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Enhanced thermal properties of epoxy composite containing cubic and hexagonal boron nitride fillers for superconducting magnet applications

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Abstract

Superconducting test coils impregnated with epoxy composites containing cubic boron nitride (cBN) particles, hexagonal boron nitride (hBN) particles, and a mixture of cBN/hBN particles were fabricated, and their thermal and electrical properties were investigated using cool-down, over-current, and repetitive-cooling tests. Micro-voids, which may act as major obstacles to the formation of thermally conductive passages, were observed in the epoxy composites containing the cBN or hBN particle fillers alone but were absent in that containing the cBN/hBN particle mixture. The coil impregnated with epoxy containing the cBN/hBN particle mixture also exhibited superior cooling performance and thermal/electrical stabilities, indicating that this composite effectively facilitated heat transfer between the coil and liquid nitrogen. Moreover, the addition of the cBN/hBN filler reduced the difference in thermal contraction between the superconducting tape and epoxy composite. Overall, the use of the epoxy composite containing the cBN/hBN filler shows potential for the development of highly stable superconducting coils with considerably enhanced thermal conductivity and low coefficients of thermal expansion.

Keywords:

A. Polymer-matrix composites (PMCs)

A. Resins

B. Thermal properties

B. Electrical properties

Boron nitride filler

1. Introduction

Epoxy resins are commonly used as adhesives, coatings, and structural materials in electronic industries because of their superior mechanical properties, easy processing, low cost, and excellent moisture/corrosion resistance. Despite these advantages, their poor thermal properties, including their high coefficient of thermal expansion (CTE) and low thermal conductivity, limit their wide application. Consequently, the inclusion of inorganic fillers is necessary to create new epoxy resin composites with desired properties [1-9].

In superconducting magnet applications, particularly for rotating machines such as motors and generators, epoxy resins are used to encapsulate the superconducting magnet to provide high mechanical reliability and dynamic stability. However, because the epoxy-impregnated magnet virtually experiences a cool-down and operation sequence or occasionally even an unexpected fault, an epoxy resin with lower CTE and higher thermal conductivity and mechanical strength is preferred [10-15].

Although continuous efforts have been made to improve the physical properties of epoxy resins by adding ceramic fillers to them [16-19], compatible epoxy-filler combinations that can resist cryogenic conditions and exhibit superior thermal and mechanical properties should be developed to effectively realize the practical applications of superconducting coils impregnated with epoxy/filler composites. Hence, much research has been focused on the development of advanced epoxy composites containing ceramic fillers for use in superconducting magnet applications [20-22].

Recently, we reported the effects of the addition of ceramic fillers such as boron nitride (BN) and aluminum nitride (AlN) to epoxy composites on the thermal and electrical characteristics of the resulting superconducting magnets [22]. This earlier study demonstrated that the addition of BN filler (i.e., epoxy/BN composites) yielded superior cooling performance as well as enhanced thermal and electrical stabilities for the magnets because of the higher thermal conductivity and lower CTE of the BN filler. However, for using epoxy/BN composites as impregnation materials for the development of highly stable superconducting coils with enhanced thermal properties, using BN particles with an appropriate shape is a pre-requisite.

In this study, three superconducting magnets impregnated with epoxy composites containing cubic BN (cBN) particles, hexagonal BN (hBN) particles, and a mixture of cBN and hBN particles were fabricated, and their thermal and electrical characteristics were investigated using conducting cool-down, over-current, and repetitive-cooling tests. Furthermore, the composition and morphology of the epoxy composites were analyzed using X-ray diffraction (XRD) and scanning electron microscopy (SEM).

2. Experimental setup

2.1. Coil construction and instrumentation

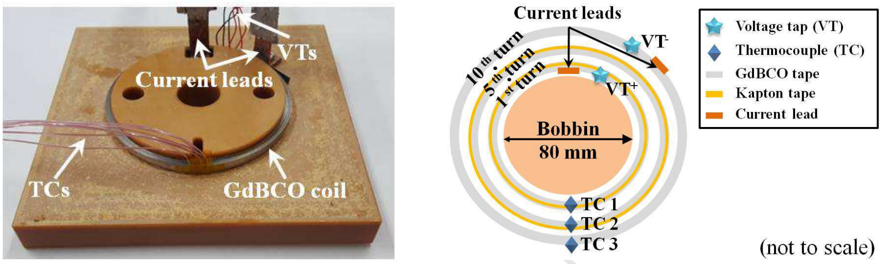
Table 1 lists the specifications of the superconducting test coil. $GdBa_2Cu_3O_7$ (GdBCO)-coated conductor (GdBCO CC) tape, a so-called second-generation high-temperature superconductor, was wound onto a fiber-reinforced Bakelite bobbin with an outer radius of 40 mm. The inner and outer diameters of the GdBCO test coil with Kapton tape as turn-to-turn insulation were 80 and 91 mm, respectively.

Fig. 1 presents a photograph of the GdBCO test coil before epoxy impregnation and a schematic drawing of the instrumentation consisting of the array of E-type thermocouples (TCs) and voltage tap (VT). The temperature profiles of the test coil during cooling testing were obtained by installing three TCs on the: 1) innermost layer (TC 1), 2) 5^{th} layer (TC 2), and 3) outermost layer (TC 3). The terminal voltage of the test coil was measured by installing the VT at both ends of the coil.

Table 1

Specifications of superconducting test coil.

Parameters		Values
Superconductor		GdBCO-coated conductor tape
Total conductor length used	[m]	2.7
Number of turns		10
Inner diameter	[mm]	80
Outer diameter	[mm]	91
Insulation material		Kapton tape
Insulation width; Thickness	[mm]	4.1; 0.05



(a) (b)

Fig. 1. Photograph of the GdBCO test coil before epoxy impregnation (a) and a schematic drawing of the instrumentation consisting of the array of E-type TCs and VT (b).

2.2. Procedure for epoxy composite preparation

Table 2 lists the specifications of the epoxy resin and filler materials [23-24]. In this study, Stycast 2850 FT manufactured by Emerson and Cuming Company was used as the epoxy resin for the impregnation of the GdBCO test coil. The thermal conductivity and CTE of Stycast 2850 FT were 1.15 W/m·K and 111.5 ppm/K, respectively. To improve the physical properties of the epoxy resin, we fabricated three lab-made epoxy composites: an epoxy/cBN composite, epoxy/hBN composite, and epoxy/cBN–hBN composite. The average size of the cBN and hBN particles was ~ 1 μm, and the thermal conductivities and CTE values for cBN and hBN were 1300 W/m·K, 4.8 ppm/K and 300 W/m·K, 0.5 ppm/K, respectively.

Table 2

Specifications of the epoxy resin and filler materials.

Parameters		Stycast 2850 FT	
Company		Emerson and Cuming Co.	
Thermal conductivity	[W/m·K]	1.15	
CTE	[ppm/K]	111.5	
Parameters		cBN	hBN
Molar mass	[g/mol]	40.99	24.82
Average particle size	[μm]	~ 1	
Shape		Cubic	Hexagonal
Thermal conductivity*	[W/m·K]	1300	300
CTE	[ppm/K]	4.8	0.5

*obtained at 293 K

The epoxy composites were fabricated using the following procedure: 1) Stycast 2850 FT (100 g) was mixed with 5 g of filler (for the epoxy/cBN–hBN composite, a mixture of cBN (2.5 g) and hBN (2.5 g) was used); 2) cure agent (Catalyst 23 LV) was added to the composites at room temperature; 3) the epoxy composites were stirred using a Thinky Mixer ARE-310 (THINKY Co.) for 20 min; and 4) the epoxy composites were degassed for 30 min [19].

2.3. Coil impregnation

Fig. 2 shows a schematic cross-sectional view of the test coil impregnated with the epoxy composite. To ensure an identical thickness of the epoxy encapsulation for each test coil, 7.1-mm-high Cu foil was installed with a gap of 3 mm from the outer radius of the coil, as illustrated in Fig. 3. The empty space (the 3-mm-gap) between the GdBCO coil and Cu foil was filled with the epoxy composite, and it was cured for 24 h at room temperature. The specifications of the epoxy-impregnated test coils are listed in Table 3. The critical currents (I_c)

of the coils were measured at 77 K using a 1 μV/cm criterion and an I_c of 122 A was observed for all the three coils. The currents and voltages were monitored and recorded using a data acquisition (DAQ; NI PXI- 1033, National Instruments Co.) system during the measurements.

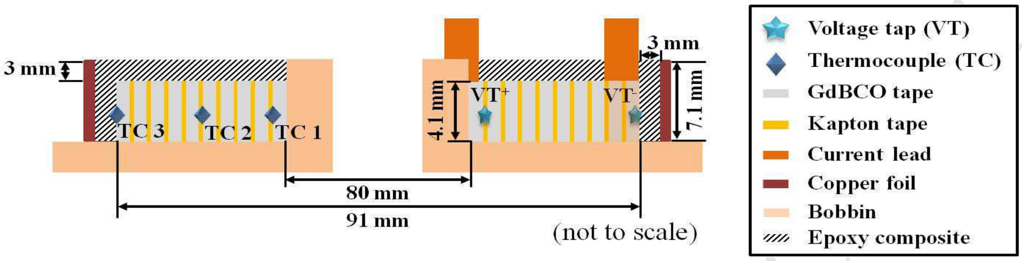


Fig. 2. Schematic cross-sectional view of the test coil impregnated with the epoxy composite.

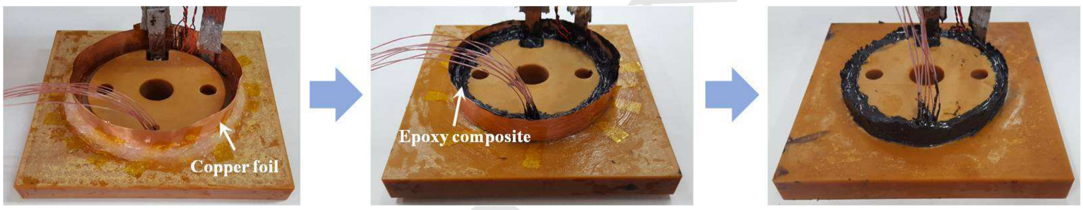


Fig. 3. Photographs of the test coil during impregnation procedure.

Table 3

Specifications of epoxy-impregnated test coils.

Parameters		Coil 1	Coil 2	Coil 3
Critical current @ 77 K	[A]		122	
Epoxy			Stycast 2850 FT	
Cure agent			Catalyst 23 LV	
Cure temperature			Room temperature	
Cure time	[h]		24	
Filler material		cBN	hBN	cBN–hBN
Filler content	[wt. %]		5	

3. Results and discussion

The composition and morphology of the three epoxy composites were analyzed using X-ray diffraction (XRD; SmartLab, Rigaku Co.) and scanning electron microscopy (SEM; Quanta 250 FEG, FEI Co.). Fig. 4 presents XRD patterns of the three epoxy composites used as impregnating materials in this study. The major diffraction peaks of carbon black, epipropidine, butyl 2, 3- epoxypropyl ether, and alumina, which are components of the Stycast 2850 FT epoxy resin, are observed in all three patterns. In addition, the cBN peak is observed at 2θ = 43.3° in Fig. 4 (a) for the epoxy/cBN composite, the hBN peak is observed at 2θ = 26.6° in Fig. 4 (b) for the epoxy/hBN composite, and both the cBN and hBN peaks are observed in Fig. 4 (c) for the epoxy/cBN–hBN composite.

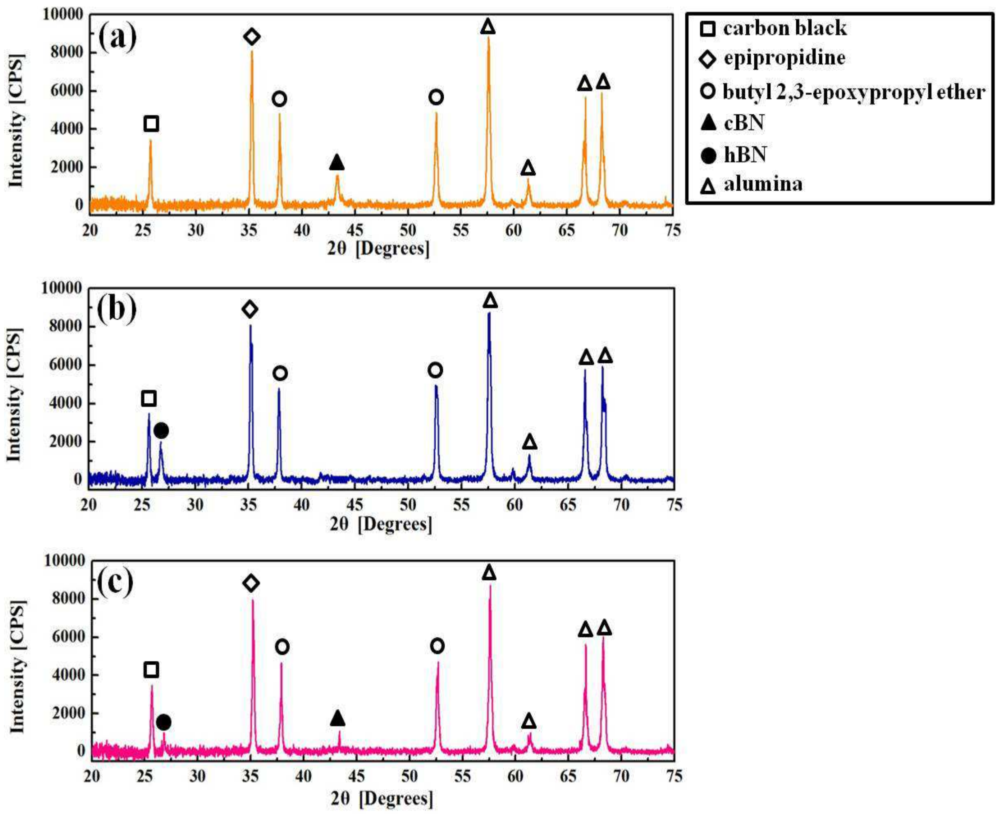
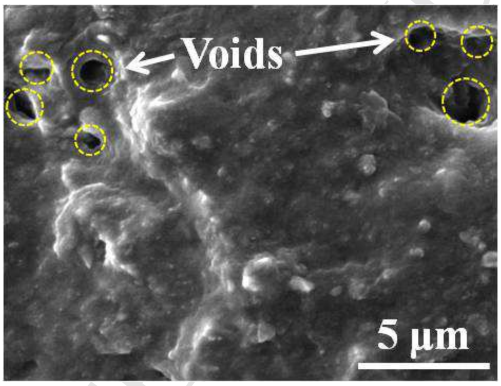
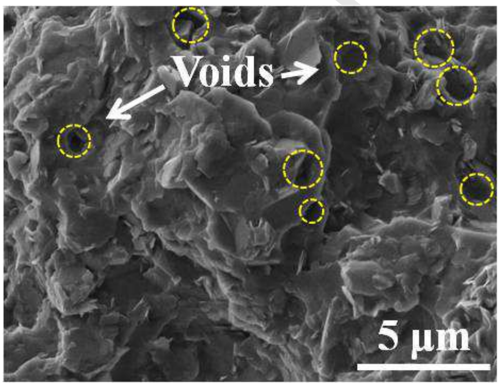


Fig. 4. XRD patterns: (a) epoxy/cBN, (b) epoxy/hBN, and (c) epoxy/cBN–hBN composites.

Fig. 5 presents SEM images of the three epoxy composites. In Figs. 5 (a) and (b), micro-voids with a lateral size ranging from 0.5 to 1.5 μm are observed. These micro-voids were attributed to the high viscosity of the epoxy resin, which could not perfectly permeate into the space between the particles. In general, the voids obstruct thermally conductive passages in the matrix, leading to a decrease in the thermal conductivity of the epoxy composites [19]. This may lead to heat accumulation within the superconducting coil, which is locally burned-out, or even cause irreversible damage to the coil. However, the SEM image for the epoxy/cBN–hBN composite does not contain any micro-voids (see Fig. 5 (c)) because the hBN particles effectively filled the voids produced between cBN particles [17-19].



(a)



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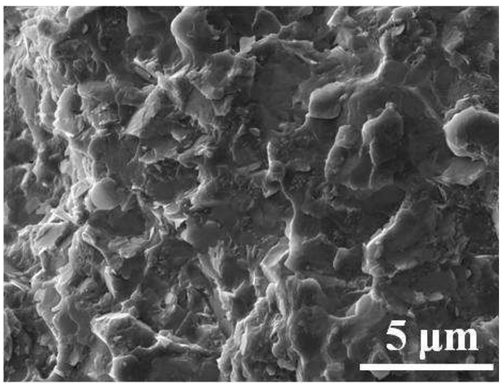
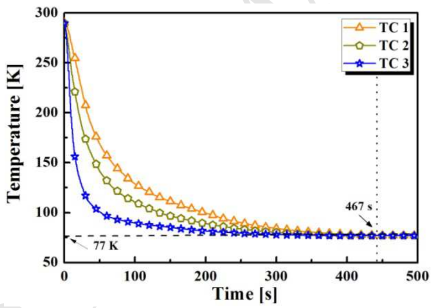


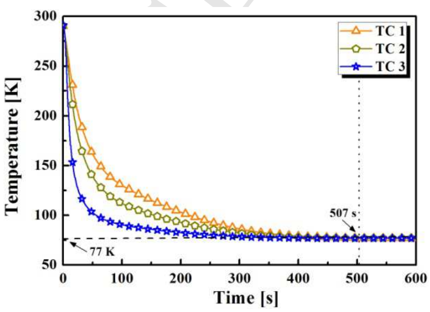
(c)

Fig. 5. SEM images of epoxy/cBN (a), epoxy/hBN (b), and epoxy/cBN–hBN composites(c).

To investigate the cooling performance of the test coils, cool-down tests were performed in a liquid nitrogen (LN_2) bath (77 K). Fig. 6 presents the temperature traces of the test coils obtained during the cool-down from room temperature to 77 K. As soon as the coils were immersed in the LN_2 bath, the temperatures of all the coils started to decrease drastically and eventually reached 77 K. The times required to reach 77 K at TCs 1, 2, and 3 for coils 1, 2, and 3 were 467, 407, and 372 s; 507, 476, and 412 s; and 343, 270, and 245 s, respectively. Thus, coil 3 exhibited faster cooling times than coils 1 and 2, indicating that the epoxy composite containing the mixture of cBN and hBN particles facilitated heat transfer between LN_2 and the coil. This effect occurred because of the absence of micro-voids, as observed in the SEM images. In other words, the micro-voids observed in the epoxy composites for coils 1 and 2 may have acted as major obstacles for the formation of thermally conductive passages within the epoxy composite, resulting in the poor thermal conduction.



(a)



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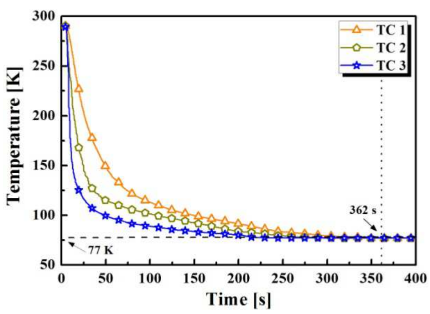


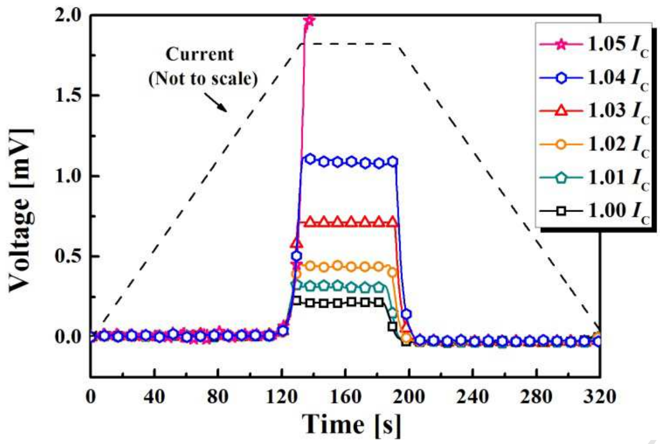
(c)

Fig. 6. Cooling test results of coil 1 (a), coil 2 (b), and coil 3 (c) obtained during cool-down from room temperature to 77 K.

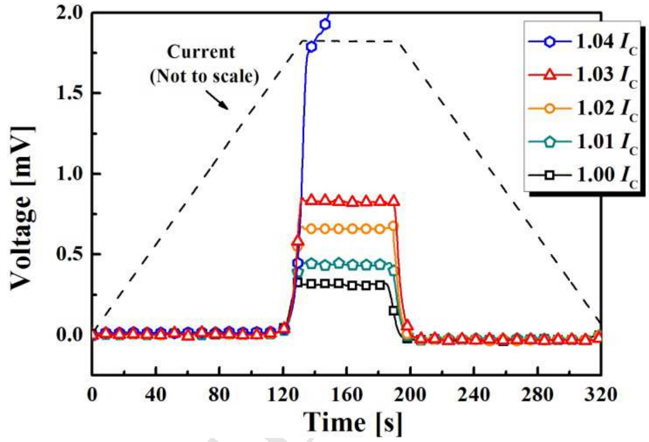
To examine the thermal and electrical stabilities of coils 1, 2, and 3 in excessive current conditions, over-current tests were performed. In the over-current tests, the operating current (I_{op}) was increased at a charging rate of 1 A/s and maintained at each target current for 60 s before being reduced to 0 A at a discharging rate of -1 A/s. When the terminal voltage reached 2.0 mV, the test was stopped to prevent damage to the coils.

Fig. 7 presents the over-current test results for the test coils. As observed in Fig. 7 (a), when the operating current of 122 A (i.e., $I_{op} = I_c$) was applied to coil 1, the total voltage initially started to increase at 117 s, reached a maximum value of 0.20 mV at 126 s, and was then stably maintained for 60 s, indicating thermal equilibrium between the LN_2 cooling and Joule heat induced by the quench. Thereafter, as I_{op} decreased, the total voltage also decreased and finally reached zero. The test results at $I_{op} = 1.01I_c$, $1.02I_c$, $1.03I_c$, and $1.04I_c$ showed similar behaviors as those obtained at $I_{op} = 1.00I_c$. The voltages started to increase abruptly, reached their maximum values, and finally decreased to zero. However, when I_{op} was further increased to $1.05I_c$, the voltage continuously increased, indicating the occurrence of a thermal runaway phenomenon that might cause permanent damage to the coil. As shown in Figs. 7 (b) and (c), thermal runaway phenomena were observed for coils 2 and 3 during the tests at $1.04I_c$ and $1.09I_c$, respectively. These results indicated that the thermal runaway current for coil 3 (i.e., 133 A) was higher than those for coil 1 (128 A) and coil 2 (127 A) because of the enhanced thermal conductivity of the coil impregnated with the epoxy/cBN–hBN composite, which allowed hot spots to be dissipated more effectively in the event of a quench. These results confirm that the epoxy/cBN–hBN composites could improve the thermal and electrical stabilities of the coil because the addition of the mixture of cBN/hBN particles inhibited the formation of voids, resulting in superior thermal conduction within the coil.

To investigate the effect of BN filler additions on the thermal contraction between the GdBCO tape and epoxy composites, repetitive-cooling tests for the coils were conducted using the following procedure: the coil was cooled to 77 K, the I_c value of the coil was measured, and then, the coil was heated to room temperature. These steps were repeated 25 times.



(a)



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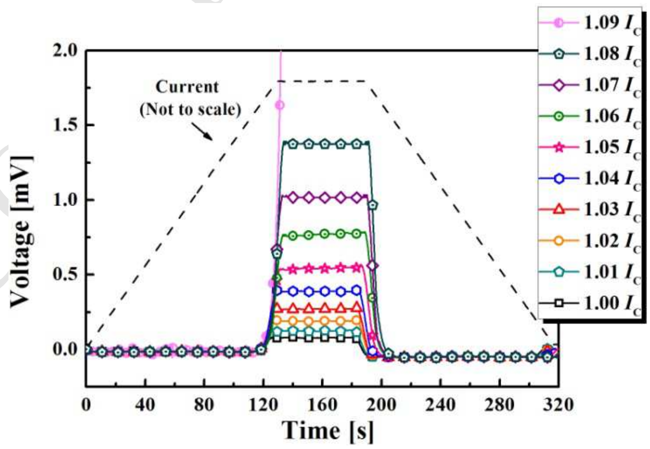

(c)

Fig. 7. The over-current test results for coil 1 (a), coil 2 (b), and coil 3 (c).

Fig. 8 presents the repetitive-cooling test results for coils 1, 2, and 3. The I_c values of coils 1, 2, and 3 after the 25th test decreased to 101, 107, and 120 A, respectively, because of the difference between the CTE of the GdBCO tape and epoxy composite. The test results indicate that coil 3 exhibited 1.72 % degradation in its I_c value, which was remarkably lower than those of coil 1 (16.3 %) and coil 2 (12.2 %). This finding was attributed to the smaller difference between the CTE of the GdBCO tape and epoxy composite for coil 3; the relatively high density of the epoxy/cBN–hBN composite without voids may have led to its low CTE. Overall, the addition of the cBN/hBN filler particles to the epoxy resin reduced the thermal contraction difference between the GdBCO CC tape and epoxy composite.

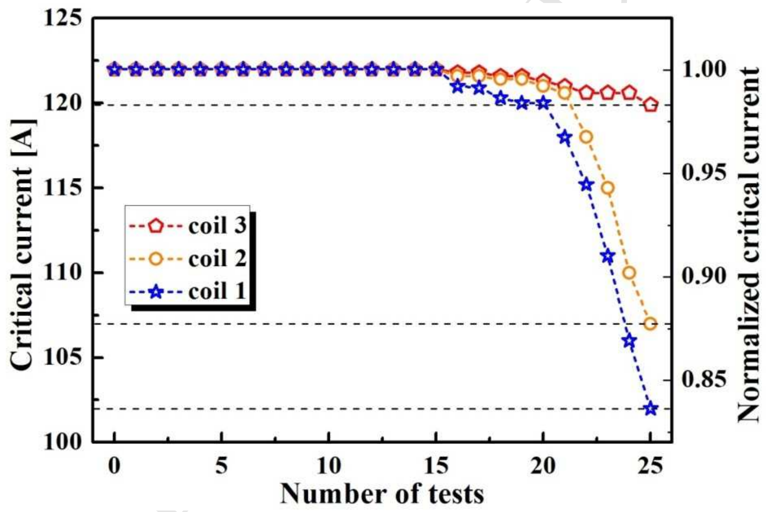

Fig. 8. Repetitive-cooling test results for coils 1, 2, and 3.

4. Conclusion

In this study, the cooling performance and thermal/electrical characteristics of GdBCO-coated superconducting coils impregnated with epoxy composites containing cBN particles (coil 1), hBN particles (coil 2), and a mixture of cBN/hBN particles (coil 3) were investigated using cool-down, over-current, and repetitive-cooling tests. The SEM images revealed that micro-voids were present only in the epoxy composites containing cBN or hBN particles alone because the epoxy resin with high viscosity could not perfectly permeate into the space between the particles. For the epoxy/cBN–hBN composite, the hBN particles effectively filled the voids produced between cBN particles, leading to the absence of micro-voids. The cool-down tests from room temperature to 77 K confirmed that coil 3 exhibited a faster cooling time than coils 1 and 2, indicating that the epoxy composite containing the mixture of cBN and hBN particles could effectively facilitate the heat transfer between LN2 and the coil. The over-current test results indicated that the current causing the thermal runaway phenomenon for coil 3 (133 A) was higher than those for coil 1 (128 A) and coil 2 (127 A), because of the enhanced thermal conductivity of the coil impregnated with the epoxy/cBN–hBN composite, which allowed hot spots to be dissipated more effectively in the event of a quench. In the repetitive-cooling test, coil 3 exhibited a 1.72 % degradation in its I_c value, which was remarkably lower than those of coil 1 (16.3 %) and coil 2 (12.2 %). The reduced degradation for coil 3 was attributed to the smaller difference between the CTE of GdBCO tape and the epoxy composite.

Overall, the addition of a mixture of cBN/hBN particles to the epoxy resin may be preferable to achieve highly stable and mechanically dense superconducting coils with high thermal conductivities and low CTEs. Further studies on the effect of the content, size, and degree of dispersion of filler particles in epoxy composites on the thermal/electrical characteristics of superconducting coils will be carried out in the future to further improve the thermal properties of the epoxy composites developed in this study.

Acknowledgment

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Figure Captions

Fig. 1. Photograph of GdBCO test coil before epoxy impregnation (a) and schematic drawing of the instrumentation consisting of the array of E-type TCs and VT (b).

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Table Captions

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