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The influence of water on the structure of hydrous sodium tetrasilicate glasses

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ABSTRACT

The structure of sodium tetrasilicate ($Na_2Si_4O_9$) glasses containing 0 to 10 wt% water was investigated by a combination of Raman, IR, and NMR methods. Both the ^{29}Si magic angle spinning NMR data and Raman spectra in the Si-O stretching region clearly show that water depolymerizes the silicate network of the glasses. Q-species distributions calculated from Raman spectra, assuming equal scattering cross sections of all bands in the Si-O stretching region, closely agree with results obtained from NMR data. At low total water contents, the silicate network is depolymerized mainly by breaking of Q^4-Q^4 bonds, whereas breaking of Q^3-Q^3 bonds dominates at high water contents. Near IR spectra show the presence of both OH groups and molecular H_2O in the glasses. The number of nonbridging O atoms per silicon atom, calculated from the near IR data, closely agrees with the results obtained from Raman and NMR, and confirms the assignment of the 4500 cm^{-1} band in the near IR to a combination mode of Si-OH groups. Moreover, the intensity of the fundamental Si-OH stretching band at 910 cm^{-1} in the Raman spectra varies proportionally to the intensity of the 4500 cm^{-1} near IR band. Both IR and Raman spectra show three main bands in the OH-stretching region, centered at 3580, 3000, and 2350 cm^{-1}, due to hydrous species with different hydrogen bond strengths. The relative intensities of these three bands are insensitive to total water content and OH/H_2O ratio, suggesting that both OH and H_2O contribute to each of these bands. This is consistent with the fine structure of the H_2O bending vibration in the IR spectra around 1640 cm^{-1} and with the polarization dependence of the OH-stretching bands in the Raman spectra. Near IR spectra of hydrous sodium tetrasilicate glasses and hydrous aluminosilicate glasses are very similar and show a similar dependence of band intensity on total water content, suggesting that there is no fundamental difference in the dissolution mechanism of water in these systems.