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Thermodynamic properties of $CaTh(PO_4)_2$ synthetic cheralite

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Abstract

The mineral cheralite [$CaTh(PO_4)_2$] allows for the incorporation of tetravalent actinides in monazite-based crystalline phases. Experimental determination of its thermodynamic properties is crucial for defining its stability and subsequent long-term ability to immobilize radionuclides. Low-temperature heat capacity from 0.5 to 300 K, enthalpy increments from 485 to 1565 K, and the enthalpy of formation of cheralite from the oxides were measured and reported on for the first time. At 298.15 K, $S°$ = (201.6 ± 2.6) J/(K·mol), which includes the configurational entropy of Ca and Th mixing, ΔH_f^{ox} = –(506.4 ± 9.5) kJ/mol, ΔH_f^{el} = –(3872.8 ± 10.2) kJ/mol, ΔG_f^{ox} = –(501.6 ± 9.6) kJ/mol, and ΔG_f^{el} = –(3635.5 ± 10.2) kJ/mol.

In aqueous environments, cheralite is able to form from whitlockite or apatite and thorianite. Under anhydrous conditions, cheralite can form by solid-state reaction only if the resultant product includes very stable Ca salts instead of CaO.

Keywords: Cheralite, actinide phosphates, thermodynamic properties, nuclear waste