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P_{H_2O}-dependent structural phase transitions in the zeolite mesolite: Real- and reciprocal-space crystal structure refinements

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Abstract

The response of the mesolite crystal structure ($Na_{16}Ca_{16}Al_{48}Si_{72}O_{240}\cdot 64H_2O$) to dehydration was evaluated as a function of temperature and partial pressure of water (i.e., P_{H_2O} or relative humidity, RH) using laboratory X-ray powder diffraction (XRD; Cu$K\alpha$ radiation) and synchrotron X-ray pair distribution function (PDF) methods. At 425 °C under low-P_{H_2O} conditions ($P_{H_2O} \leq$ ~1.3 mbar), dehydrated mesolite preserved the long-range ordered aluminosilicate framework structure, which has not been previously observed. This new dehydrated phase, x-metamesolite, has unit-cell parameters [a = 16.731(3) Å, b = 17.822(2) Å, c = 6.312(1) Å, V = 1882.5(5) Å^3, and possible space group $Fdd2$] similar to those of other dehydrated natrolite phases (either α1- or α2-metanatrolite). Conversely, under high-P_{H_2O} conditions ($P_{H_2O} >$ ~1.3 mbar), dehydrated mesolite became amorphous (amorphous T_5O_{10}) at 425 °C. The local structure of amorphous T_5O_{10} was characterized by PDF analyses, which showed the formation of twisted T_5O_{10} nano-fibers [with dimensions (LWH) of ~6.9 × 6.9 × 6.3 Å] resulting from breakage of the mesolite aluminosilicate framework. The two distinct high-temperature P_{H_2O}-dependent phase transition paths illustrate the importance of considering the combined effects of temperature and P_{H_2O} in mesolite. In addition, the low-temperature phase transition in mesolite, involving order-disorder of the extraframework cations, also showed a P_{H_2O}-dependent transition temperature. Although, there is no path dependence on P_{H_2O} for this transition, the local arrangement of Na, Ca, and vacancies in disordered metamesolite (formed through the extraframework cation order-disorder phase transition) likely influences the thermal stability of the aluminosilicate framework during further heating.

Keywords: Phase transition, mesolite, XRD data, crystal structure