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Elastic behavior and phase stability of pollucite, a potential host for nuclear waste

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Abstract

The elastic behavior and the phase stability of natural pollucite, $(Cs,Na)_{16}Al_{16}Si_{32}O_{96}{\cdot}nH_2O$, were investigated at hydrostatic pressure by in situ single-crystal X-ray diffraction with a diamond-anvil cell. Pollucite experiences a *P*-induced phase transition, not previously reported in the literature, at $P = 0.66 \pm 0.12$ GPa from cubic ($Ia\bar{3}d$) to triclinic symmetry ($P\bar{1}$). The phase transition is completely reversible and without any appreciable hysteresis effect. No further phase transition has been observed up to 9 GPa. Fitting the pressure-volume data of the low-pressure cubic polymorph with a second-order Birch-Murnaghan Equation-of-State (BM-EoS), we obtain $V_0 = 2558.3(4)$ Å^3, $K_{T0} = 41(2)$ GPa, and $K'_T = 4$ (fixed). For the high-pressure triclinic polymorph, a third-order BM-EoS fit gives $V_0 = 2577.5(40)$ Å^3, $K_{T0} = 25.1(9)$ GPa, and $K'_T = 6.5(4)$. The axial bulk moduli of the high-pressure triclinic polymorph were calculated with a third-order "linearized" BM-EoS. The EoS parameters are $a_0 = 13.699(12)$ Å, $K_{T0}(a) = 25.5(17)$ GPa, and $K'_T(a) = 6.8(6)$ for the *a* axis; $b_0 = 13.728(12)$ Å, $K_{T0}(b) = 23.2(15)$ GPa, and $K'_T(b) = 7.7(7)$ for the *b* axis; $c_0 = 13.710(7)$ Å, $K_{T0}(c) = 25.2(10)$ GPa, and $K'_T(c) = 6.8(4)$ for the *c* axis [$K_{T0}(a):K_{T0}(b):K_{T0}(c) = 1.10:1:1.09$]. Brillouin light-scattering was used to investigate the single-crystal elastic properties of pollucite at ambient conditions. The aggregate adiabatic bulk modulus (K_s) and shear modulus (G), calculated using the Voigt-Reuss-Hill averaging procedures, are $K_s = 52.1(10)$ GPa and $G = 31.5(6)$ GPa. The elastic response of pollucite and other isotypic materials (e.g., analcime, leucite, and wairakite) is compared. The high thermo-elastic stability of pollucite, reflected by the preservation of crystallinity at least up to 9 GPa (at room *T*) and 1470 K (at room *P*) in elastic regime, the large amount of Cs hosted in this material ($Cs_2O \sim 30$ wt%), the immobility of Cs at high-temperature and high-pressure conditions, and the extremely low leaching rate of Cs, make of this open-framework silicate a functional material with potential use for fixation and deposition of Cs radioisotopes in high-level nuclear waste.

Keywords: Pollucite, single-crystal X-ray diffraction, high-pressure, compressibility, phase transition, nuclear waste disposal material