

LETTER

**Structuration under pressure: Spatial separation of inserted water during pressure-induced hydration in mesolite**

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**ABSTRACT**

In situ high-pressure single-crystal X-ray diffraction studies of mesolite, an aluminosilicate composed of stacks of Na<sup>+</sup>-containing natrolite and Ca<sup>2+</sup>-containing scolecite layers in the ratio of 1:2, showed two discrete steps of pressure-induced hydration (PIH): first H<sub>2</sub>O molecules are inserted into the natrolite layers between ~0.5 and ~1.5 GPa and subsequently into the scolecite layers. During the PIH in the natrolite layers, the coordination environment of Na<sup>+</sup> changes from six to seven, the same as that of Ca<sup>2+</sup> in the scolecite layers. While the natrolite layers behave as in the mineral natrolite, the scolecite layers show a different behavior from the mineral scolecite by adopting the super-hydrated natrolite-type structure at higher pressure, as a larger distortion is not favorable in the 1:2 layered framework. This spatial separation of inserted H<sub>2</sub>O during PIH and the growing structural similarity of the two layers result in a weakening of  $k \neq 3n$  reflections maintaining the 1:2 layer configuration. Our study of this unique behavior of mesolite provides a simple model of structuration under pressure, and the implications of our experimental findings are discussed.

**Keywords:** Mesolite, structuration, high-pressure, pressure-induced hydration, natrolite, scolecite; Microporous Materials: Crystal-chemistry, Properties, and Utilizations