

U004-P03

Room: Convention Hall

Time: May 24 17:15-18:45

Speciation of N-O-H volatiles in coexisting fluids and silicate melts determined in-situ under reduced conditions

Shigeru Yamashita^{1*}, Bjorn O. Mysen²

¹ISEI, Okayama University, ²Geophysical Laboratory, CIW

Speciation of N-O-H volatiles in fluids and silicate melts is important to understand mechanism of incorporation and transport of volatile components in magmas and Earth's interior. Experimental results from in-situ observation of the N-O-H speciation in coexisting fluids and silicate melts in the system $\text{Na}_2\text{Si}_4\text{O}_9$ (haploandesite)-N-O-H at pressures to 870 MPa and at temperatures to 800 degree C under reduced conditions are presented. Raman bands assigned to the fundamental N-N, N-H, O-H stretch vibrations (around 2300 to 3600 cm^{-1}) and near infrared band assigned to the first overtone of O-H stretch (around 7000 cm^{-1}) were investigated using an externally heated hydrothermal diamond anvil cell (HDAC) fitted to confocal micro-Raman and micro-FTIR spectrometers. The $f_{\text{H}_2}/f_{\text{H}_2\text{O}}$ ratio was controlled near 1 log unit above that of the iron-wustite buffer with the aid of the reaction $\text{Mo} + 2\text{H}_2\text{O} = \text{MoO}_2 + 2\text{H}_2$. Pressure in the HDAC was monitored with the pressure- and temperature-dependent Raman shift of synthetic ^{13}C diamond. This Raman shift was calibrated with the equation-of-state of pure H_2O (IAPWS-95) with ^{13}C and H_2O only loaded in the HDAC. Temperature was controlled to plus-minus 1 degree C. To ensure the highest precision of the Raman shift, the 585 nm Ne line was acquired simultaneously as reference. The estimated uncertainty in the pressure is plus-minus 100 MPa. In the silicate + N-O-H experiments, N-O-H fluids and silicate melts coexist above 400 degree C and 300 MPa. Below these conditions the fluid becomes a less dense vapor phase. The N-O-H speciation of fluid remains approximately unchanged at temperatures between 400 and 800 degree C and at pressures between 300 and 870 MPa. These N-bearing species exist in a variety of redox states, from molecular N_2 through NH_2 groups to molecular NH_3 . In melts on the other hand, the N-bearing species are dominantly molecular NH_3 . The average redox state of nitrogen in the melt is, therefore, more reducing than in coexisting fluid. Thus, the nitrogen solubility in silicate melts is likely greater the more reducing the conditions. The different redox states of nitrogen in fluids and melts may also affect nitrogen partitioning and nitrogen isotope fractionation behavior between fluids and melts.

Keywords: fluid, silicate melt, speciation, N-O-H volatile, in-situ observation