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First Structural Investigation of a Super-hydrated Zeolite

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Powder diffraction data of natrolite, $\text{Na}_{16}\text{Al}_{16}\text{Si}_{24}\text{O}_{80}\cdot 16\text{H}_2\text{O}$ [1], were measured as a function of pressure up to 5.0 GPa using a diamond-anvil cell and a 200 μm -focused monochromatic synchrotron X-ray beam [2]. Upon pressure increase, there is an abrupt volume expansion (ca. 2.5 %) between 0.8 - 1.5 GPa but no change in space group ($Fdd2$). The pressure-induced swelling is caused by the expansion of the unit cell along the a - and b -axes while the c -axis shows a normal contraction throughout the volume expansion period. Rietveld refinements using these data and framework geometrical constraints suggest that this anomalous swelling is due to the *selective sorption* of water into the expanded channels from the alcohol-based pressure-transmitting media. This gives rise to a "superhydrated" phase of natrolite with an approximated formula of $\text{Na}_{16}\text{Al}_{16}\text{Si}_{24}\text{O}_{80}\cdot 32\text{H}_2\text{O}$, containing infinite chains of hydrogen-bonded water molecules along the channels (see Figure). The calculated bulk modulus of the large-volume natrolite is slightly smaller than that of the normal natrolite, illustrating the increased compressibility for this high-water-content phase. The mechanism of the superhydration will be investigated in subsequent high-pressure X-ray and neutron diffraction studies.

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References:

- [1] Artioli, G.; Smith, J.V.; Kvik, A. *Acta Cryst.* **1984**, *C40*, 1658-1662.
[2] Lee, Y.; Hriljac, J.A.; Vogt, T.; Parise, J.B.; Artioli, G. *J. Am. Chem. Soc.* **submitted**.

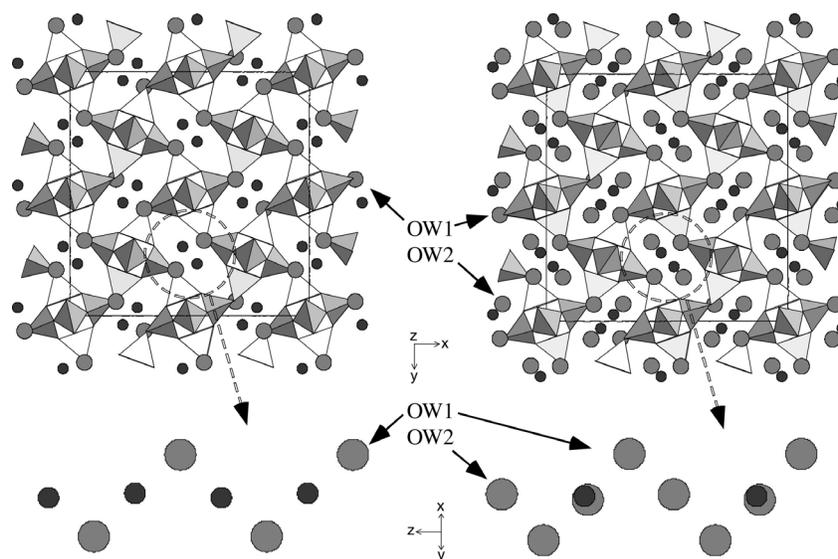


Figure. Polyhedral representations of natrolite at 0.40 GPa (left) and at 1.51 GPa (right) viewed along [001], the chain/channel axis. Dark-large and light-small circles represent water molecules and sodium cations, respectively. Chains of water and sodium are viewed along [110] below each model. Dark (light) tetrahedra illustrate an ordered distribution of Si (Al) atoms in the framework. Straight lines define the unit cell.