

Thermoelastic properties and phase transition of natural pollucite

J. Schreuer¹, M. Münchhalphen¹, E. Hartman¹

¹Ruhr-Universität Bochum, Institut für Geologie, Mineralogie und Geophysik,
Universitätsstraße 150, Bochum, 44801, Germany
e-mail: schreuer@rub.de

Pollucite (Cs,Na)AlSi₂O₆·yH₂O, a cesium-bearing zeolite belonging to the analcim group characterized by ANA topology of its tetrahedral framework, has long been considered to be a suitable material for use in fixation and deposition of radioactive Cs isotopes in high-level nuclear waste. Particularly favorable properties are the stability and low thermal expansion at high temperatures, the capability to host large amounts of Cs, and the low leaching rate of Cs. However, below about 500 K, the three-dimensional framework gradually changes from an "expanded form" to a "collapsed form" which is accompanied by a significant shrinkage of the unit cell volume without a change of symmetry (e.g., Kobayashi et al. 1997). Phase transformations from the cubic phase to a low-temperature tetragonal phase are also known (e.g., Kobayashi et al. 2006). Both processes can negatively affect the mechanical integrity of pollucite crystals and ceramics. The aim of our work is, therefore, to investigate the thermoelastic behavior of pollucite between 100 K and 673 K using dilatometry and resonant ultrasound spectroscopy.

The elastic properties of our pollucite samples at room temperature are in reasonable agreement with values reported by Sanchez-Valle et al. (2010). The temperature coefficients of the three independent elastic stiffnesses are uncharacteristically positive for stable materials. In the course of the framework "collapse", the longitudinal stiffness c_{11} and the shear resistance c_{44} soften between 673 K and 133 K by nearly 50% and 35%, respectively. At the same time, the deviations from the Cauchy-relations take on negative values, indicating the increasing importance of directional interactions.

Sanchez-Valle C, Chio C-H, Gatta, GD (2010): Single-crystal elastic properties of (Cs,Na)AlSi₂O₆·H₂O pollucite: A zeolite with potential use for long-term storage of Cs radioisotopes. - J Appl Phys 108, 093509

Kobayashi H, Yanase I, Mitamura T (1997): A new model for the pollucite thermal expansion mechanism. - J Am Ceram Soc 80, 2161-2164

Kobayashi H, Sumino S, Tamai S, Yanase I (2006): Phase transition and lattice thermal expansion of Cs-deficient pollucite, Cs_{1-x}Al_{1-x}Si_{2+x}O₆ ($x \leq 0.25$). - J Am Ceram Soc 89, 3157-3161