

Determination of water speciation in hydrous Na₂Si₂O₅ melt at high temperature and high pressure

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Though in situ measurements of water speciation were widely conducted for rhyolitic melts and their analogues [e.g., 1, 2, 3], only limited data are available for depolymerized silicate melts. We performed in situ near-infrared and Raman spectroscopic measurements of Na₂Si₂O₅ (structural analogue of depolymerized, basaltic melt) with a fixed water content (5.9 wt%) in an externally heated diamond anvil cell (HDAC) [4]. Experiments were carried out at temperatures to 800 degree C and at pressures to 2 GPa. Temperature was controlled to plus-minus 1 degree C with chromel-alumel thermocouples in contact with the anvils near the sample chamber of the HDAC. Pressure inside the sample chamber was monitored with the pressure- and temperature-dependent Raman shift of the ¹³C diamond marker [5]. Only single, homogeneous liquid phase was stable at temperatures above 650 degree C and at pressures above 1.6 GPa; neither crystallization nor fluid exsolution was observed. At lower temperatures (hence at lower pressures) crystallization of solid Na₂Si₂O₅ was observed and conformed by Raman spectroscopy. Near-infrared spectra of the homogeneous liquid phase contain absorption peaks corresponding to both molecular H₂O (near 5200 cm⁻¹) and structurally bound OH groups (near 4500 cm⁻¹). Assuming a constant ratio of the molar absorptivities for these bands, OH/H₂O is estimated to be 0.9 in the quenched glass at ambient conditions and 1.9 in the homogeneous liquid phase at 800 degree C (both OH and molecular H₂O as H₂O, in wt%). Thus, one may expect that in the homogeneous liquid phase, the OH/H₂O increases as temperature increases. This trend is similar to those reported for highly polymerized silicate melts [1, 2].

References: [1] Sowerby J.R., Keppler H., *Am. Mineral.* 84, 1843 (1999); [2] Nowak M., Behrens H., *Earth. Planet. Sci. Lett.* 184, 515 (2001); [3] Shen A.H., Keppler H., *Am. Mineral.* 80, 1335 (1995); [4] Bassett W.A., Shen A.H., Bucknum M., Chou I.M., *Rev. Sci. Instr.* 64, 2340 (1993); [5] Mysen B.O., Yamashita S., *Geochim. Cosmochim. Acta.* 74, 4577 (2010).