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Designing and preparing for a 288 °C heat resistant and inhibition of gas production resin matrix nuclear radiation shielding material

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ABSTRACT
A resin matrix nuclear radiation shielding material for performing in high temperature is designed and prepared, which consists of epoxy resin, elemental boron, tungsten carbide, and graphene oxide. It was manufactured under room temperature, but it can be serviced below 288 °C, and the gas production could be retained while working. The material will be applied to a land nuclear reactor, transportable nuclear reactor, or macronuclear reactor as a neutron and gamma ray shielding material. The component of the material was designed by a genetic algorithm in combination with the Monte Carlo N-Particle Transport (MCNP) software. Several blocks of the material were prepared. The TG/DTG/DSC (Thermogravimetric analysis, Derivative Thermogravimetry, and Differential Scanning Calorimeter) experiment was performed with the outgas of CO2, NH3, and H2O from the material being observed. The shielding performance of the material was examined by the MCNP software and compared with polyethylene and concrete, which, respectively, work below 90 °C and 60 °C. The scanning electron microscope experiment of the material was carried out, and the results are presented.

I. INTRODUCTION
Neutrons and gamma rays are produced when operating nuclear devices, which are harmful to nuclear workers because ionizing radiation will occur after the interaction of neutrons and gamma rays with the human body. Therefore, neutrons and gamma rays should be absorbed. Because neutrons and gamma rays are electrically neutral, their intensity and energy are difficult to be changed by electric and magnetic fields. One effective way is to interact with the material, so different kinds of shielding material were developed.1,2

Nowadays, many kinds of material, such as polyethylene-based shielding material and concrete, are active. Although polyethylene-based shielding material has been popular all these years,1,2 it fails to satisfy the need of temperature resistance. Furthermore, the main shielding material of the land nuclear reactor,3 concrete, is very heavy and can only work below 60 °C. Hence, a neutron and gamma ray shielding material that is resistant to temperatures as high as 288 °C and inhibits gas production is designed and prepared. The material is shown in Fig. 1, which consists of epoxy resin, boron, and tungsten carbide (WC). The gas release not only brings hidden danger for the material in a closed environment but also takes away matter which will reduce the shielding performance of the material. Therefore, inhibition of gas production of the material is needed. Nowadays, many kinds of material, such as polyethylene-based shielding material and concrete, are active. Although polyethylene-based shielding material has been popular all these years,1,2 it fails to satisfy the need of temperature resistance. Furthermore, the main shielding material of the land nuclear reactor,3 concrete, is very heavy and can only work below 60 °C. Hence, a neutron and gamma ray shielding material that is resistant to temperatures as high as 288 °C and inhibits gas production is designed and prepared. The material is shown in Fig. 1, which consists of epoxy resin, boron, and tungsten carbide (WC).
of neutrons and gamma rays with atoms in the material. Neutrons mainly undergo inelastic scattering, elastic scattering, and absorbing. Because of the good elastic scattering cross section of H, C, and O and good thermal neutron absorbing cross section of B, secondary gamma rays will be produced after the good inelastic cross section of W. Gamma rays mainly undergo the Compton effect, electron-pair effect, and photoelectric effect with different electronics being produced. W and Pb have good gamma ray shielding performance. The covalent bond will break after the gamma rays interact with the electronics which is responsible for its formation. As a result, the mechanical performance and high temperature resistant performance will gradually degenerate.

The chemical structure of graphene oxide is shown in Fig. 3. There are many active groups in graphene oxide, such as the hydroxyl (–OH), epoxy, and carboxyl (–COOH) group. They can react with resin and curing agents. The spatial reticular structure of the material is denser. Indirectly, the high temperature resistant performance and mechanical performance may be improved. Therefore, a resin matrix nuclear radiation shielding material modified by graphene oxide was studied, aiming to obtain a material with good shielding performance, heat resistance, and good mechanical strength.

II. MATERIAL DESIGN AND MANUFACTURE

A. Material design

Each element has a different shielding performance for a different energy neutron or gamma ray. There exists an optimal combination of the element, which makes the material have better shielding performance than that made by others in combination. Therefore, selection and design of the component should be done before preparing the material.
The components of the material are as follows: First, epoxy resin is selected as the matrix material to slow down fast neutrons through elastic scattering. Then, boron (B) is selected as the reinforced phase material because it can absorb thermal neutrons despite secondary gamma ray production. As a consequence, tungsten carbide (WC) is also selected as a reinforced phase material because it can not only slow down fast neutrons through inelastic scattering but it can also shield gamma rays. Finally, graphene oxide is selected as the modified material to improve the temperature resistance of the material.

The intensity and energy of neutrons and gamma rays are complex. When interacting with the atoms in the material, their intensity and energy will change gradually and so will the optimal component. Therefore, a design method was established using a genetic algorithm (GA) in combination with the Monte Carlo N-Particle Transport (MCNP) software on paper. The version is MCNP4C. The objective is to fulfill the lowest dose equivalent of neutrons and $\gamma$-rays at the position of the detector.

The objective function of the method is as follows:

$$\min H(A) = \min[H_n(A) + H_\gamma(A)].$$

The constraint condition of this method is as follows:

$$\sum_{i=1}^{n} A_i = 1,$$

$$\rho_1 < \rho_{\text{eff}} < \rho_2,$$

where $A$ is the mass component matrix in the composite material, a variable vector; $H(A)$ is the total dose equivalent of neutrons and $\gamma$-rays; $H_n(A)$ is the dose equivalent of neutrons; $H_\gamma(A)$ is the dose equivalent of $\gamma$-rays; $\rho_{\text{eff}}$ is the density of the new shielding material; and $\rho_1$ and $\rho_2$ are the vectors of the density range.

The crossover rate and mutation rate of the genetic algorithm were selected reasonably through try calculations. The population size was 200, and the generation was 500.

The flow chart of the method is shown in Fig. 5, with the five steps as follows:

1. Input the parameters expressing the thickness of the shield and the components of the material.
2. Produce the “inpn” file and “inpp” file for simulating the neutron and $\gamma$-ray transmitting in the material.
3. The “inpn” file and “inpp” file are calculated by MCNP, and the “outpn” file and “outpp” file are produced.
(4) Extract the data expressing the dose equivalent of the neutron in the “outpn” file and \( \gamma \)-rays in the “outpp” file.

(5) The program stops when the fitness value is not changed or when the iteration times reach \( N_0 \) (the generation number). If not, a new thickness and component will be produced and then, the next calculation starts.

The optimal component was obtained as shown in Table I. The density of the new shielding material is 2.25 g/cm\(^3\).

### B. Manufacture of the material

There are three main steps in manufacturing the material.

(1) After the pretreatment, mix the resin with the curing agent, WC particles, B particles, and graphene oxide. The components are consistent with the design results shown in Table I.

(2) Stir the mixed material in vacuum to make the particles uniform and reduce the air bubble in the shielding material. This step is supported by ultrasound and some additives as it benefits in making each component mix uniformly and reducing the gas generation when the curing reaction occurs.

(3) Pour the mixed material into a mould under room temperature. Remove the mould after hours, and the sample of the new shielding material is obtained as shown in Fig. 6. In this step, the microwave can be adopted to accelerate the curing of the material. It only takes 2 min to cure the material under microwave radiation.

### III. RESULT AND DISCUSSION

#### A. Thermal property testing result of the material

The TG/DTG/DSC (Thermogravimetric analysis, Derivative Thermogravimetry, and Differential Scanning Calorimeter) experiment of the material was performed from 30 °C to 500 °C. The temperature rising rate was 10 °C/min. The purge gas was nitrogen. The measurement results of the TG/DTG/DSC experiment are shown in Fig. 7. The TG result of the material was the black line. The mass of the resin decreased by 1%, 5.0%, and 10.0% at 213.7 °C, 310.1 °C, and 335.4 °C, respectively.
The decomposition temperature was the point where the tangent at the point of 213.7 °C (1% mass lost) intersects with the other tangent at the point of 310.1 °C (5% mass lost). The decomposition temperature of the resin was around 288.3 °C.

The decomposition temperature of the chemical compound into its constituent elements was 288.3 °C. The point of 10% decomposition loss was 312.0 °C, at which the resin can be used. The red line was the TG result of the material without graphene oxide. The result showed that graphene oxide improved the temperature resistance of the material.

**B. Outgassing property testing result of the material**

To understand the outgassing performance of the material, the TG-IR experiment was performed from 30 °C to 500 °C. The IR results at six different temperatures are shown in Fig. 8. The outgassing performance of the material is shown in Fig. 8. The gas which included water (H₂O) vapor, carbon monoxide (CO), and methane (CH₄) was observed. In Fig. 9, the result of water (H₂O) vapor, CO, and CH₄ was only a tendency, which had no unit. According to the trend result, measures such as adding modified materials could be taken to reduce the gas generation.

**C. Shielding performance testing result of the material**

The shielding performance of the material was simulated by MCNP and compared with concrete and polyethylene. Figure 10 shows the result of the epoxy resin based material, concrete, and polyethylene. Figures 10(a)–10(d) show the results of the shielding performance result of neutrons, secondary gamma rays, primary gamma rays, and neutrons and gamma rays together, respectively. According to the results, polyethylene had good neutron shielding performance. Resin had good gamma ray shielding performance.
FIG. 10. Shielding performance of the material. (a) Dose of neutrons; (b) dose of secondary gamma rays; (c) dose of primary gamma rays; (d) dose of neutrons and gamma rays together.

When the thickness of the material was less than 20 cm, the total shielding performance of the resin matrix material and polyethylene was close. When the thickness of the material exceeded 20 cm, the total shielding performance of the resin matrix material was better than that of polyethylene.

D. The scanning electron microscope (SEM) experiment

Figure 11 shows the scanning electron microscope (SEM) experiment of the material. In the blue circle, there were enhanced phase particles. In the red circle, there were some bubbles. When

FIG. 11. SEM result of the material. (a) Result at position one; (b) result at position two.
preparing the material, no vacuum was evacuated, and exothermic reaction occurred during the curing process. Therefore, bubbles were produced. This reduces the shielding performance of the material.

IV. CONCLUSION

A new composite nuclear radiation shielding material for in situ casting is developed. Its features are summarized as follows:

(a) The material consists of epoxy resin, elemental boron, tungsten carbide, and graphene oxide. It can be manufactured under room temperature and fit for in situ casting.
(b) The shielding performance of the material is better than that of concrete and polyethylene.
(c) The outgassing property is observed in the experiment. The tendency of the outgassing H₂O vapor, CH₄, and CO is measured.
(d) The thermal property is tested, and the resolved temperature is 288.3 °C.
(e) Graphene oxide improves the temperature resistance of the material.

It can be concluded that this material will be a good alternative as an in situ casting shielding material.

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