

Chemical vapor infiltration SiC matrix densification of fully ceramic micro-encapsulated fuel fabricated by additive manufacturing

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1. Introduction

After the Fukushima nuclear accident in 2011, fully ceramic-microencapsulated (FCM) fuel was proposed to enhance the damage tolerance in nuclear reactor [1]. The FCM fuel is made up Tri-structural ISotropic (TRISO) particle in silicon carbide (SiC) matrix. TRISO particle is composed uranium dioxide (UO₂) fuel kernel-coated with buffer layer / inner pyrolytic carbon (IPyC) / SiC / Outer pyrolytic carbon (OPyC). TRISO particles have extremely low nuclear fission product and high safety. The SiC matrix has a low neutron absorption cross-section, excellent irradiation resistance, and high specific strength at elevated temperatures. It also prevents nuclear fission products inside the particles and helps maintain the structure of the TRISO particles at high temperatures.

The SiC matrix of FCM fuel is typically manufactured through hot pressure sintering. The theoretical density of SiC can be achieved by hot pressing at 1700~1800 °C for 1~5 hours [2]. However, if the TRISO particles are unevenly distributed, hot pressure sintering can be damaged by the fuels [3]. To overcome this issue, the FCM fuel is manufactured by additive manufacturing and then filled up the SiC matrix from chemical vapor infiltration (CVI). This process can form FCM fuel without damaging the TRISO particle.

In this study, we manufactured the FCM fuel from additive manufacturing and the CVI process.

2. Methods

We manufactured the FCM fuel shell through self-developed 3D binder jet printing (BJT). The particle size distribution, phase, and shape of the SiC powder used in the 3D BJT are shown in Fig. 1. The green SiC powder has an alpha-SiC phase (ICDD card No.: 01-074-1302), and the D₅₀ of the powder is ~63 μm. The FCM fuel process sequence is shown in Fig. 2. First, the FCM fuel shell was designed by 3D Computer-Aided Design (CAD) software. The shell has an empty space inside cylinder to contain TRISO particle. It produced an FCM shell using a 3D BJT. In 3D BJT, SiC powder spread on flat substrate and then polymeric binder drops into the desired layer shapes. The binder is cured by infrared (IR) radiation. This sequential process is repeated several times to form the desired geometry.

Printed parts are obtained by removing loose powder, this called de-powdering. (Fig. 3).

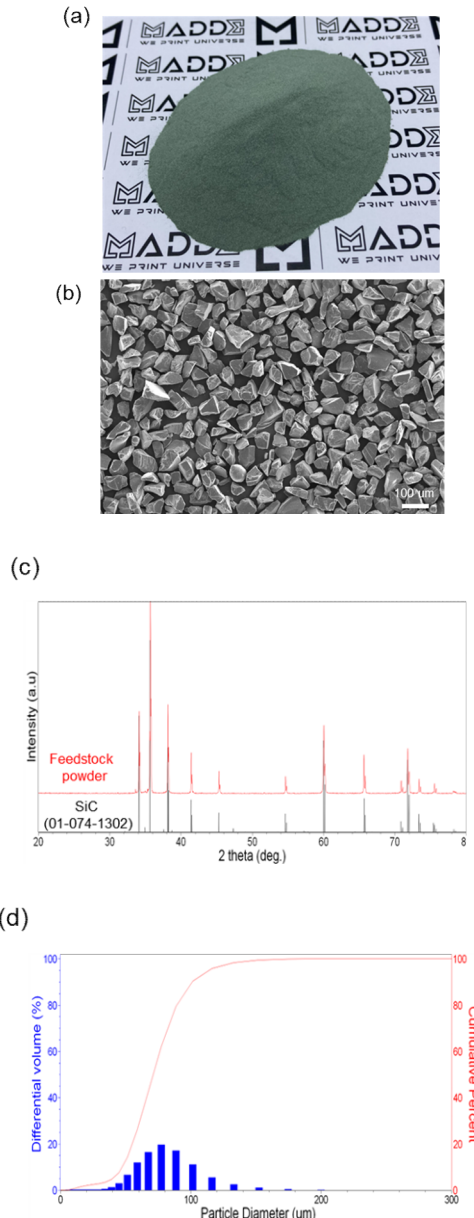


Fig. 1. (a) Green SiC powder used as feedstock for FCM fuel shell, (b) microstructure of SiC powder, (c) X-ray Diffraction pattern of SiC powder, and (d) particle size distribution of SiC powder

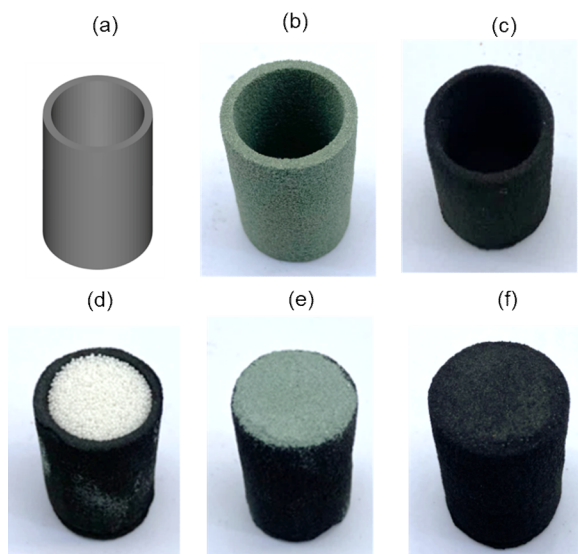


Fig. 2. The FCM process sequence (a) 3D model of the FCM shell, (b) 3D BJT SiC shell, (c) partial densified shell after CVI, (d) shell filled with ZrO_2 ball, (e) cladding with interstitial spaces filled with SiC powder, and (f) FCM after CVI.

The green body, which SiC particle is weakly bonded by the binder, be partially densified by a CVI process to prevent it from breaking during handling. ZrO_2 balls, used as a replacement for TRISO fuel, poured into the partially densified shell. To fill the large void between the ZrO_2 balls, SiC powder of the same size used in 3D BJT was poured on ZrO_2 ball and the filled FCM shell tapped repeatedly. Finally, the SiC matrix was densified by CVI. To densify SiC in the CVI process, a methyltrichlorosilane (MTS, CH_3SiCl_3) precursor containing silicon and carbon, was used, with hydrogen as a dilution gas. The process temperature, MTS/ H_2 ratio, and pressure were fixed at 1150 °C, 1/3, and 50 Torr, respectively.

3. Results

Fig. 4 shows the microstructure of the green body manufactured by 3D BJT. The binder is not only coated on the SiC surface but also bridges the particles to each other. In BJT, the binder wets the particle surface or penetrates between particles by capillary force. After IR radiation, the binder covers the particle surface or connects the particles. The green body, which is weakly bound by the binder, is easily broken during the filling process with ZrO_2 balls or SiC powder. These problems can be solved by performing a partial densification process with CVI.

Fig. 5 shows the microstructure of a FCM fuel shell partially densified by CVI. The SiC formed by CVI appear as protrusions on the particle surface and fill the voids between SiC particles. The binders are removed when the CVI process temperature is reached. The voids between the SiC particles are filled with a SiC matrix during CVI. The SiC matrix increases the bonding strength of FCM fuel shell by filling the empty

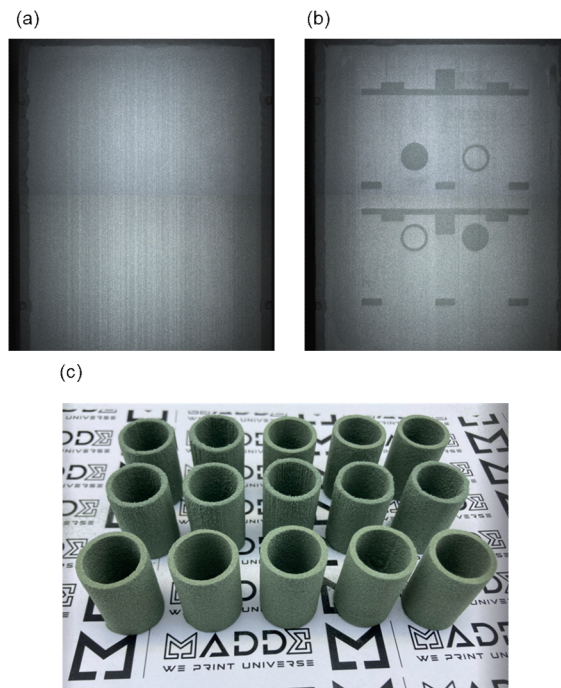


Fig. 3. Method for fabricating FCM shell using binder jet additive manufacturing process (a) after SiC powder spreading, (b) 2D patterning image and (c) after de-powdering.

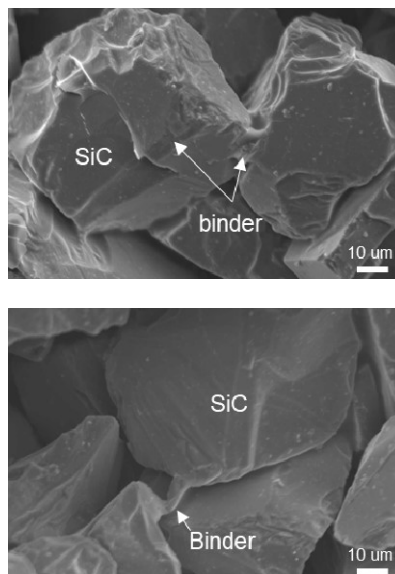


Fig. 4. Microstructure of SiC particles covered and bridged by binder.

spaces and connecting particles. The partially densified FCM shell contains many pores between SiC particles. The pores within the FCM fuel shell provide channels for the MTS precursor to infiltrate into areas filled with ZrO_2 balls and SiC powder, facilitating the formation of a dense SiC matrix inside the FCM fuel. The Fig.6 (a) shows the microstructure of FCM shell formed with SiC matrix by CVI for 20 hours.

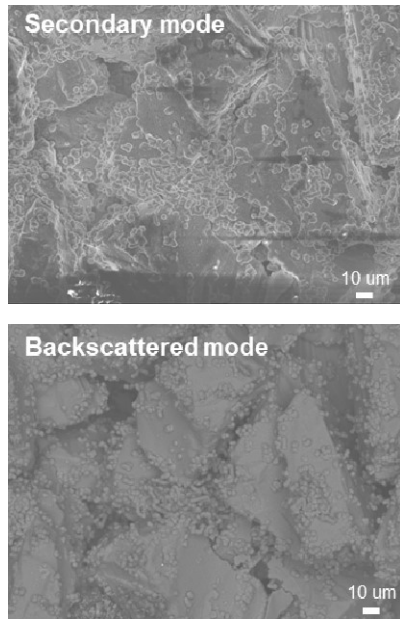


Fig. 5. Microstructure of partially densified FCM shell.

There are regions where a dense SiC matrix was formed by CVI, as well as residual pores. Pores within the FCM shell can be divided into open pores and closed pores. Open pores are formed between widely spaced SiC particles and provide channels for gas precursors to flow into the structure. In contrast, closed pores impede the flow of gas precursors, preventing the formation of a SiC matrix within the FCM fuel. This can block the flow of MTS gas precursor inside FCM fuel. Fig. 6 (b) shows the microstructure of inner FCM fuel. SiC powder is filled between ZrO₂ balls, and there are many pores between SiC particles. It is expected that a dense SiC matrix is formed in FCM shell, preventing MTS from penetrating into the inner region. This result in less filling of the SiC matrix inside FCM fuel. It was confirmed that process conditions need to be controlled to form a dense SiC matrix by CVI.

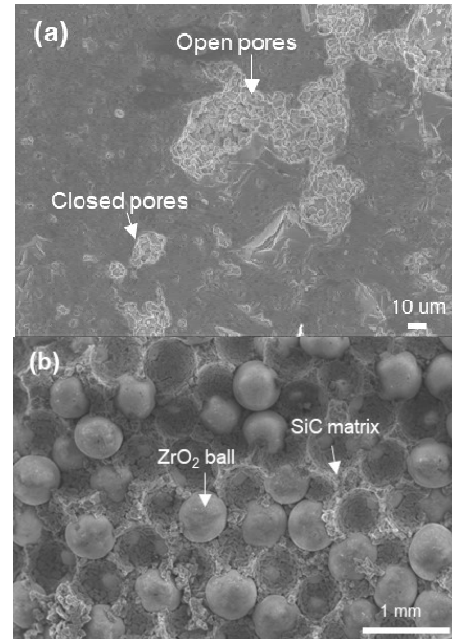


Fig. 6. Microstructure of FCM fuel after CVI densification for 20 hours (a) FCM shell, (b) inner FCM fuel.

4. Conclusions

In this study, an FCM fuel was manufactured from a 3D BJT and CVI process. During the CVI process, the dense SiC matrix in the FCM fuel shell does not allow gas precursors to penetrate deep inside, forming many pores inside. It was confirmed that the CVI process conditions must be controllable to form a dense SiC matrix. In the future, it is expected that 3D BJT will be helpful in manufacturing complex FCM fuel architecture and optimizing CVI process conditions to improve efficiency and manufacture densely SiC matrix FCM fuel.

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