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Thermodynamics of uranyl minerals: Enthalpies of formation of uranyl oxide hydrates

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ABSTRACT

The enthalpies of formation of seven uranyl oxide hydrate phases and one uranate have been determined using high-temperature oxide melt solution calorimetry: $[(UO_2)_4O(OH)_6](H_2O)_5$, metaschoepite; β-$UO_2(OH)_2$; $CaUO_4$; $Ca(UO_2)_6O_4(OH)_6(H_2O)_8$, becquerelite; $Ca(UO_2)_4O_3(OH)_4(H_2O)_2$; $Na(UO_2)O(OH)$, clarkeite; $Na_2(UO_2)_6O_4(OH)_6(H_2O)_7$, the sodium analogue of compreignacite, and $Pb_3(UO_2)_8O_8(OH)_6(H_2O)_2$, curite. The enthalpy of formation from the binary oxides, $\Delta H_{f\text{-ox}}$, at 298 K was calculated for each compound from the respective drop solution enthalpy, ΔH_{ds}. The standard enthalpies of formation from the elements, ΔH_f°, at 298 K are –1791.0 ± 3.2, –1536.2 ± 2.8, –2002.0 ± 3.2, –11389.2 ± 13.5, –6653.1 ± 13.8, –1724.7 ± 5.1, –10936.4 ± 14.5, and –13163.2 ± 34.4 kJ/mol, respectively. These values are useful in exploring the stability of uranyl oxide hydrates in auxiliary chemical systems, such as those expected in U-contaminated environments.

Keywords: Calorimetry, becquerelite, enthalpy, compreignacite, metaschoepite, uranyl, curite, uranium