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Synthesis and crystal structure of the feldspathoid $CsAlSiO_4$: An open-framework silicate and potential nuclear waste disposal phase

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Abstract

Crystalline $CsAlSiO_4$ was synthesized from a stoichiometric mixture of $Al_2O_3 + SiO_2 + Cs_2O$ (plus excess water) in Ag-capsules at hydrostatic pressure of 0.1 GPa and temperature of 695 °C. The duration of synthesis was 46 h. The crystal structure of $CsAlSiO_4$ was investigated by single-crystal X-ray diffraction. The structure is orthorhombic with $Pc2_1n$ space group and lattice parameters: $a = 9.414(1)$, $b = 5.435(1)$, and $c = 8.875(1)$ Å. Because of the orthohexagonal relation between b and a ($a \approx b\sqrt{3}$), within the standard uncertainty on the lattice parameters, a hexagonal superlattice exists, which is responsible for twinning. The crystals are twinned by reflection, with twin planes (110) and (310): twinning in both cases is by reticular merohedry with twin index 2 and hexagonal twin lattice ($\mathbf{L}_T$). The transformation from the lattice of the individual ($\mathbf{L}_{ind}$) to $\mathbf{L}_T$ is given by: $\mathbf{a}_T = \mathbf{a}_{ind} - \mathbf{b}_{ind}$, $\mathbf{b}_T = 2\mathbf{b}_{ind}$, and $\mathbf{c}_T = \mathbf{c}_{ind}$. The refinement was initiated using the previously published atomic coordinates for $RbAlSiO_4$. The final least-square cycles were conducted with anisotropic displacement parameters. $R_1 = 3.04\%$ for 66 parameters and 2531 unique reflections. For a more reliable crystallographic comparison the crystal structure of $RbAlSiO_4$ is reinvestigated here adopting the same data collection and least-squares refinement strategy as for $CsAlSiO_4$.

The crystal structure of the $CsAlSiO_4$ feldspathoid is built on an ABW framework type, showing a fully ordered Si/Al-distribution in the tetrahedral framework. The only extra-framework site is occupied by Cs, lying off-center in the 8mR-channels. $CsAlSiO_4$ is more likely to retain Cs when immersed in a fluid phase, relative to several other Cs-bearing zeolites. The topological configuration of the Cs-polyhedron (and its bonding environment), the small dimension of the pores and the high flexibility of the ABW framework type would imply a better thermal and elastic stability of $CsAlSiO_4$ than those of the zeolitic Cs-aluminosilicates. In this light, $CsAlSiO_4$ can be considered as a functional material potentially usable for fixation and deposition of radioactive isotopes of Cs and can also be considered as a potential solid host for a ^{137}Cs γ-radiation source to be used in sterilization applications.

Keywords: $CsAlSiO_4$, $RbAlSiO_4$, ABW framework type, feldspathoid, crystal structure, nuclear waste disposal phase