2), M. Borchert (1), F. Farges (3), A. Simionovici (4) (1) Institut f. Geowissenschaften, Universität Potsdam, D-14415 Potsdam (2) GeoForschungsZentrum Potsdam, D-14473 Potsdam (3) Laboratoire des Géomateriaux, Université Marne-la-vallee, F-77454 Marne la Vallée cedex 2 (4) Laboratoire des Sciences de la Terre, ENS Lyon, F-69007 Lyon (max@geo.uni-potsdam.de)

Insight to the structural units of melts is an important key to model properties of magmas. The effect of water and pressure on the local structure around minor to trace elements in silicate melts was investigated at in-situ conditions by means of XANES spectroscopy using a diamond anvil-cell. This was done to avoid spurious effects invoked by quenching [1] and to understand better the processes occurring during the quench. We present results of comparative in-situ XANES measurements of compatible and incompatible elements such as iron, uranium and thorium in hydrous silicate melt up to 1 GPa and 700°C. This study was conducted to reveal the effect of water and pressure on the local structure of Fe, Th and U in silicate melts. In-situ XANES spectra were recorded at the ESRF (Grenoble France), beamline ID 22, using a hydrothermal diamond anvil cell with a design optimized for such measurements [2], i.e. recesses on the front and the back-side of one of the diamond anvils that provide the possibility to collect spectra at relatively low energies (down to 7 keV) and relatively low concentrations (0.1-1 wt%). In-situ Fe K-edge XANES spectra of Fe(II) in hydrous haplogranitc melt at 700°C and 500 MPa suggests that the local structure around Fe in hydrous glass observed previously is probably due to ordering during the quench. Additionally, the XANES is very similar to in-situ spectra taken on Fe(II) in anhydrous haplogranitic melt at 1150°C and ambient pressure. This indicates that the combined effect of water and pressure (0-500 MPa range) does not influence drastically the local structure of Fe in this type of melt composition. In-situ LIII-edge XANES of U in hydrous haplogranitic melt (1 wt% U) at 700°C and 620 MPa show that, upon reduction, U precipitated as uraninite. This suggests a low amount of NBO's (to which tetravalent actinides preferentially bond [3]) in this water-saturated melt. In contrast, U-bearing (1000 ppm) hydrous sodium-tri-silicate melt shows the presence of U(IV) dissolved in the melt as 6-7 coordinated species, as in dry glasses [3]. Similar structural information is obtained for Th(IV). Spectra taken above and below the complete miscibility of the silicate and aqueous phase (ca. 460°C) also reveal no difference in speciation. The aqueous fluid measured at ambient conditions after the run did not show any significant amount of dissolved tetravalent actinides [1] Wilke et al. (2002) Chem. Geol., 189, 55-67. [2] Schmidt C., Rickers K. (2003) Am. Mineral., 88, 288-292. [3] Farges F. (1991) Geochim. Cosmochim. Acta, 55, 3303-3319.