Evolution of Uranium Release in Slip-Cast and Pressed UO_2 Pellets Over Extended Leaching Periods

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***Keywords :** Uranium dioxide (UO_2), Leaching behavior, Microstructure, Pellet fabrication

1. Introduction

The chemical durability of uranium dioxide (UO_2) is vital for the safe disposal of spent nuclear fuel in geological repositories. Previous studies often focused on dissolution kinetics [1], but the influence of pellet size and fabrication-induced microstructure on leaching behavior is less explored. Differences in pellet mass and manufacturing methods can alter grain size, porosity, and reactive interfaces, profoundly impacting dissolution rates. This study compares two distinct UO_2 specimens: a large 70 g slip-cast block and a small 3.5 g pressed and sintered pellet. By combining long-term leaching experiments with microstructural analysis, the study elucidates how mass and microstructure influence uranium release in disposal environments.

2. Experimental

Leaching tests followed a modified ASTM C1308 [2] semi-dynamic approach (Figure 1). Two UO_2 pellet types were examined: a 3.5 g pressed/sintered pellet leached in 35 mL deionized water at 60 °C, and a 70 g slip-cast block leached in 250 mL deionized water at room temperature (25 °C). The leachant volumes were selected to maintain a solution-to-surface-area ratio of approximately 10 mL/cm², equivalent to a surface area-to-leachant volume ratio of 0.1 cm^{-1}, ensuring consistent leaching conditions for each specimen. Both samples were placed in PTFE vessels to avoid contamination. The tests spanned 112 days with sampling at 3 hours, 1, 2, 3, 7, 14, 28, 56, and 112 days. After each sampling, the leachant was fully replaced to maintain sink conditions.

Leachate samples were filtered through 0.45 µm PTFE filters and acidified with 1% HNO_3. Uranium concentrations were measured via inductively coupled plasma–mass spectrometry (ICP–MS; Analytik Jena PlasmaQuant MS 9100) at KAERI. Uranium was the sole analyte monitored, as pure UO_2 was used. Microstructural characterization utilized scanning electron microscopy (SEM; Hitachi SU 8010) to assess grain size and boundary morphology. These observations enabled interpretation of dissolution behaviors related to structural features.

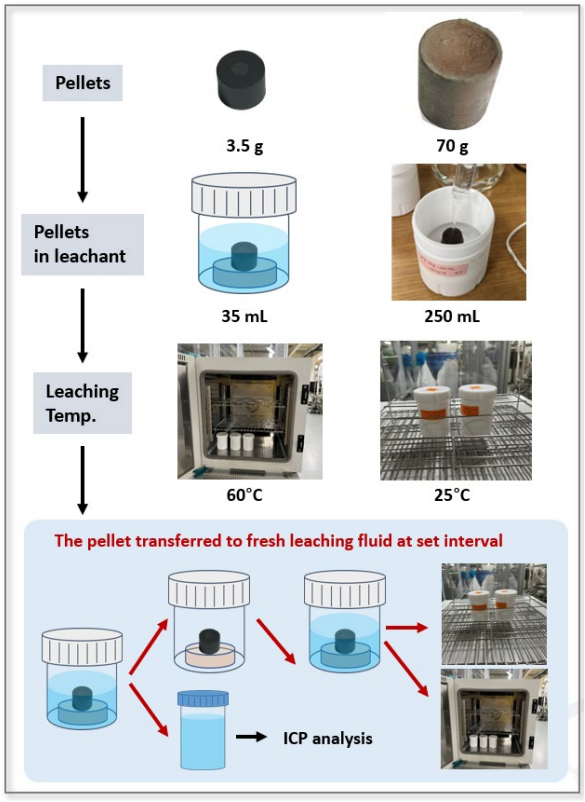

Figure 1. Schematic of the semi-dynamic leaching test showing the procedure applied to two UO_2 pellet types with different sizes and fabrication methods.

3. Results and Discussion

Scanning electron microscopy (SEM) micrographs of the UO_2 pellets are shown in Figure 2. The pressed and sintered 3.5 g pellet exhibits relatively large grains with fewer grain boundaries, as visible in Figures 2a. This microstructure limits the reactive interfaces accessible for dissolution. In contrast, the slip-cast 70 g block (Figures 2b) has a finer grain size and a higher density of grain boundaries, providing increased reactive surface area that facilitates uranium release. These

microstructural differences, driven by fabrication methods, are expected to have a significant influence on leaching behavior.

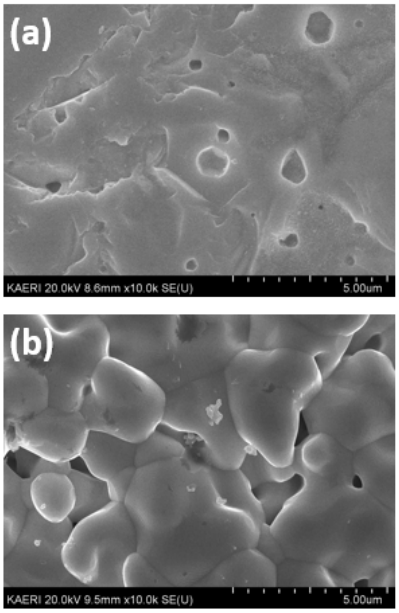

Figure 2. SEM images of UO_2 pellets: (a) pressed and sintered 3.5 g pellet, (b) slip-cast 70 g pellet .

Figure 3 presents the cumulative uranium release from the two pellets during the 112 day leaching test. Initially and throughout the intermediate stages, the slip-cast 70 g pellet released uranium at a higher rate than the smaller pressed pellet, in line with its finer microstructure and increased dissolution pathways. However, after 112 days, the cumulative uranium releases from both pellets converged to nearly the same value. This indicates that although microstructure strongly governs the early and intermediate kinetic differences in uranium leaching, the long-term cumulative release is ultimately limited by bulk properties such as total uranium content and matrix characteristics.

Thus, the combined interpretation of microstructural and leaching data highlights that pellet fabrication influences initial dissolution kinetics, while extended leaching leads to similar uranium release endpoints irrespective of pellet size. These insights emphasize the need to incorporate both microstructural and bulk considerations in evaluating spent fuel durability for disposal.

4. Conclusion

This study shows that uranium leaching from UO_2 pellets is strongly influenced by microstructure and fabrication method. The slip-cast 70 g pellet with finer grains initially released uranium faster than the larger-grained pressed 3.5 g pellet. However, after extended leaching, cumulative uranium release from both pellets converged. These results highlight that microstructural features control early dissolution kinetics, while long-term release is governed by bulk properties. Understanding both aspects is essential for accurate prediction of spent fuel durability in disposal environments.

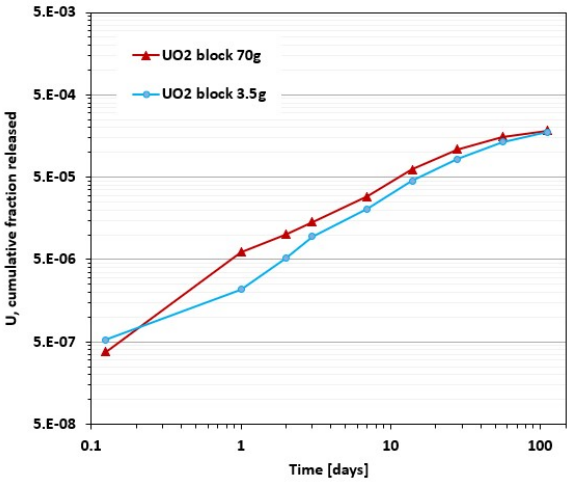

Figure 3. Cumulative fraction of uranium released as a function of time for the slip-cast 70 g and pressed 3.5 g pellets.

ACKNOWLEDGEMENTS

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korean government (Grant No. 2021M2A7A1080748)

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