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Dissolution behavior of MgO based inert matrix fuel for the transmutation of plutonium and minor actinides

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For the transmutation in accelerator-driven systems fuels with a high content of Pu and minor actinides (MA = Np, Am, and Cm) are favored. To increase the burn-up of transuranium elements (TRU) and to reduce the formation of new TRU inert matrix fuels (IMF)[1-3] are preferred. These are ceramic substrates or metallic matrices with high thermal conductivity, which are free of uranium and also have small cross sections for reactions with neutrons. Experimental work on the reprocessability of IMF is being performed within the EU project ASGARD[4]. (Pu,MA)-Oxide within a metallic 92Mo matrix (CERMET) and (Pu,MA)-oxide within a ceramic MgO matrix (CERCER) are under investigation. The influence of the major amounts of matrix material on the reprocessing processes such as dissolution and different extraction steps needs to be investigated. A complete dissolution of the actinide oxide and the matrix material and the actinide oxide, or a selective dissolution, where the matrix remains undissolved, can be considered.

MgO pellets as well as MgO pellets containing different amounts of CeO2 (5, 10, 25, and 40 wt%), which serves as surrogate for plutonium dioxide, were produced and characterized. Here, we present new experimental data on the dissolution kinetics of the matrix material in nitric acid as a head-end step prior to partitioning by solvent extraction. The dissolution rate was studied in macroscopic experiments as a function of acid concentration and volume, temperature, stirring velocity, and pellet density (85, 90, 96, and 99%TD). MgO is soluble even under mild conditions (RT, 2.5 mol/L HNO3). The dissolution rates of MgO at different acid concentrations are rather similar, whereas the dissolution rate is strongly dependent on the temperature.

Additionally, the dissolution process was investigated following a microscopic approach. Detailed SEM investigations show a heterogeneous reactivity of the MgO pellet's surface. A model was developed to describe the evolution of the pellet surface area. The additional surface area of the pellets caused by the pores created during dissolution was estimated under the assumption that they are all cylindrical with a diameter equal to the height.[5] The obtained cylinder surfaces were summed up and related to the total geometrical surface area of the pellet. Considering the additional surface area obtained by estimation of the development of pores in the pellet surface resulted in a more or less constant dissolution rate of approximately 0.02 gs-1m-2.

Moreover, dissolution rates of the inert matrix fuel containing CeO2 as surrogate for the (Pu,MA)-oxide were determined as a function of the acid concentration and temperature. During the dissolution of MgO/CeO2 pellets the MgO dissolves completely, while the bulk of CeO2 (>99%) remains undissolved.

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