Carbothermic Reduction and Sintering of UCO kernels

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1. Introduction

Uranium oxycarbide (UCO, UO_2–UC_x) fuel kernels have been adopted as a key fuel form for TRISO particles, particularly through the U.S. Department of Energy (DOE)-led Advanced Gas Reactor (AGR) program, owing to their ability to suppress carbon monoxide (CO) gas generation during irradiation [1]. UCO kernels are fabricated from UO_3–C gel microspheres prepared by the internal gelation process, followed by carefully controlled heat treatment, carbothermic reduction, and sintering [2,3]. In this study, UCO kernels were produced by heat treatment to obtain the desired carbon and oxygen contents and kernel density. The results of the carbothermic reduction and sintering processes used to obtain UCO kernels with the desired properties are presented.

2. Methods and Results

2.1 Preparation of UO_3-C microsphere

After carbon black was dispersed in a mixed solution of hexamethylenetetramine (HMTA) and urea, acid-deficient uranyl nitrate (ADUN), with a composition of $UO_2(NO_3)_{2-x}$, was added. The resulting mixture was then injected through vibrating needles using a gear pump to form droplets for the internal gelation process. The droplets were dropped into hot silicone oil, where gelation occurred, followed by aging, washing, and drying to produce UO_3–C gel spheres.

2.2 Heat treatment

The dried UO_3–C gel spheres were calcined in a furnace at 650 °C under an Ar–H_2 atmosphere to remove residual organics and convert the uranium oxide phase to UO_2. The temperature then increased to 1790 °C to promote carbothermic reduction and sintering, resulting in the formation of dense UCO kernels. The fabricated UCO kernels were characterized by Archimedes density measurements, X-ray diffraction (XRD), and scanning electron microscopy (SEM).

After heat treatment, the UCO kernels were uniformly fabricated with a diameter of approximately 425 μm, as shown in Fig. 1, and their density was measured to be approximately 10.6 g/cm³ using the Archimedes method. XRD analysis (Fig. 2) revealed that the UCO kernels consisted of UO_2, β-UC_2, and UC, with phase fractions of 73.1, 19.5, and 7.4 mol%, respectively. Based on the calculated theoretical density of 11.44 g/cm³, the fabricated UCO kernels achieved approximately 92.7% of theoretical density (%TD).

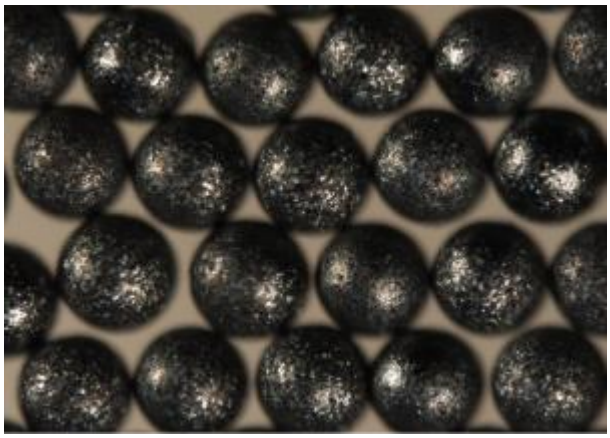


Fig. 1. Optical images of UCO kernels after heat treatment.

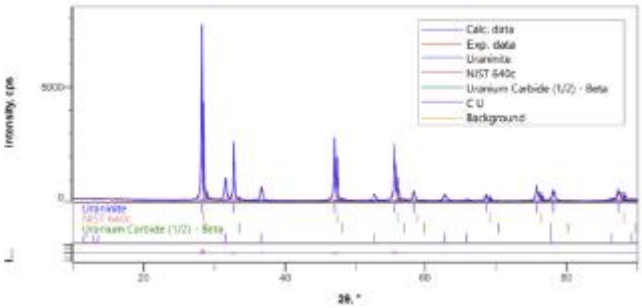


Fig. 2. XRD patterns of UCO kernels and their Rietveld refinement

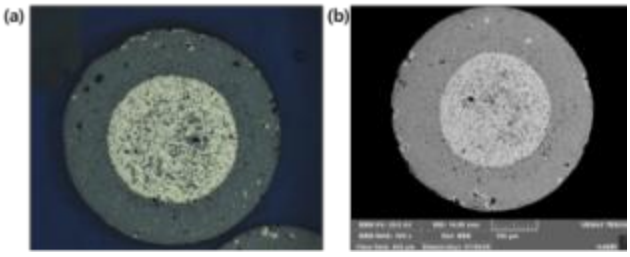


Fig. 3. (a) Optical microscopy and (b) back scattered electron (BSE) micrscopy of cross section of UCO kernels

As shown in the optical and BSE images in Fig. 3, the UCO kernels exhibited a core–shell structure, with the oxide phase appearing dark and the carbide phase appearing bright. It should be noted that UC and UC_2 could not be distinguished from each other based on image contrast alone. This core–shell morphology is presumed to result from the reaction between UC_x and UO_2 in the outer region, leading to the formation of a UC_xO_γ oxycarbide solid-solution phase.

3. Conclusions

In this study, heat-treatment conditions were investigated to optimize the fabrication of UCO kernels. Carbothermic reduction and sintering were carried out at temperatures up to 1790 °C. The fabricated UCO kernels achieved a density of 92.7% of the theoretical density (TD) and a total carbide phase fraction of 26.9 mol%. Future work will focus on achieving a density higher than 95% TD, controlling the carbide phase fraction to approximately 25 mol%, and minimizing the shell region in the core–shell structure.

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