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Dissolution studies on molybdenum-based inert matrix fuel targets for the transmutation of minor actinides

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Plutonium and minor actinides (MA = Np, Am, and Cm) are of particular concern with respect to long term radio toxicity, heat load issues and proliferation risks. One possibility to cope with this problem may be the transmutation of plutonium and minor actinides in subcritical accelerator-driven reactors (ADS). Fuels with high Pu and MA contents are preferred for transmutation. To increase the burn-up of transuranium elements (TRU) and to reduce the formation of new TRU a non-fissile, inert matrix is added to the fuel to improve the thermal properties. These so-called inert matrix fuels (IMF) [1-3] contain ceramic substrates or metallic matrices with high thermal conductivity, which are free of uranium and neutron transparent. The reprocessability of (Pu,MA)-oxide within a metallic ^{92}Mo matrix (CERMET) is under investigation within the EU project ASGARD [4]. A major difficulty will be to optimize the dissolution process of the used fuel after ADS operation as a head-end step prior to partitioning by solvent extraction. Mo as an inert matrix poses additional challenges with respect to its REDOX-chemistry. Mo is soluble in nitric acid but is oxidized to MoO_3 which then precipitates.[5] There is a need to avoid coprecipitation with Pu, thus leading to Pu losses. This work focuses on the dissolution kinetics of molybdenum as matrix material in nitric acid. Molybdenum pellets as well as Mo pellets containing different amounts of cerium dioxide (5, 10, 25, and 40 wt%), which serves as surrogate for plutonium dioxide, were produced. The pellets were compacted at 640 MPa and sintered at 1600 °C. The dissolution rate was investigated as a function of acid concentration and temperature. The dissolution rate of the Mo-pellets strongly depends on the acid concentration. Higher acid concentrations result in faster dissolution but also in more precipitation. An increase in temperature also results in a faster dissolution and larger amount of precipitation. Iron (III) is known to significantly increase the solubility of uranyl molybdate in nitric acid. Anion exchange resin studies indicate that a negatively charged iron-molybdenum complex ion is present in such solutions. [6,7] The addition of ferric nitrate to the acid increases the dissolution rate of Mo-pellets in 1M HNO_3 due to complexation. The pellet can be completely dissolved in 1 mol/L HNO_3 containing two equivalents Fe(III). An increase of temperature results in faster dissolution kinetics, but at temperatures above room temperature precipitates occur even in the presence of iron.

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