frontiers | Frontiers in Nuclear Engineering

TYPE Editorial
PUBLISHED 01 June 2023
DOI 10.3389/fnuen.2023.1214113

OPEN ACCESS

EDITED AND REVIEWED BY
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RECEIVED 28 April 2023
ACCEPTED 18 May 2023
PUBLISHED 01 June 2023

CITATION
Léchelle J, Luzzi L and Zhang L (2023), Editorial: Experimental and model-based assessment of diffusion kinetics of actinides and oxygen during fuel fabrication and of fission products over in-pile use.
Front. Nucl. Eng. 2:1214113.
doi: 10.3389/fnuen.2023.1214113

Editorial: Experimental and model-based assessment of diffusion kinetics of actinides and oxygen during fuel fabrication and of fission products over in-pile use

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KEYWORDS

diffusion coefficient, experiment, modeling, nuclear fuel, oxygen diffusion, actinide diffusion

Editorial on the Research Topic
Experimental and model-based assessment of diffusion kinetics of actinides and oxygen during fuel fabrication and of fission products over in-pile use

During the nuclear fuel cycle, stages of fabrication and in-pile processing imply diffusion phenomena are taking place in mixed oxide ceramics. These diffusion phenomena change the microstructure and key properties of the fuel, such as thermal conductivity and creep behavior. For instance, in the fabrication process, the interdiffusion of uranium and plutonium cations leads to a homogeneous solid solution and an increased grain size. Diffusion of oxygen anions leads to deviation from the stoichiometry of the fuel, which controls physical properties such as thermal conductivity. These phenomena can also occur during the in-pile stay of the fuel with an impact on creep properties. Gaseous fission products tend to remain in the bulk of fuel pellets. Thus, understanding diffusion phenomena makes it possible to avoid bad cooling of the fuel (due to the degradation of the fuel-cladding gap thermal conductance) at the beginning of in-pile operation and an increase in rod or pin internal pressure at the end of fuel life, which may lead to potential sheath embrittlement. The time required for the fission gas to reach grain boundaries depends on grain size. For all these reasons, it is necessary to be able to quantify these diffusion phenomena that occur either in the bulk of grains, at their surfaces, or along grain boundaries. This series of articles provides insight into the current knowledge in this field and can help in fuel performance codes, such as BISON (Yu et al., 2021), SCIANTIX (Pizzocri et al., 2020), or PLEIADES (Marelle et al., 2017).

For future reactors with higher levels of Pu and, to a lesser extent, Am and Np, deviation from stoichiometry may reach higher values with an oxygen-to-metal ratio between 1.5 and 2.0, which occurs due to the behavior of the 5f electrons involved in the chemical binding. Deviation from stoichiometry may also lead to a reordering of oxygen vacancies and even to a different crystal structure.

These issues are presented in the article by Charlton et al., which is based on an experimental study of analogous 4f-type chemical elements. A soft chemistry fabrication route implying sol–gel processes and freeze-drying can be designed to overcome the slow diffusion of cations in these compounds. The transition from one crystal structure to another occurs through cation diffusion, which is the limiting step of the evolution of these materials. In nuclear fuels during operation, radiation fields dramatically increase the concentration of point defects, leading to enhanced diffusion. Therefore, defect models for radiation-induced diffusion are required for advanced fuel design.

To study deviation from the stoichiometry of UO_2, which is the main constituent of nuclear fuel, Watanabe and Kato performed high-temperature gas equilibrium experiments (between 1,673 K and 1,873 K). These experiments enabled the analysis of the type of point defect that can occur with a specific oxygen potential value as well as determining a precise value of deviation from stoichiometry under these conditions. The value of the oxygen chemical diffusion coefficient was obtained from the speed at which such deviations from stoichiometry were reached.

A similar study by Kato et al. extends to other actinide oxides, such as ThO_2, (U, Pu)O_2, and to a lanthanide oxide, CeO_2, to evaluate point defect concentrations as well as the oxygen self-diffusion and chemical diffusion coefficients. The contribution of Frenkel pairs to the heat capacity is also evaluated therein and was found to be noticeable in the case of CeO_2 and ThO_2. This study also provides insight into the mechanisms of reduction and oxidation, which turned out to be different.

A thorough literature review was carried out by Vauchy et al. for the specific case of the study of cation thermal interdiffusion in uranium–plutonium mixed oxide fuels. This study relies upon published experimental results from 1963 to 2013 that consider both temperature and deviation from stoichiometry effects. Although of the highest importance, cation interdiffusion coefficients remain scarce and scattered. This scattering is mainly due to difficulties linked to the mastering of the oxygen potential value during the experiment as well as to the effect of extra cations, such as americium, due to ^{241}Pu radioactive decay. However, it can be concluded that interstitial oxygen enhances interdiffusion, whereas oxygen vacancies may cluster and limit interdiffusion.

Oxygen diffusion is better understood than the diffusion of the much slower cations, and oxygen diffusion is especially sensitive to extra cations and oxygen potential. Much remains to be carried out in this latter field to identify reliable cation diffusion coefficients both in bulk grains and at grain boundaries. More experimentally dedicated work is required, but simulation-based assessments, such as the kinetic cluster expansion method (Schuler et al., 2020), could help provide diffusion coefficient values. Precise knowledge of the effect of radiation upon point defects in fuel is also necessary to help future simulations.

Author contributions

LL, LZ, and JL worked as co-topic editors. The Research Topic description was written by JL; all three co-topic editors encouraged potential contribution directors, selected reviewers for each article in this Research Topic, and followed the reviewing process. The first draft of the editorial was written by JL and corrected by LL and LZ. All authors contributed to the article and approved the submitted version.

Conflict of interest

The author(s) JL, LL, and LZ declared that they were editorial board members of Frontiers at the time of submission. This had no impact on the peer review process and the final decision.

Publisher's note

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