Carbon-Boron Composites for the Canisters in Dry Spent Nuclear Fuel Casks

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1. Introduction

The disposal of spent nuclear fuel is a critical issue in the nuclear industry. Typically, spent nuclear fuel is stored in pools near nuclear reactors. However, the storage capacity is nearly full with increased spent nuclear fuel [1]. The use of dry spent nuclear fuel storage is an option to replace the pools. However, the stainless steel canister in the core of dry cask storage systems has limited properties regarding thermal conductivity and radiation shielding properties [2]. The hinder of emission of heat, α -, and β - radiation from spent nuclear fuel is fundamentally required. Therefore, there is a need to develop materials that can more effectively shield radiation and dissipate heat to safely utilize dry spent nuclear fuel storage system. It is also expected to yield more compact storage facilities

This study aims to investigate the manufacture of boron and graphite composites for use in canisters because the boron has excellent neutron absorption properties and the graphite shows a high-level thermal conductivity, which could contribute to safer and more compact dry spent nuclear fuel cask storage system [3, 4].

2. Experimental procedure

2.1 Manufacture of graphite-boron

For the manufacture of graphite-boron composites, both boron nitride (BN_{HCP}) and boron carbide (B_4C) powders were used in conjunction with graphite powder as the base materials. The graphite powder was mixed with phenol resin to enhance inter-particle adhesion during manufacture process. To investigate the particle size effects on the packing rate and density of composites various particle sizes including 45 µm boron nitride, and 45 µm and 10 µm boron carbide powders were employed. Fig. 1 shows scanning electron microscopies (SEM) image of as received boron carbide and nitride powder. The graphite and boron powders were mixed in a weight ratio of 1:1 with an aid of zirconia balls for uniform mixing for 2 hours. The powder was then pressed at under a pressure of 147~294 MPa (1.5-3.0 ton/cm²) for 1.5 min to produce cylindrical pellets with a diameter of 13.3 mm and height of 10-12 mm. The fabricated pellets were heated 1800°C for 2 hours under vacuum conditions.



Fig. 1. SEM morphologies of (a) BC_10 μ m, (b) BC_45 μ m and (c) BN 10 μ m powder.

2.2 Characterization

After heat-treatment, the weight and volume changes of each pellet were measured to calculate the packing density of the heat treated pellets. The manufactured cylindrical pellets exhibit anisotropy due to the uniaxial compaction method. Therefore, the microstructure and mechanical properties were analyzed according to the direction of compaction. For example, the direction parallel to pressure direction is referred to H-direction, while its perpendicular direction is referred to Vdirection as shown in Fig. 2. Their microstructures were analyzed using optical and scanning electron microscopies (OM/SEM). Additionally, the elemental distribution of the fabricated graphite-boron composites was analyzed using energy dispersive spectroscopy (EDS). Hardness measurements were also conducted to evaluate the mechanical property of heat treated pellets.



Fig. 2. schematic of BC, BN compacts prepared for microstructural and hardness analysis.

3. Results and Discussion

Fig. 3 shows the density changes and packing fractions of the composites before and after heat-treatment. The density of fabricated pellets was decreased after heat treatment, which is attributed to the vaporization of phenol resin added to the graphite powder. As the pressure increased, the density and packing fraction of the heated pellets also increased. BC 10 µm had the lowest density and packing fraction, while BN 45 µm achieved over 80% packing density and fraction after heat treatment. The packing fraction and density changes are affected by particle size and compaction pressure. However, other factors including crystal structure, particle shape, particle distribution, and interactions with other components should be considered. It is also necessary to consider the increase in the surface area as the particle becomes smaller. In particular, boron carbide generally obtains low self-diffusion coefficient, low plasticity, high resistance to grain boundary sliding, and low surface tension. These properties generally hinder the densification of particles [5].



Fig. 3. Before and after heat-treatment of BN_45 μ m and BC_10, 45 μ m: (a) packing fraction, (b) Density.

Fig. 4 shows the microstructure of the composites after heat treatment. BN compacts show intact and smooth microstructures without noticeable pores and defects. Moreover, BN particles are uniformly distributed in the graphite matrix as shown in Fig. 4 (g) and (h). However, the needle-like shaped BN particles were aligned to a direction perpendicular to the pressure direction. Interestingly, BN composite microstructures show different morphology. Surface is rough with various empty spots probably due to separations during mechanical polishing. It is because the packing fraction and density of BN composites were a higher level compared to those of BC 10 µm composite (see Fig. 3). It is noted that BC has identical molecular structure with graphite matrix that may yield similar needle-like shape particles.



Fig. 4. SEM images of microstructures and EDS analysis of elemental distribution after heat-treatment. (a, b) BC_10 μ m H and V-direction under 294 MPa, (c, d) BC_45 μ m H and V-direction under 294 MPa, (e, f, g, h) BN_45 μ m H and V-direction under 294 MPa.

Fig. 5 presents the hardness measurement results. For BC composites, the hardness in the V-direction was higher than that in the H-direction. Anisotropy was observed as uniaxial pressure was applied during the fabrication process of the pre-heated pellets. In the case of BC 10um, the hardness decreased compared to larger particles, attributed to differences in particle packing density. Despite having high density and particle packing fraction, BN 45 µm exhibited the lowest hardness. This result is attributed to the crystal structure of graphite and BN, which consists of a single thin platelet in the hexagonal close-packed (HCP) structure [6]. This lower hardness could be a clue of particle separations during mechanical polishing as already shown in Fig. 4. Therefore, it could be concluded that BC is more attractive candidate for the composite due to a higher compatibility with graphite matrix.

The canister must possess both good mechanical and thermal properties to provide structural supports and dissipate heat generated from spent nuclear fuel. Therefore, we aim to vary particle sizes and investigate different manufacture methods to increase density and packing fraction of composites. Thermal conductivity measurements will be performed using a laser flash instrument.



Fig. 5. Hardness measurement results after heat treatment.

4. Conclusion

In this study, we investigated the manufacture processing for dry cask canister materials, and the following results were obtained:

1) The BC composites obtain higher compatibility with graphite matrix in terms of HCP structures and particle shapes when compared to those of BN. BC composites, therefore, show intact microstructure without particle separations and higher hardness compared to those of BN composites.

2) BC_45 μ m composite has higher packing fraction and density than those of BC_10 μ m composite. It is probably due to the hinderance of graphite matrix densification from smaller particles.

3) Microstructural analysis and hardness measurements revealed anisotropic property in both H and V-directions for BC composites. The influence of the anisotropic properties on the thermal conductivity will be performed.

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