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AMORPHOUS MATERIALS: PROPERTIES, STRUCTURE, AND DURABILITY†

Quantitative Raman spectroscopy: Speciation of Na-silicate glasses and melts

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ABSTRACT

In situ, high-temperature Raman spectroscopy was used to study the Q^n speciation in binary Na-silicate glasses and melts. Over 300 Raman spectra in the compositional range from 25 to 40 mol% Na_2O were collected at room and high temperatures between 800 and 1200 K. Quantitative information on the relative abundances of species in melts was obtained from the Raman spectra through a quantification procedure that does not require any a priori assumptions about the line shapes or external calibration of the Raman scattering efficiencies for the various Q^n species. The $\Delta H°$ associated with the speciation reaction $2Q^3 = Q^4 + Q^2$ was found to be 20.3 ± 7.9 kJ/mol. For a given temperature, the speciation is more disordered in sodium than in potassium silicate melts. Because of the smaller temperature dependence of the speciation in the sodium silicate system, the difference in the speciation for the sodium and potassium silicate system decreases with increasing temperature. In addition to the speciation data, the partial Raman spectra for the different species were obtained. The experimentally observed variation of the partial Raman spectra with temperature, and, to a minor extent, with composition, should stimulate future theoretical studies on the vibrational properties of silicate glasses and melts.

Keywords: Silicate melts, Na-silicate glasses, speciation, Raman spectroscopy, partial Raman spectra