Thermodynamic Stabilities of MO_{2+x}(s) (M = U, Np, Pu and Am), Pourbaix diagrams.

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The experimental solubilities of the hydrated amorphous freshly precipitated $M(OH)_z$(am) and $MO_2(OH)_z$(am) compounds are often used as an upper limit for the safety assessments of deep waste repositories, since these compounds slowly transform to less soluble ones, as typically $M(OH)_4$(am) to MO_2(cr). Solubility (vs. redox potential) at pH=8, and E-pH predominance diagrams are plotted in aqueous solutions at 25°C by using thermodynamic data recently selected by the NEA-TDB review, or estimated by using classical chemical analogies for the non-redox reactions. The solubilities and relative stabilities are also calculated for the MO_{2+x}(s) crystalline compounds of known stabilities: U_4O_9(s), U_3O_7(s), U_3O_8(s) and Np_2O_5(s) where 2+x = 2.25, 2.33, 2.67 and 2.5 respectively. The stabilities of the other MO_{2+x}(s) compounds are estimated by analogy: M_4O_9(s) (M=U, Np, Pu), M_3O_7(s) and M_3O_8(s) (M=U, Pu) ,and M_2O_5(s) (M=Np, Am) are predicted to be more stable (*i.e.* less soluble), than the amorphous hydroxides. However their precipitation have never been observed at room temperature possibly for kinetic reasons or difficulties in interpreting solubility experiments.

KEYWORDS: Uranium, Neptunium, Plutonium, Americium, thermodynamic, redox, hydrolysis, solubility