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Development of 300 °C heat resistant boron-loaded resin for neutron shielding

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Abstract

A new neutron shielding material resistant to temperatures up to 300 °C is developed, consisting of a phenol-based resin with 6 wt% boron. The resin will be applied around the vacuum vessel of the DD plasma device to suppress the streaming neutrons and to reduce the nuclear heating of the superconducting coils. The neutron shielding performance of the newly developed resin, examined by the ^{252}Cf neutron source, is almost the same as that of polyethylene, which is not effective above 100 °C. The new resin maintains its mechanical strength in the high temperature region. The outgas of CO_2, NH_3 and H_2O from the resin have been measured, however, the neutron shielding performance of the resin after 200 °C baking was almost the same as that before baking. Thirteen kinds of organic gases have been observed at ~300 °C.

1. Introduction

Through a nation wide collaboration in Japan the National Centralized Tokamak (NCT) program will demonstrate the production of a steady-state high-β plasma. The recent design of the NCT facility is described in Ref. [1]. The NCT will be operated with deuterium plasmas and a deuterium beam, but no blanket is planned. Therefore, some shielding structure is required for the DD neutrons ($E_n = 2.45$ MeV) in order to suppress nuclear heating at the superconducting coil. The DD neutrons will be shielded mainly by the water in the double walled vacuum vessel. However at the port duct where the double wall structure is not available, another neutron shield material will be required. Such a neutron shielding material is required to be resistant to the baking temperature of the vacuum vessel of 150–300 °C. The neutron shielding material such as resin will be installed outside the vacuum vessel of the NCT between the port wall and the superconducting coils. The required temperature range is higher than the heatproof temperatures of presently available neutron shielding resins, e.g. 90 °C in polyethylene, 150 °C in KRAFTON [2,3] and 200 °C in EPONITE

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[4]. Therefore, a boron-loaded resin, which is heat resistant to 300 °C has been developed as a neutron shielding material.

2. Design of neutron shielding resin

To decrease the nuclear heating it is essential to attenuate the DD neutrons as well as the secondary γ-rays emitted from the recoil nucleus due to elastic scattering. For effective neutron attenuation the shielding material should be both a 'moderator' and an 'absorber'. That is fast neutrons should be thermalized at first by the 'moderator' through elastic scattering, and then absorbed by the 'absorber'. The most effective 'moderator' is hydrogen because it has almost the same mass as the neutron. The 'absorber' should be selected so as to minimize the effects of capture γ-ray emission during absorption of the thermalized neutron. Boron is effective as the 'absorber'.

To decide the amount of boron to be included in the neutron shielding material for NCT, the neutron and the γ-ray flux through the resin were evaluated using the 1D model analysis code, ANISN [5]. The neutron and γ-ray fluxes were calculated using the group constant set FUSION-40 [6], which consists of 42 neutron and 21 γ-ray groups, based on JENDL3.1 [7]. Multiplying these fluxes by the KERMA (Kinetic Energy Released in MAterials) factors [8] based on FUSION-40, the nuclear heating at the TF coil of NCT was obtained. In the calculation of radiation shielding and nuclear heating, the neutron emission rate of DD neutrons of 4×10^{17} n/s and DT neutrons of 1.2×10^{16} n/s (3% of DD neutrons) are defined [9]. DT neutrons fraction is estimated by the result on JT-60U [10].

Fig. 1 shows the calculated neutron flux and γ-ray flux at the front surface of the TF coil as a function of the boron density in the designed resin with 8 cm thickness. The γ-ray flux is drastically reduced with increasing boron concentration in the range of 0–2 wt%. On the other hand, the neutron flux decreases only slightly in the range of 0–9 wt% B. At 10 wt% B, the total neutron flux increases significantly due evidently to decreasing hydrogen in the resin as the amount of the boron increased.

Fig. 2 shows the calculated nuclear heating rates at the front surface of the TF coil as a function of the boron concentration. The total nuclear heating, where the γ-ray heating is dominant, is drastically reduced with increasing boron concentration in the range of 0–2 wt%. In the range of 2–10 wt% of boron, the total nuclear heating is almost constant. The nuclear heating efficiency, indicated by the KERMA factor, depends on the capture γ-ray energy (E_γ). Compared to the hydrogen ($E_\gamma=$ 2.225 MeV), boron has much lower capture energy ($E_\gamma=0.48$ MeV). The nuclear heating at the superconducting coil is generated mostly by the copper wire of the cable in the conduit. The KERMA factor of copper in the γ-ray by hydrogen is about three times larger than that of boron. Thus boron doping is an effective method to decrease the nuclear heating of the coil. For these reasons, the amount of boron was chosen to be 5–9 wt% to minimize the nuclear heating of the superconducting coil.

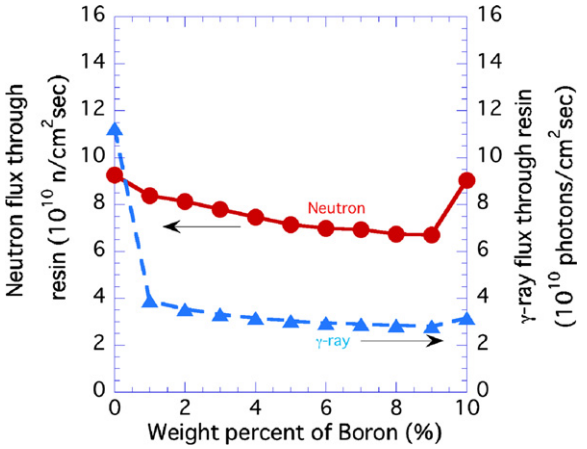

Fig. 1. Neutron and γ-ray flux through the trial product resin (250WD).

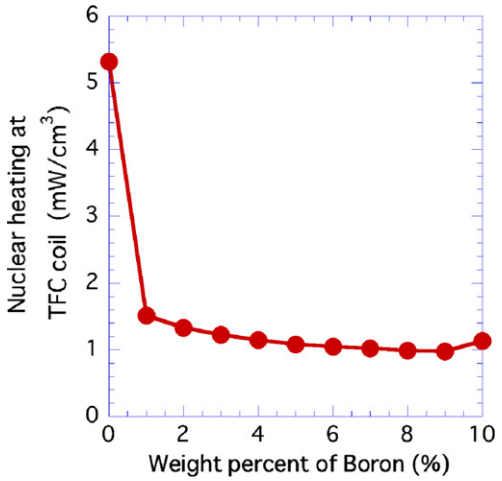

Fig. 2. Nuclear heating rate of the TF coil (250WD).

3. Characteristics of the newly developed resin

We have applied these findings and current technology to produce high heat resistance resin for neutron shielding. Also, it is important to understand the mechanical characteristics and the neutron shielding performance in the high temperature region.

3.1. Making of test pieces of the resin

Ten kinds of test materials were produced. Six consist of epoxy-based resin, two consist of the glass fiber, one consists of polyurethane, and the last one consists of phenol-based resin. Only the phenol-based resin and the glass fiber are useful above 250 °C. From a viewpoint of the cost, further improvement of the phenol-based resin was carried out.

In the next step, the purpose of mixing the boron is to capture the thermal neutron. At first, we tried to mix boric acid into the resin. However, it was difficult to produce neutron shielding material having more than 5 cm thickness. So, the neutron shielding resin was developed by mixing boron carbide (B_4C) with phenol-based resin that has improved heat resistance. The measured chemical compositions of the newly developed resin is summarized in Table 1. The density of the resin was 1.8 g/cm^3.

3.2. Mechanical characteristics and heat resistant temperature

The mechanical characteristics and heat resistant temperature of the developed resin are summarized in Table 2. The tensile strength, bending strength, and compressive strength were measured at room temperature and 250 °C by the examination method of the Japanese Industrial Standard (JIS). It was confirmed that the developed resin has enough mechanical strength at 250 °C. The heat resistance temperature was more than 300 °C by the determination of the temperature of deflection under load on the JIS.

Table 2
Mechanical characteristics of the developed resin (250WD)

	Room temperature (23 °C)	250 °C
Tensile strength (MPa)	83.9	38.5
Bending strength (MPa)	139	65.9
Compressive strength (MPa)	300	135

3.3. Neutron shielding performance

The neutron shielding characteristic was measured by neutron penetration tests with a ^{252}Cf neutron source (0.3 MBq, average energy: $\langle E_n \rangle \sim 2.1$ MeV) on samples ($40 \times 40 \times 5$ cm^3) of the developed resin and polyethylene at room temperature (~20 °C). The neutron penetration was measured using a neutron rem counter (ALNOR 2202D). The number of penetration neutrons was measured by pulse counting for the neutron source. Fig. 3 shows the neutron dose attenuation of the developed resin and the polyethylene as a function of the thickness of the sample. The neutron shielding performance of the resin is almost the same as that of the polyethylene. The neutron shielding characteristic was also estimated by the 3D Monte Carlo Code MCNP-4C2 [11] using the continuous energy cross section data sets based on the JENDL-3.2 [12] for the composition of the developed resin based on Table 1. The experimental analysis was faithfully modeled by the 3D calculation. The neutron flux through the surface in the center part of the detector was calculated. The neutron dose was estimated by multiplying the conversion coefficient of the calibrated rem counter. The calculation result agrees well with the experimental result as shown in Fig. 3.

4. Out gas property at high temperature

When resin is used at the high temperature region, it is important to assess the amount and the element of the outgas from resin. The H_2O is one of the outgas to shield as for the neutron. The

Table 1
Chemical composition of the developed resin (250WD)

Element	C	H	N	O	B	Ca	Al	Si
Contents (wt%)	29.4	1.94	0.31	30.4	6.1	7.4	6.8	14

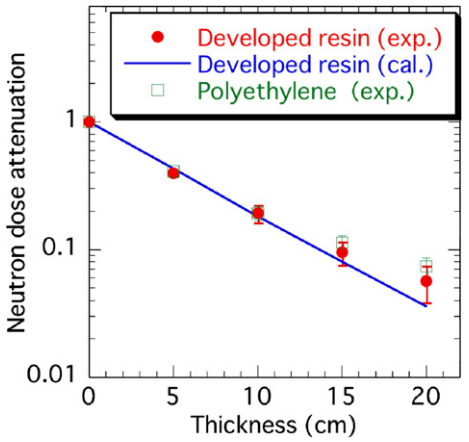

Fig. 3. Neutron dose attenuation in boron-loaded resin and polyethylene (250WD).

organic gas that influences the human body might be included in the outgas.

4.1. TG/DTA analysis

To understand the amount of the outgas from the developed resin in the high temperature region, thermogravimetric (TG)/differential thermal analysis (DTA) was applied to the resin. The temperature was from 20 °C to 300 °C. The temperature rise rate was 10 °C/min. The purge gas was nitrogen. The initial weight of the resin was 21.5 mg, and the weight of the resin decreased by 1.1% at ~300 °C.

4.2. TDS analysis

To identify the gases released from the developed resin in the high temperature region, the mass spectrum of the outgas was measured at 20 °C, 50 °C, 100 °C, 150 °C, 200 °C, 250 °C and 300 °C by thermal desorption spectroscopy (TDS). The observed mass numbers were 2, 17, 18, 28, 32, and 44, which corresponds to hydrogen (H_2), ammonia (NH_3), water (H_2O), carbon monoxide (CO), oxygen (O_2), and carbon dioxide (CO_2), respectively. The main outgas components from the resin at 100–150 °C were NH_3 and H_2O. The measured amounts of NH_3 and H_2O are 2.692 l/kg, 2.698 l/kg, respectively. H_2O is an essential element in order to shield the neutron, however, the neutron shielding performance of the resin after 200 °C baking was almost the same as that before baking. In a second baking, the outgases from the resin were less than the limitation of identification.

4.3. TPD–GC/MS analysis

The quantitative analysis of the outgas from the resin in the high temperature region was done by the temperature programmed desorption (TPD)–gas chromatography and mass spectrometry (GC/MS). For the first time, the outgas generated within the range of temperatures from 20 °C to 150 °C was gathered by a cold trap. It was maintained for 1 h at 150 °C. The gases CO_2, NH_3, and H_2O were observed. In the second time, the outgas generated within the range of temperatures from 150 °C to 300 °C was gathered by a cold trap. It was maintained for 1 h at 300 °C. The gases CO_2, NH_3, H_2O and the 13 organic gases listed in Table 3 were observed in very small amounts of μg/g by GC/MS measurements. Thus we have confirmed that the very small amount of organic gases emitted at ~300 °C will have little effect when the area of the newly developed resin is small, such as in the NCT.

5. Simulation of neutron penetration at 300 °C

The neutron shielding performance of the developed resin at 300 °C was simulated by 3D analysis. The resonance cross section of the nucleus is broad at the high temperature region by the Doppler

Table 3
Detected organic compound of the developed resin by GC/MS (500WD)

	Detected organic compound	Retention time (min)	μg/g
1	Pyridine	5.53	3.9
		5.84	
		6.17	
2	Unknown	7.26	0.76
3	Pyridine, 2-methyl	7.91	12.9
		9.51	
4	Formamide, *N*-methyl	8.58	3.3
5	Acetamide, *N*-methyl	9.91	4.8
6	Phenol	10.6	13
		10.83	
7	Pyridine, 2,5-dimethyl	11.2	3.4
		12.57	
8	Phenol, 2-methyl	12.32	21.7
		12.46	
		12.78	
9	Phenol, 2,5-methyl	13.83	9.4
		14.29	
10	Phenol, 2,3,5-trimethyl	15.68	1.9
11	1.4-Benzenedicarbonitrile	17.38	1.8
12	Hexadecanenitrial	25.79	7.4
13	Octadecanenitrial	27.97	6.6

effect. The cross section of the temperature dependent library for MCNP based on JENDL-3.2 is prepared at 327 °C [13].

The 3D analysis result by using the library at 327 °C agrees with the experimental result of the developed resin as shown in Fig. 3. The calculation results using the 327 °C library and the 20 °C library are almost the same.

6. Conclusion

A new 300 °C heat resistant neutron shielding resin has been successfully developed. Its features are summarized as follows:

- The neutron shielding resin was produced by mixing of boron carbide (B_4C) and phenol-based resin.
- The neutron shielding is almost the same as that of the polyethylene as measured with a ^{252}Cf neutron source.
- Outgasing of CO_2, NH_3, H_2O and 13 kinds of organic gases have been observed at ~300 °C temperature regions.

It can be concluded that this high heat resistance resin for neutron shielding is very attractive as a neutron shielding material.

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