Fabrication of UO_2-based model systems for spent nuclear fuel

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Spent nuclear fuel (SNF) is a highly complex and heterogeneous material. Therefore, it is challenging to gain mechanistic insights into the role of individual contributors to the long-term matrix corrosion and several corrosion mechanisms are convoluted in a macroscopically observable radionuclide release rate. So far model systems such as SIMFUEL [1-2] or pure UO_2 were either too complex or not suitable to enable single effect studies of SNF corrosion. The challenge is to design an appropriate chemical system and to develop a suitable synthesis route for the fabrication of simplified UO_2-based model systems. Here, we present a bottom-up approach to fabricate UO_2-based model systems, from simple to more complex systems. As a first step UO_2 pellets containing 1 to 2 wt% Nd were synthesized. Nd serves as fission product surrogate of the complete lanthanide series and the lanthanide concentrations in SNF are mimicked.

Various wet chemical approaches such as the weak acid resin process, sol-gel routes as well as coprecipitation routes were applied and compared. Well-densified pellets (> 95% of the theoretical density) were obtained with all methods. Samples exhibit grain sizes (~8 μm) relatively similar to that of SNF [3]. Examinations by XRD and SEM-EDX revealed that all ceramics are single phased with the cubic fluorite structure and exhibit a homogeneous Nd-distribution.

To go one step beyond the pure doping of UO_2 ceramics with lanthanides, Pd was introduced into the matrix to mimic the metallic epsilon particles which are suspected to play a catalytic role in the redox behavior of SNF. Therefore, as a first approach pure UO_2 powder from a precipitation route was ball-milled and coated with a suspension of $Pd(NO_3)_2$ followed by a reductive calcination step [4]. The obtained ceramics were well-densified with a similar microstructure as the Nd-doped ceramics. Precipitates of ~1 μm Pd are observed mainly in the grain boundaries but intra-granular precipitates occurred also.

[1] Lucuta et al. (1991) J. Nucl. Mater. **178**, 48-60. [2] Hiezl et al. (2015) J. Nucl. Mater. **456**, 74-84. [3] Carbol et al. (2012) Comprehensive Nuclear Materials 389-420. [4] Baena et al. (2015) J. Nucl. Mater. **461**, 271-281.