

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

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### ABSTRACT

The structure of sodium tetrasilicate ( $\text{Na}_2\text{Si}_4\text{O}_9$ ) glasses containing 0 to 10 wt% water was investigated by a combination of Raman, IR, and NMR methods. Both the  $^{29}\text{Si}$  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  $\text{Q}^4\text{-Q}^4$  bonds, whereas breaking of  $\text{Q}^3\text{-Q}^3$  bonds dominates at high water contents. Near IR spectra show the presence of both OH groups and molecular  $\text{H}_2\text{O}$  in the glasses. The number of non-bridging 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  $4500\text{ cm}^{-1}$  band in the near IR to a combination mode of Si-OH groups. Moreover, the intensity of the fundamental Si-OH stretching band at  $910\text{ cm}^{-1}$  in the Raman spectra varies proportionally to the intensity of the  $4500\text{ cm}^{-1}$  near IR band. Both IR and Raman spectra show three main bands in the OH-stretching region, centered at 3580, 3000, and  $2350\text{ cm}^{-1}$ , due to hydrous species with different hydrogen bond strengths. The relative intensities of these three bands are insensitive to total water content and OH/ $\text{H}_2\text{O}$  ratio, suggesting that both OH and  $\text{H}_2\text{O}$  contribute to each of these bands. This is consistent with the fine structure of the  $\text{H}_2\text{O}$  bending vibration in the IR spectra around  $1640\text{ cm}^{-1}$  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.