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Impact of lanthanide elements on the dissolution of uranium dioxide

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Due to the concomitant formation of oxygen vacancies in the fluorite structure, it was found that the incorporation of trivalent lanthanide elements in ThO_2 or CeO_2 strongly affect their chemical durability during dissolution or leaching tests [1-4]. On the contrary, the impact of such elements on the dissolution of UO_2 remains largely unknown. For this purpose, several lanthanide-uranium mixed based samples incorporating 1 to 20 mol.% of a mixture of lanthanide elements (with the mole ratio : 13 % La ; 7.4 % Y ; 25.6 % Ce ; 12 % Pr ; 42 % Nd) were prepared through quantitative oxalate precipitation. After conversion to the final oxides then sintering, they were characterized from chemical and microstructural points of view and finally submitted to multiparametric dissolution tests in several nitric acid solutions and at various temperatures. Contrarily to Th- and Ce-based mixed oxides, the presence of lanthanide elements in UO_2 did not affect significantly the normalized dissolution rates in concentrated nitric acid solutions ($C > 1\text{M}$). On the contrary, the effect was more important in less concentrated solutions (e.g. a factor of 100 compared to pure UO_2 observed in 0.5M HNO_3) when the role of surface-controlling reactions on the overall dissolution become predominant. Such observations related to the macroscopic description of the dissolution were connected to in operando following of the evolving solid/liquid interface that underlined the preferential dissolution of lanthanide enriched grain boundaries and triple junctions.

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