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Water speciation in rhyolitic melt determined by in-situ infrared spectroscopy

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ABSTRACT

Near-IR spectra obtained from hydrous silicate melts, close to natural rhyolite, were acquired at both high temperature using a heating stage and simultaneously at high pressure and temperature using a hydrothermal diamond cell. The temperature dependence of the extinction coefficients for the peaks due to OH and H_2O is negligible. In both sets of experiments, the speciation reaction $H_2O + O = 2OH$ is shifted to the right with increasing temperature above the glass transition but changes below this are negligible, within experimental uncertainty. For a sample containing 3.93 wt% total water, the temperature dependence of the speciation equilibrium can be described by two equations with temperature in K: $\ln k = -36.74/T - 4.02$ for the glass phase (giving enthalpy and entropy of reaction values of $\Delta H = 0.31$ kJ/mol and $\Delta S = -33.45$ J/mol-K), and $\ln k = -3821.83/T + 1.61$ for the melt phase (where $\Delta H = 31.77$ kJ/mol and $\Delta S = 13.41$ J/mol-K) respectively). The glass transition temperature of 670 K is defined by the intersection of these curves. Our values are in good agreement with previously published glass transition temperatures for similar compositions. Similar values for the enthalpy and entropy of reaction were obtained from all the other experiments including those at pressures up to 4.5, 5, and 10 kbars, suggesting that the speciation depends negligibly on pressure for this pressure range.