Key Issues in the Fabrication of Oxycarbide Kernels for TRISO Fuel

Minseok Lee[a,b], Injin Sa[a], Yeon-Ku Kim[a], Eung-Seon Kim[b]*

[a]*Advanced Nuclear Fuel Technology Development Division, Korea Atomic Energy Research Institute, 111, Daedeok-daero 989 beon-gil, Yuseong-gu, Daejeon, Republic of Korea 34141*
[b]*Korea Advanced Institute of Science and Technology, Nuclear & Quantum Engineering Dept., 291 Daehakro, Yuseong, 34141, Republic of Korea*
**E-mail: kimes@kaeri.re.kr*

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1. Introduction

HTGRs designed to operate at outlet temperatures exceeding 750 °C or higher, are regarded as next-generation reactors with the potential for hydrogen production. which comprises spherical uranium oxycarbide (UCO) kernels—mixture of UO_2-UC_2—and coated with inner pyrolytic carbon (IPyC), SiC, and outer pyrolytic carbon (OPyC) layers to prevent diffusion of fission products. The key challenges in fuel kernel fabrication include achieving high densification, a well-formed UC_2 phase, uniform fuel kernel size distribution, and high sphericity [1]. To accomplish these goals, optimization of fuel kernel fabrication process. This study reports on key technical issues encountered during process experiments for oxycarbide fuel kernel fabrication.

2. Methods and Results

2.1. Dispersing carbon black

UCO kernels are fabricated through the carbothermic reduction of UO_3 hydroxide microsphere gel and uniformly dispersed carbon black. Therefore, ensuring homogeneous dispersion of carbon black in the broth solution prior to drop-casting is critical for achieving high densification and UC_2 phase [2,3]. Since carbon black is dispersed well under basic conditions, it is typically first dispersed in a hexamethylenetetramine (HMTA)-urea solution before being combined with the uranium solution [3].

To evaluate the dispersion state of carbon black before internal gelation, particle size distribution was measured via dynamic light scattering. Raven 5000 (Birla Carbon, USA), with a BET surface area of 583 m^2/g and a primary particle size of 8 nm, was dispersed in HMTA-urea solutions using an anionic dispersing agent, Tamol SN. As shown in Fig. 1, the particle size distribution in the carbon black dispersion exhibits a bimodal distribution, where each peak corresponds to agglomerates (>1 μm) and aggregates (<100 nm). Large agglomerates lead to the formation of non-uniform UC_2 phases, free carbon, and cracks. Therefore, it is essential to optimize dispersion conditions to effectively mitigate agglomeration.

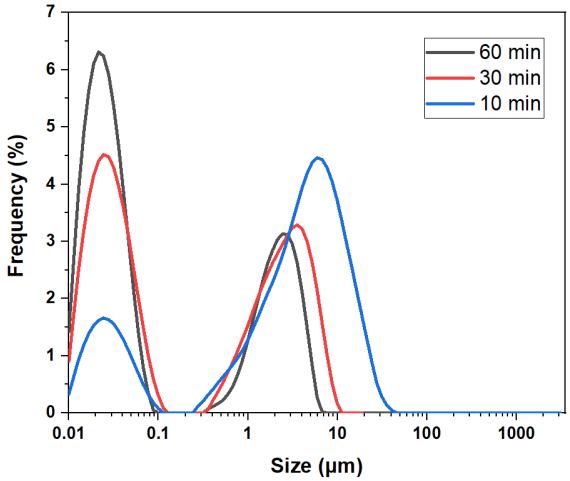

Fig. 1. Particle size distribution of carbon black aggregates in HMTA-urea solution with Tamol SN at different ultrasonication time.

2.2. Fabrication of gel microspheres

The broth solution for internal gelation was prepared by mixing acid-deficient uranium nitrates (ADUN) with HMTA-urea (and carbon black for the UCO kernel) [5]. After chilling at 0°C, the broth solution is drop-cast onto hot silicon oil (90°C) using a vibrating nozzle to produce UO_3 microsphere gel as shown in Fig.2. Due to waste management concerns, ADUN solutions containing carbon black were not yet used. The drop-casting process involves applying a controlled vibrational frequency to a laminar liquid jet as it is extruded through a nozzle, inducing jet breakup into uniformly sized droplets. The most effective breakup occurs when the vibrational frequency matches the natural instability frequency of the jet. Droplet size is primarily determined by the nozzle diameter and liquid flow rate, with additional control achieved through vibrational frequency adjustments. Visual monitoring techniques, such as stroboscopic illumination, aid in fine-tuning the droplet formation process.

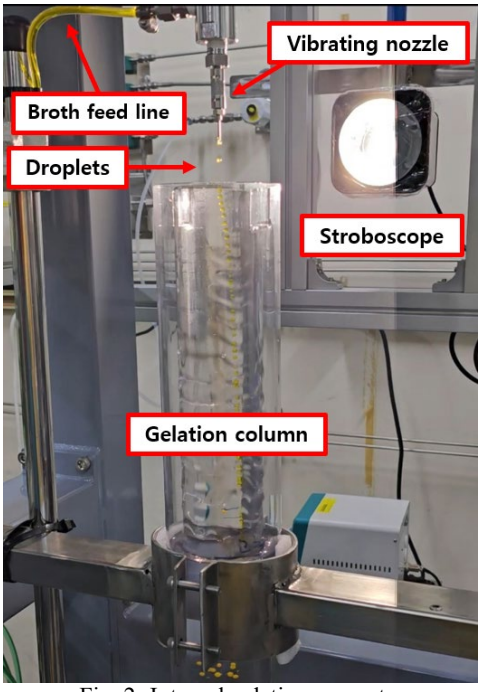

Fig. 2. Internal gelation apparatus.

2.3. Heat treatment

The UO_3-C microsphere gel, prepared through drop casting and washing, undergoes carbothermic reduction to the UC_2 phase and sintering of the UO_2 matrix through high-temperature heat treatment above 1550°C. During this process, the reaction between UO_2 and UC_2 must be controlled by introducing CO into the heat treatment atmosphere [3]. In consideration of nuclear regulatory restrictions, Zr was used as a surrogate in preliminary heat treatment experment before handling U [5]. Fig. 3 presents the results of a ZrO_2-ZrC surrogate nuclear fuel experiment. In an atmosphere without CO, a reaction between ZrO_2 and ZrC occurs, resulting in a white surface. In contrast, the surrogate nuclear fuel heat-treated in a CO atmosphere exhibits a black surface, indicating that the reaction between ZrO_2 and ZrC has been suppressed.

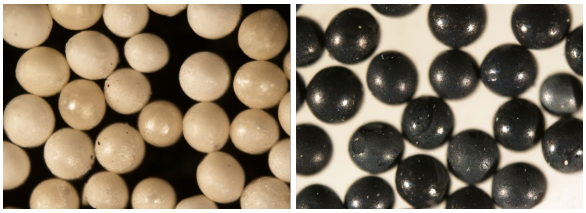
Fig. 3. Optical microscopy images of ZrO_2-ZrC surrogate fuel after heat treatment without CO (a) and with CO (b).

3. Conclusion

In this study, we investigated the internal gelation process for fuel kernel fabrication and identified several key challenges. However, the issues discussed are not exhaustive, as additional challenges may arise in various stages, including aging, washing, drying, and calcination. Future work will focus on optimizing key processes to further improve the quality and reproducibility of fuel kernel fabrication.

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