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Dissolution of Mo-based CERMET fuel: ESI-TOF MS Speciation in nitric acid medium

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The main long term contributors to spent fuel radiotoxicity are plutonium followed by the minor actinides (MA = Np, Am, and Cm). A possibility to reduce the radiotoxic inventory and the footprint of the repository is to separate the most radiotoxic and long-lived elements from spent fuel and to transmute them into non-radioactive elements or elements with a much shorter lifetime. For the transmutation in accelerator-driven systems (ADS) fuels with a high content of Pu and MA are preferred. To increase the burn-up of transuranium elements (TRU) and to reduce the formation of new TRU inert matrix fuels (IMF) [1-3] are favored. 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. The reprocessability of (Pu,MA)-oxide within a metallic 92Mo matrix (CERMET) is under investigation within the EU project ASGARD [4].
Dissolution in nitric acid is the first step in reprocessing. Therefore, detailed knowledge of the speciation of molybdenum in nitric acid medium is crucial on the one hand to understand this dissolution process and on the other hand as a basis for the design of a tailored extraction process. The speciation of molybdenum has been studied intensively in hydrochloric and perchloric acid medium[5,6], but little is known about the speciation in nitric acid medium, especially at high acidities, which are relevant for the dissolution of IMF. The solution species of molybdenum in strongly acidic nitric acid medium need to be extensively characterized and quantified. Therefore, electrospray ionization mass spectrometry, which can probe the stoichiometry and relative abundances of solution species, was applied.
Here, we present new experimental data on the speciation of molybdenum as a function of nitric acid concentration. Isotopically pure 98Mo powder was dissolved in nitric acid and measured with the ALBATROS ESI-TOF [7]. Monomeric, dimeric, trimeric, tetrameric, and pentameric cationic molybdenum species have been detected. Besides the presence of hexavalent Mo species the spectra show that pentavalent Mo species are present in solution in spite of the oxidizing condition in strong nitric acid.

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