In situ observation of silicate speciation in liquids in the system K2Si4O9-H2O at high temperature and high pressure

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Knowledge of the speciation of silica structural units (Q unit speciation) in hydrous silicate liquids is of importance for understanding the mechanism of incorporation and transport of H2O in magmas. Although many experimental studies on the Q unit speciation in silicate glasses are available, recent advance in experimental techniques revealed that the structural relaxation of silicate liquids is so fast that the Q unit speciation is not quenchable from high temperatures and high pressures. This is particularly in the case for hydrous silicate liquids as the presence of H2O causes in a drastic decrease in the liquid-glass transition temperature. We present here results from in situ observation of the Q unit speciation in liquids in the system K2Si4O9-H2O at high temperatures and high pressures. Raman shift bands due to the fundamental Si-O stretch vibrations (700-1200 cm-1) and near infrared absorption bands due to the first overtone O-H stretch vibration (˜7000 cm-1) were investigated to 800 degree C and ˜1.4GPa for the liquids in the system, using an externally heated diamond anvil cell fitted to micro-Raman and micro-FTIR spectrometers. The relative abundance of each Q unit in the liquids was estimated based on the integrated intensity of respective Si-O stretch vibration band. The concentration of H2O (both OH groups and molecular H2O as H2O) was determined from the integrated absorbance intensity of the first overtone O-H stretch vibration band, provided that the integrated absorption coefficient is independent of temperature and pressure. Our results show that at compositions containing 35wt% and 15wt% of H2O in the system, only liquid phase is present at temperatures above 600 degree C and at pressures above ˜1.0GPa, but below this, crystalline quartz coexists. In the only liquid region, Q0 (˜780 cm-1) dominates by far any other structural units in the 35wt% H2O liquid, whereas Q3 (1050 cm-1) is major structural unit in the 15wt% H2O liquid. The Q unit speciation remains approximately unchanged at temperature between 600 and 800 degree C and at pressure between ˜1.0 and ˜1.4GPa. This leads to that the Q unit speciation in the liquids is mainly governed by the reaction that the more (and the most) depolymerized product is strongly favored by increasing activity of H2O component in the liquids.