American Mineralogist, Volume 94, pages 156–165, 2009

Structural position of H_2O molecules and hydrogen bonding in anomalous 11 Å tobermorite

SERGEY V. CHURAKOV*

Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland

ABSTRACT

For the first time, the structure and dynamics of H_2O 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 H_2O molecules and OH groups. The calculated structural parameters of the tobermorite building blocks are in good agreement with the experimental model of Merlino 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 H_2O 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