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Fabrication of Boron Nitride Nanotube/Polymer Composites for Neutron shielding

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1. Introduction

Shielding of neutrons, which have higher permeability than alpha and beta rays, is essential for maintaining the health of radiation workers. In general, conventional methods of shielding neutrons have involved using materials that contain large amounts of cadmium, hafnium, boron, or hydrogen elements, which interact strongly with neutrons. However, polymer shields that use high hydrogen content have the disadvantage of being very bulky and heavy, with limited physical properties and stability as a material. Hafnium and cadmium are heavy metals with many restrictions on their use due to their strong toxicity and low annual yield. Although boron is light and has a high neutron absorption rate, its processability is very poor. However, boron nitride nanotubes (BNNTs), which are chemically stable due to bonding between boron and nitrogen, are lightweight and possess high mechanical strength and thermal conductivity (approximately 5000 W/mK). Moreover, they are highly heat-resistant and can maintain a stable shape at temperatures up to 900°C. Additionally, BNNTs have a wide bandgap (5-6 eV) and exhibit electrical insulator properties. [1-7] Therefore, due to their high boron content and neutron absorption rate, BNNTs are ideal for use in neutron shielding applications. As a result, composite materials made with BNNTs exhibit remarkable durability at elevated temperatures, efficient heat dissipation, and effective shielding against neutrons generated by nuclear reactions. Additionally, BNNTs may also serve as a supplementary breeding material for nuclear fusion power generation, owing to their boron content. [8-9]

Agarose, a hydrophilic polymer, forms a structure where each molecule is bonded through hydrogen bonds and chains are entangled. This unique property allows agarose polymer to undergo a sol-gel phase transition with temperature change in aqueous solutions, making it a versatile material for composites with various shapes. [10] To achieve uniform and stable mixing of Agarose, a hydrophilic polymer, and BNNTs with hydrophobic surfaces in an aqueous solution, it was necessary to modify the hydrophobic surfaces of the BNNTs to be hydrophilic. Cetyltrimetylammonium 4-vinylbenzate (CTVB), an amphiphilic surfactant, was used to solve the problem of aggregation in aqueous solution due to the strong van der Waals attraction and hydrophobic surface of BNNTs in aqueous solution. Thus, the surface of BNNTs was modified to be hydrophilic, and a polymerization process was performed using an VA-044 initiator for individual and stable dispersion. Neutron shields of various shapes were fabricated through a sol-gel reaction by mixing hydrophilic modified BNNTs (p-BNNTs) and agarose polymers. Small angle X-ray scattering (SAXS) and optical microscopy (OM) measurements were performed on the prepared Agarose/p-BNNT composite thin film to confirm structural changes and aggregation according to the p-BNNT content. A neutron transmittance measurement experiment was performed to confirm the neutron transmittance according to the increase of the p-BNNT content. As a result of the measurement, it was found that the Agarose/p-BNNT composite exhibits excellent neutron absorption while maintaining a lightweight, which makes it highly processable and formable. The composite also possesses excellent mechanical, electrical, and thermal properties, making it a multifunctional neutron shielding material.

2. Methods and Results

2.1 Sample preparation

In order to solve the problem of strong van der Waals attraction and hydrophobic surface of BNNT, the surface of BNNTs was modified to be hydrophilic using amphiphilic molecules. 0.1 wt % of BNNTs powder and 0.5 wt % of CTVB, an amphiphilic surfactant, were mixed in an aqueous solution, followed by tip sonication for 1 hour to disperse them. Thereafter, VA-044 was injected to prepare hydrophilic surface-modified BNNTs (p-BNNTs) through polymerization. Centrifugation was performed to remove bundled BNNTs. Then, the supernatant was taken out and freeze-dried to obtain p-BNNT powder.

A composite thin film of agarose and p-BNNT was prepared by mixing agarose and p-BNNT, preparing a hydrogel through a sol-gel reaction, and drying for several days. A composite thin film of agarose and p-BNNT was prepared through the following procedure. The p-BNNT powder was added to ultrapure water at 3 to 50 wt % of agarose and dispersed for 1 hour by magnetic stirring at 80 °C. The total concentration of the mixture was fixed at about 2 wt%. The mixture was poured onto a clean quartz plate and dried at room temperature for 2 to 3 days to obtain an agarose/p-BNNT composite thin film.

2.2 Neutron shielding of Agarose/p-BNNT composites

To investigate the structural changes of the Agarose/p-BNNT composite thin film according to the p-BNNT content, a SAXS measurements were conducted (Figure 1a). The SAXS measurements confirmed the presence of interaction peaks resulting from the aggregation of p-BNNTs at 30 and 50 wt% of p-BNNT. Furthermore, as the p-BNNT content increased, the degree of opacity of the thin film increased, and p-BNNT aggregation was observed (Figure 1b).

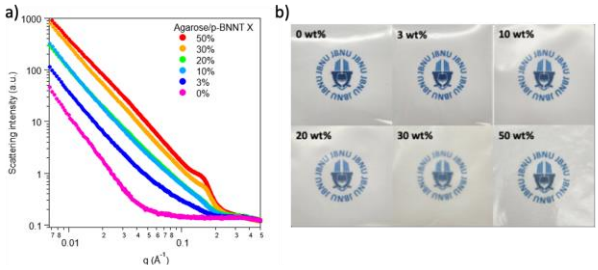

Figure 1. a) SAXS intensities and b) photographs of the Agarose/p-BNNT composite thin film according to the p-BNNT concentration

In addition, OM measurements confirmed the phenomenon of aggregation as a result of increasing p-BNNT content. Agglomerates of hundreds of micrometers were observed at 10 wt% p-BNNT content or higher, while the widest area of agglomeration was seen at 50 wt%. SAXS and OM measurements verified that p-BNNT aggregation occurred when content exceeded 10 wt% .

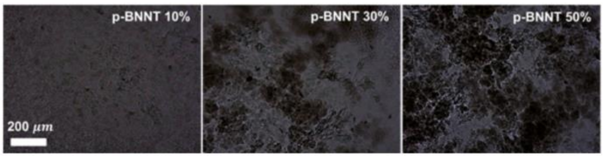

Figure 2. OM images of the Agarose/p-BNNT composite thin film.

By comparing the pure agarose thin film with the Agarose/p-BNNT composite thin film, we measured the change in neutron transmittance as a function of the increase in p-BNNT content. We confirmed that the neutron transmittance tended to decrease with an increase in the thickness of the thin film. Moreover, we observed a relatively greater decrease in neutron transmittance with an increase in p-BNNT content in the composite. (Figure 3a) The linear attenuation coefficients of the Agarose/p-BNNT composite films ranged from 533 ± 0.04 mm^{-1} to 0.765 ± 0.062 mm^{-1} from 0 wt % to 50 wt % p-BNNT content. However, upon comparison with the linear attenuation coefficient value calculated from the neutron absorption table provided by the US NIST, the p-BNNT content exhibited similar values up to 10 wt %. Subsequently, it was confirmed that the linear attenuation coefficient value decreased when the p-BNNT content increased from 10 wt % to 50 wt % and was lower than the theoretically calculated value.

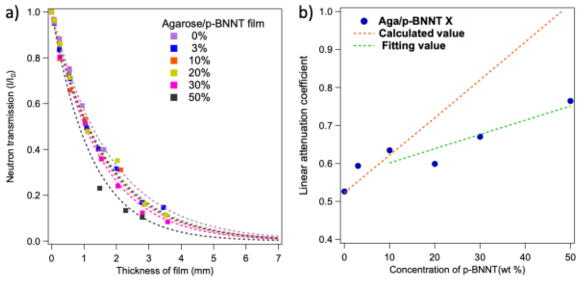

Figure 3. (a) Neutron transmittance (b) Linear attenuation coefficient of agarose/p-BNNT composite thin films with different p-BNNT contents

3. Conclusions

In conclusion, this study exhibits the potential of Agarose/p-BNNT composites for neutron shielding applications. SAXS and OM measurements confirmed that the Agarose/p-BNNT composite thin film formed an agglomerated phase of p-BNNT particles when the p-BNNT content exceeded 10 wt %. However, the neutron transmittance decreased with an increase in p-BNNT concentration, demonstrating the effectiveness of p-BNNT as a neutron shielding material. Overall, this study confirmed the efficacy of Agarose/p-BNNT composites as a neutron shielding material and provides a basis for the formation of hydrophilic polymer complexes through surface modification of one-dimensional nanoparticles.

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