

Structural position of H₂O molecules and hydrogen bonding in anomalous 11 Å tobermorite

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ABSTRACT

For the first time, the structure and dynamics of H₂O in the interlayer of anomalous 11 Å tobermorite have been analyzed based on ab initio molecular dynamics simulations. The simulations provide detailed information on the structure of the hydrogen bonds formed by H₂O molecules and OH groups. The calculated structural parameters of the tobermorite building blocks are in good agreement with the experimental model of Merlini et al. (1999), which is based on X-ray diffraction (XRD) measurements. However, in contrast to the measurements, the simulations suggest that the W1 and W3 sites are split between two general positions with 50% occupancy. It is proposed that the experimental studies provide only averaged coordinates of these sites due to the limitations imposed by the polytypic structures. Analysis of the H₂O dynamics at 321 and 506 K suggest the possibility of a temperature induced order-disorder transition associated with the orientation of O6H··W1 and O6H··W3 hydrogen bonds in the structure of anomalous 11 Å tobermorite. The experimental IR and Raman spectra of 11 Å tobermorite are interpreted based on analyses of the vibrational density of states.

Keywords: Tobermorite, C-S-H phases, water, molecular dynamics