Effects of Carbon Black Dispersion on Oxycarbide Fuel Kernels

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1. Introduction

Uranium oxycarbide (UCO) kernels are highly promising nuclear fuels in TRISO due to their excellent mitigation of CO gas release and superior fission product retention [1]. These kernels are fabricated via carbothermic reduction using carbon black as the carbon source, where the uniform dispersion of carbon is critical for the final density and stoichiometry [2-3]. For this study, zirconium (Zr) was used as a temporary surrogate for uranium to enable systematic investigation [4]. We evaluated the effects of carbon black dispersion states on the overall quality, including the density and C/Zr ratio, of the surrogate Zr oxycarbide (ZrCO) fuel kernels.

2. Methods and Results

2.1. Preparation of carbon black dispersion

Two types of carbon black (C-1 and C-2) were dispersed in a 3.2 M HMTA (hexamethylenetetramine)-urea solution using a ultrasonication probe. The molar ratio of HMTA-to-carbon was 1.3. Tamol SN was added as a dispersing agent for C-1, whereas C-2 required no dispersant due to its surface modification with sulfonate groups. The degree of dispersion was controlled by varying the ultrasonication time (10, 30, and 60 min).

As shown in Fig. 1, the aggregate size of the carbon black gradually decreases with increasing ultrasonication time. Specifically, the median particle size (D50) decreased from 0.054 μm to 0.037 μm for C-1, and from 0.028 μm to 0.024 μm for C-2. This is accompanied by a gradual decrease in viscosity, as depicted in Fig. 2, indicating that the dispersion becomes more uniform.

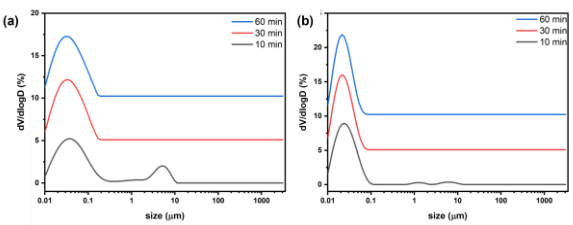

Fig. 1. Particle size distributions of carbon black dispersion with dispersion times: (a) C-1 and (b) C-2.

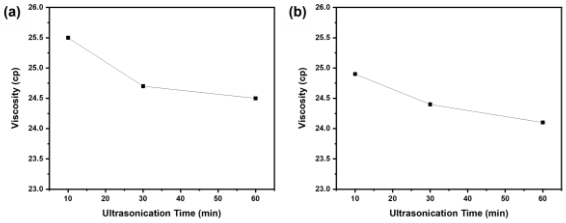

Fig. 2. Change in viscosity of carbon black dispersion with dispersion time: (a) C-1 and (b) C-2.

2.2. Fabrication of ZrCO kernels

ZrCO kernels were fabricated by mixing a carbon black dispersion, prepared by 30 min of ultrasonication, with a $ZrO(NO_3)_2$ solution and HNO_3 at 0 °C for 30 min. The C/Zr molar ratio was 1. The mixture was then injected through a 1.0 mm nozzle into hot silicone oil (90 °C) under vibration at ~10 Hz to generate droplets. During this process, HMTA thermally decomposed to produce ammonia, which induced gelation of ZrO_2^+ ions to form microsphere gels.

The as-formed microsphere gels were sequentially washed with TCE, ammonia solution, pressurized water, DI water, and IPA, followed by drying. The dried microspheres were then heat-treated at 1600 °C under an Ar/CO mixed gas atmosphere to obtain ZrCO kernels via a carbothermic reaction.

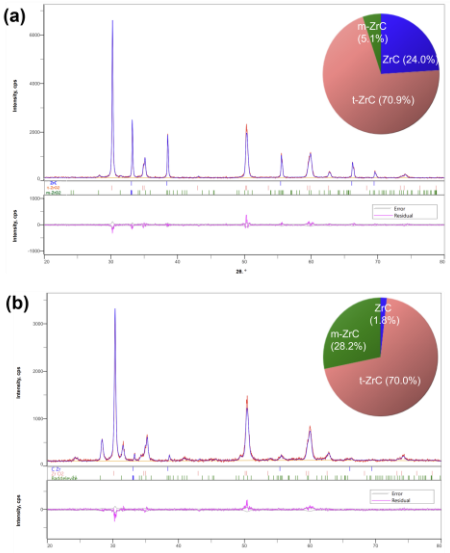

Fig. 3. XRD patterns and Rietveld phase analysis of the fabricated ZrCO: (a) C-1 and (b) C-2.

As shown in Fig. 3, approximately 24 wt% of ZrC was formed in the ZrCO fabricated from the C-1 source, whereas only 1.8 wt% of ZrC was produced when the C-2 source was used. This is thought to be due to the very small size of the dispersed carbon black aggregates of C-2, which likely led to the rapid consumption of carbon. ZrCO synthesis experiments under different dispersion conditions are currently in progress.

Fig. 4 shows the cross-sectional microstructure of the ZrCO kernel synthesized from C-1. The outer region of the microsphere consists of ZrO_2, while the interior exhibits a mixed region of ZrO_2 and ZrC. This morphology may also be influenced by the dispersion behavior of the carbon black and will be further investigated in future work.

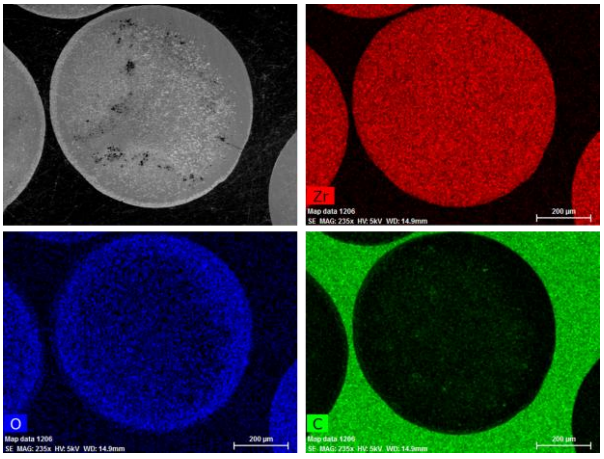

Fig. 4. SEM image and EDS analysis of ZrCO fabricated with C-1

3. Conclusions

In summary, we investigated how carbon black dispersion states affect the properties of oxycarbide fuel, focusing on ZrCO synthesis. The findings highlight the critical role of carbon black dispersion in determining phase formation and microstructure. Future studies will explore a broader range of dispersion conditions to systematically assess their impact on key properties, including the C/Zr ratio, density, and microstructure.

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