

Characterization of EPDM/micro-Silica/micro-Silicon Carbide (SiC)/nano-hBN Composites for HVDC Insulation

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Characterization of EPDM/micro-silica/micro-silicon carbide (SiC)/nano-hBN composites for HVDC insulation

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Abstract

Nano-sized hexagonal boron nitride (nano-hBN) was incorporated into an ethylene propylene diene monomer (EPDM)/micro-sized silicon carbide (micro-SiC) composite in order to prepare an electrical insulation material for high voltage direct current (HVDC). The surface of nano-hBN and micro-SiC was organized with 1,3-divinyl-1,1,3,3-tetramethyldisilazane, and the effects of nano-hBN content on HVDC and HVAC electrical breakdown strength, thermal conductivity and tensile characteristics were studied. EPDM/micro-SiC/nano-hBN composite was prepared by a two-roll mill in the weight ratio of EPDM : micro-SiC : nano-hBN = 100g : 25g : 0-30g. As nano-hBN content increased, thermal conductivity of EPDM/micro-SiC/nano-hBN composite increased, thereby effectively dissipating heat and preventing the thermal degradation of polymer chain. When 20g of nano-hBN particle was incorporated into EPDM(100g)/micro-SiC(25g) composite, HVDC breakdown strength and HVAC breakdown strength were maximum. (+)HVDC breakdown strength for the composite without nano-hBN was 87.3 kV/mm and that for the composite with nano-hBN(20 phr) was 95.4 kV/mm, which was 9.3% higher. (-)HVDC breakdown strength for the composite without nano-hBN was 89.3 kV/mm and that for the composite with nano-hBN(20 phr (parts per hundred epoxy resin)) was 98.2 kV/mm, which was 10.0% higher.

Keywords Electrical insulator · EPDM · Micro silicon carbide · Nano hexagonal boron nitride · HVDC insulation · Tensile strength

Statements and Declarations

Competing interests

The authors declare that they have no competing interests.

Introduction

The development of polymer composites for insulation materials in high voltage direct current (HVDC) equipment is essential to address the unique characteristics of the DC environment, which differs from conventional alternating current (AC), and to advance high-voltage and high-capacity technology [1-3]. Key developments can be summarized in three key areas: (1) improving electrical performance, (2) establishing thermal stability, and (3) ensuring environmental friendliness and cost-effectiveness.

In the first, the greatest technical challenge facing HVDC equipment is the accumulation of space charge at the interfaces between insulating components or within the insulation materials. This accumulation distorts the electric field within the insulator, leading to insulation breakdown. Therefore, many researchers are studying new methods to form composites by dispersing semiconducting powders, such as SiC, ZnO, graphene oxide (GO) and carbon nanotubes (CNT) within a polymer matrix in order to solve the problem of local accumulation by dispersing space charges evenly within the insulator [4-7].

In the second, HVDC equipment requires heat resistance to withstand high-temperature operating environments for higher power transmission, and to achieve this, research is being conducted on the selection of high-heat-resistant materials and the optimization of structural design. However, it is very difficult and costly to secure structural design flexibility by employing high-heat-resistant materials. To solve this problem, a technology is being adopted to composite materials by mixing high thermally conductive powders, such as hexagonal boron nitride (hBN), with polymers. Such composites improve the low thermal conductivity of polymer insulation materials and effectively dissipate heat generated in the conductors inside the HVDC equipment, thereby preventing the degradation of insulation performance (thermal degradation) caused by heat accumulation [8-10].

In the third, many researchers are conducting studies to develop polymer composites using biodegradable resins instead of conventional petroleum-based resins to solve global environmental problems, but the current research level is in its early stages [11,12].

EPDM is being widely studied as an insulating material for HVDC equipment due to its excellent electrical properties and physical durability [13-16]. Since this material exhibits excellent insulation performance and low dielectric loss even in high-voltage environments, it can minimize power loss when used in high-voltage DC transmission. In addition, because it is stable over a wide temperature range from -50°C to +150°C, it is suitable for high-voltage equipment that generates a lot of heat. Furthermore, due to its excellent ozone resistance, water resistance, and chemical corrosion resistance, it can be used in equipment that requires harsh external

environments for a long time, such as outdoor HVDC cable connectors or bushings for power facilities. Also, as explained above, space charge can be accumulated in HVDC systems and cause insulation degradation, and EPDM is a material that can effectively manage or suppress the accumulation of such space charge.

Many researchers are studying on introducing semiconducting SiC (electrical conductivity: ca. 10^1 - 10^3 S/cm) powder to EPDM (electrical conductivity: ca. 10^{-14} - 10^{-17} S/cm) matrix in order to evenly disperse space charge within the insulating materials. Our previous work investigated the space charge behaviors in EPDM/micro-SiC/nano-SiC composite and we found that an appropriate content of nano-SiC makes the space charge disperse throughout the EPDM matrix, greatly improving HVDC breakdown strength [15]. Li et al showed the space charge behaviors in the interface between LDPE and EPDM/SiC composite for HVDC cable accessory and they found that the approximate SiC incorporation can availably suppress the interface charge accumulation between the interface and offers a possible method for the improvement of cable accessory performance [17]. Zhang et al found that SiC doping to EPDM can effectively suppress the interface charge accumulation at the interface between low-density polyethylene (LDPE) and EPDM [18].

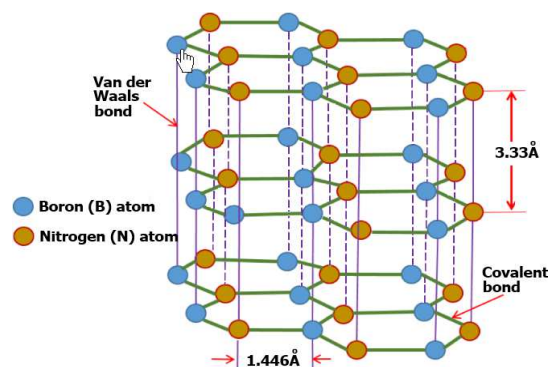


Fig. 1 Crystal structure of hexagonal boron nitride (hBN)

Hexagonal boron nitride (hBN) is structurally similar to graphite as shown in Fig. 1 and is therefore called ‘white graphite’ [19]. However, unlike graphite, which conducts electricity, it is a perfect insulator with a wide band gap of approximately 5.9 eV. Its electrical insulation properties include a high electrical resistivity of about 10^{14} - 10^{16} $\Omega\cdot\text{cm}$ at room temperature and a high dielectric breakdown strength of about 30-40 kV/mm, making it suitable for high-voltage insulation. Additionally, to improve the heat dissipation characteristics of EPDM (thermal conductivity: 0.1-0.2 W/m·K), hBN powder with excellent thermal conductivity (250-600 W/m·K) is being introduced to the EPDM matrix in order to rapidly dissipate heat generated within the components to prevent degradation of EPDM polymer chains. Since semiconducting SiC powder used for space charge dispersion also has very high thermal conductivity (120-490 W/m·K), mixing it with hBN powder can produce a

synergistic effect on heat dissipation characteristic [20-23].

In this study, HVDC insulating material was developed by introducing semiconductive micro-SiC powder and high thermal conductive nano-hBN into an EPDM matrix and its HVDC breakdown strength and tensile strength were studied.

Experimental part

Materials

An EPDM rubber was purchased from Dow Inc. whose product name was NORDEL™ IP 4640 Hydrocarbon Rubber. It was designed for molding and extruding applications. The weight ratio of ethylene : propylene : ethylidene norbornene (ENB) was 55 : 40 : 5 wt%. Vinyl group in the ENB crosslinked in the presence of dicumyl peroxide (DCP), sulfur and zinc oxide (ZnO) microparticle, and they were obtained from Merck. β -type micro-SiC powder was purchased from US Research Nanomaterials, Inc. (USA). Its electrical conductivity was 33.3-100 S/cm at 25°C, particle size was 2-100 μm and the purity was 99 wt%. Nano-hBN powder was also purchased from US Research Nanomaterials, Inc. (USA). Its thermal conductivity was 700-758 W/m·K at 25°C, particle size was 70-80 nm and purity was 99.8 wt%. To modify the surface of micro-SiC and nano-hBN particles, 1,3-divinyl-1,1,3,3-tetramethyldisilazane (DVTMDS, Gelest Inc. Japan) was used.

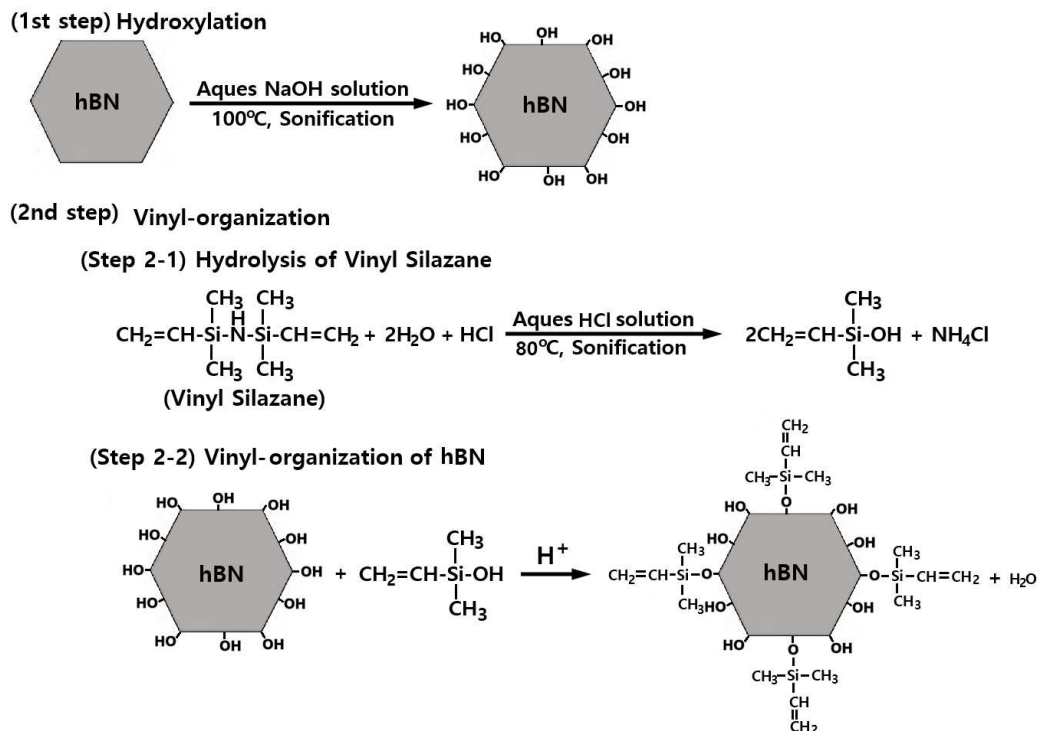


Fig. 2 Surface organization of nano-hBN with vinyl-silane coupling agent

Surface organization of micro-SiC and nano-hBN particles

As shown our previous work, Micro-SiC was modified with DVTMDS having vinyl group in order to improve the interfacial bonding between micro-SiC particles and EPDM via the hydrosilylation reaction [15, 24]. This process proceeded in two steps in succession: Step (1) generating hydroxyl groups on the surface of SiC powder and Step (2) organization of the SiC surface by the reaction of hydroxyl groups and silane coupling agents.

Similar to the surface organization process of micro-SiC particles [8, 15], the surface organization of nano-hBN was also carried out in two steps, as shown in Fig. 2. In the 1st step, hydroxyl groups were generated on surface of nano-hBN particles by treating nano-hBN(1g) in NaOH(5g)/distilled water(200g) solution at 100°C for 24 h under ultrasonic energy (26.4 kHz, 300W). Then, the nano-hBN particles were washed several times with deionized water and filtered under reduced pressure until the filtered water was neutral. It was dried at 60°C under vacuum overnight. In the 2nd step, the hydroxyl groups on the nano-hBN particles(1g) were reacted with DVTMDS(3g) in HCl(2.2g)/distilled water(150g) solution at 80°C for 5h under ultrasonic energy (26.4 kHz, 300W), resulting in vinyl-organized nano-hBN particles. It was washed several times with deionized water and filtered under reduced pressure until the filtered water was neutral. It was dried at 60°C under vacuum overnight and stored in a desiccator. The weight of vinyl silane group on the organized nano-hBN was estimated by thermogravimetric analysis (TGA) and Fourier-transform infrared spectroscopy (FT-IR).

Specimen preparation

EPDM(100g), vinyl-organized micro-SiC(25g), vinyl-organized nano-hBN(0-30g), ZnO(6g, crosslink activator for sulfur) and stearic acid(13g, crosslink activator for sulfur) were mixed well during the two-roll mill process. These materials were sequentially mixed little by little at 120°C for 1h, and DCP(1.7g, crosslinker) and sulfur(0.3g, co-crosslinker) were mixed well at 80°C for 30 min using a two-roll mill. The obtained master batch was then cured by a compressive method at 200°C and 4 MPa.

Electrical insulation breakdown tests under HVDC and HVAC conditions

Specimen dimension was 100 mm in diameter and 1 mm in thickness, and it was kept in 25°C insulating oil for 2 h before test. HVDC breakdown test was carried out using a DC Endurance Voltage Tester (Sungmin Electronics Co., Ltd., Korea), which could generate maximum voltage of 250 kV. A specimen was arranged between two spherical copper electrodes (diameter 7.40 mm), and then DC was applied at a rising speed of 2.5

kV/s until breakdown. All HVDC electrical breakdown data were statistically evaluated using a Weibull statistical analysis (Weibull++7.0 program of ReliaSoft (HBM Prentice Inc.) [25, 26].

HVAC breakdown test was conducted using a specimen of the same specifications as that used in the HVDC test. HVDC breakdown tester was an AC Endurance Voltage Tester (Haefely, Germany), which was controlled at the frequency of 60 Hz with the maximum voltage of 400 kV. A specimen was kept in 25°C insulating oil for 2 h before test and it was arranged between two spherical copper electrodes (diameter 7.40 mm). AC (60 Hz) was applied at a rising speed of 1.0 kV/s until the specimen breakdown. All HVAC electrical breakdown data were also statistically evaluated using a Weibull statistical analysis [25, 26].

Instrumental analyses

To estimate the amount of vinyl silane group on the organized nano-hBN, TGA (TG-121, Cahn) was carried out. About 10 mg of the organized nano-hBN was placed in a furnace and dynamic run was carried out from room temperature to 600°C at a heating rates 10 °C/min. N₂ purge gas flows at 50 ml/min to prevent the effect of sample oxidation. FT-IR (Nicolet iS 10, Thermo Fisher Scientific Inc.) analysis was performed for unorganized, OH-modified and vinyl-organized nano-hBN at a resolution of 2 cm⁻¹. Transmission electron microscopy (TEM, Hitachi S-4100) was used to observe the dispersion morphology of organized nano-hBN in EPDM matrix. Ultrathin section (60-90 nm in thickness) was prepared by cutting the EPDM/vinyl-organized nano-hBN (20 phr (parts per hundred epoxy resin)) nanocomposite at temperature below -60°C in liquid nitrogen using a cryo-ultramicrotomy (PT-X Power Tome Ultramicrotomes, Boeckeler Instruments). This ultrathin section was placed on a 200 mesh carbon-coated copper grid and observed. Thermal conductivity was measured according to the ASTM D5470 standard test method using a vertical type thermal conductivity meter (TCM-100V, SEO Com., Korea). Two specimens were prepared for each composition, each with a diameter of 3 cm and different thicknesses of 1 mm and 2 mm. After measuring the thermal resistance of two specimens under thermal equilibrium, the thermal conductivity was calculated by excluding the influence of interfacial resistance. Tensile strength and elongation were measured by universal testing machine (LR5K-plus, Lloyd Instruments Ltd., England). Tensile specimen was prepared under the recommendation of JIS B7502 and they were tested at a crosshead speed of 10 mm/min at 23°C and 50% relative humidity. All tensile and flexural data were statistically evaluated using a Weibull statistical analysis [25, 26].

Results and discussion

To measure the weight of vinyl silane group on the surface of organized nano-hBN particle, TGA was carried out and TGA curves for unorganized and vinyl-organized nano-hBN are compared as shown in Fig. 3. The thermogram of unorganized hBN shows a typical thermal decomposition behavior and weight-loss of 0.23 wt% at 600°C, attributable to the loss of chemisorbed humidity [27]. Vinyl-organized nano-hBN starts to decompose at about 310°C and its weight-loss is 2.99 wt% at 600°C. The difference is 2.76 wt%, which corresponds to the chemical bonding weight of DVTMDS on the nano-hBN.

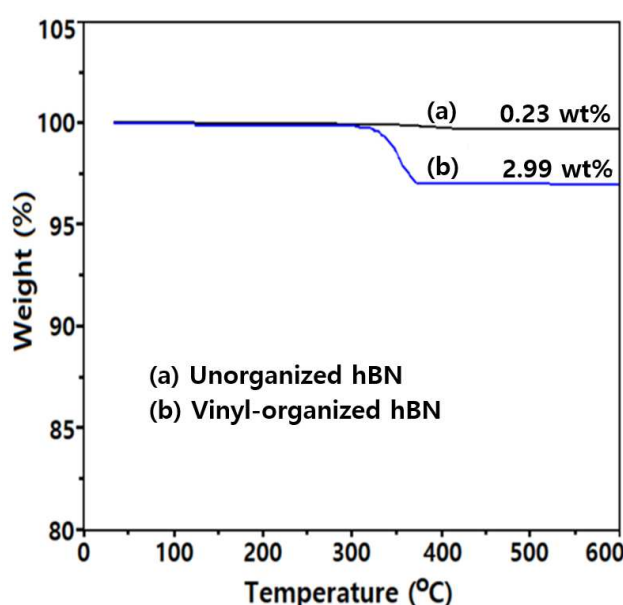


Fig. 3 TGA thermograms for unorganized and vinyl-organized nano-hBN

In order to characterize the chemical difference among unorganized hBN, OH-modified hBN and vinyl-organized nano-hBN, the three dried samples were analyzed by a FT-IR spectrometer. As shown in Fig. 4(a), unorganized hBN shows two characteristic peaks around 785 cm^{-1} and 1375 cm^{-1} , corresponding to the B–N–B out-of-plane bending vibration and B–N in-plane stretching vibration, respectively [27,28]. As the surface of hBN is modified with -OH group, a broad peak around 3500 cm^{-1} and a weak peak at 932 cm^{-1} newly appear, as shown in Fig. 4(b). The former is attributed to the stretching vibration of hydroxyl groups (–OH) and the latter is attributed to the B–O stretching vibration, indicating that the surface of nano-hBN particle is successfully functionalized with hydroxyl groups. For vinyl-organized nano-hBN, FT-IR spectrum (Fig. 4(c)) shows new peaks at 918, 1085, 2991 and 3015 cm^{-1} . The first two peaks are corresponded to the stretching vibration of

silanol group (Si-OH) and the stretching vibration of Si-O-Si, and the last two peaks are corresponded to -CH=CH₂ and -CH₃ groups originating from DVTMDS on the nano-hBN surface. These results indicated that the surface of nano-hBN particle is successfully organized with vinyl groups.

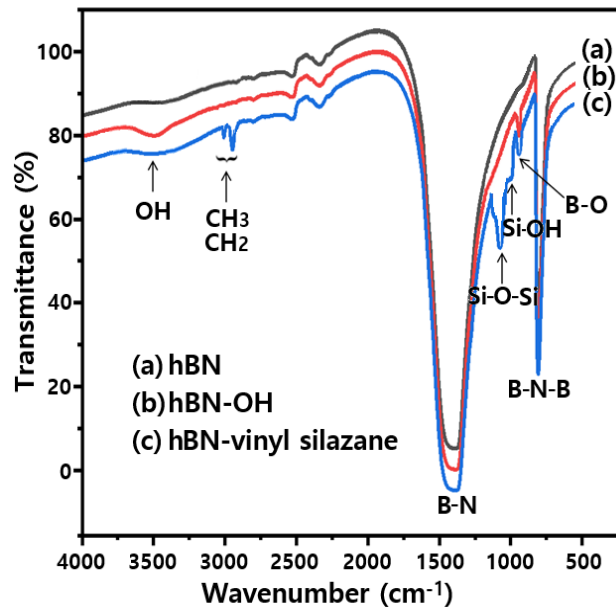


Fig. 4 FT-IR spectra of (a) unorganized, (b) OH-modified and (c) vinyl-organized nano-hBN

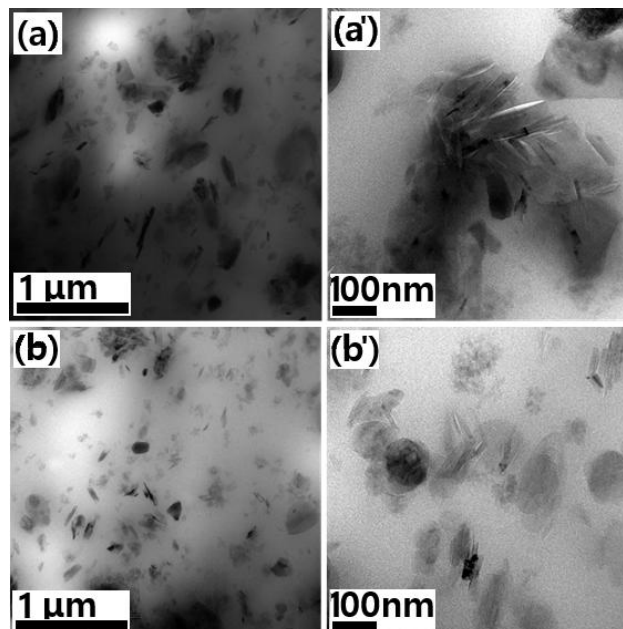


Fig. 5 TEM images of vinyl-organized nano-hBN dispersed in EPDM. The amounts of nano-hBN particles were 20 phr. (a) unorganized (b) vinyl-organized nano-hBN. Prime (') is for high magnification image

To compare the dispersion morphologies of nano-hBN (20 phr) in EPDM, TEM observations were carried out. As shown in Fig. 5(a), unorganized nano-hBN particles are not evenly dispersed in EPDM matrix because they are not compatible with EPDM, resulting in some large aggregates. The aggregate is observed under high magnification in Fig. 5(a'), and it can be seen that the layered hexagonal hBN particle is not exfoliated. In contrast, as shown in Fig. 5(b) and 5(b'), vinyl-organized nano-hBN is evenly dispersed EPDM matrix in exfoliated small state. These results indicate that DVTMDS achieve a good effect on the dispersion of nano-hBN particles in the EPDM matrix, because the surface of the nano-hBN is changed to EPDM-friendly surface and vinyl group on the surface of nano-hBN is reacted with vinyl group of EPDM matrix through addition polymerization, improving the interfacial characteristics.

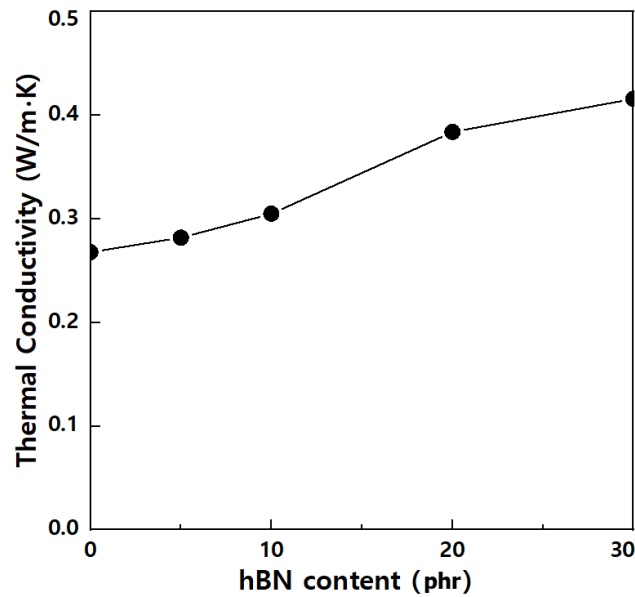


Fig. 6 Thermal conductivity of EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Fig. 6 shows the effect of nano-hBN content on the thermal conductivity of EPDM/micro-SiC(25 phr)/nano-hBN composites. As expected, thermal conductivity increases with increasing nano-hBN content. Thermal conductivity for EPDM/micro-SiC(25 phr) system without nano-hBN is 2.68×10^{-1} W/m·K and that for EPDM/micro-SiC(25 phr) system with nano-hBN(30 phr) is 4.16×10^{-1} W/m·K, which was 1.55 times higher. Therefore nano-hBN powder can increase the thermal conductivity of EPDM and effectively dissipate heat generated inside the HVDC equipment, thereby preventing the thermal degradation of polymer chains.

Table 1 (+)HVDC insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Specimen Number											
	1	2	3	4	5	6	7	8	9	10	11	12
0	97.8	95.0	91.4	90.1	87.2	86.4	81.2	79.9	79.3	73.0	72.2	69.6
5	99.5	95.0	91.7	91.3	91.1	90.1	87.9	85.5	83.8	80.4	79.9	75.9
10	96.8	96.6	93.2	92.2	90.6	90.1	87.0	86.7	85.8	85.7	84.8	84.7
20	101.8	96.7	95.8	95.4	94.8	94.0	92.5	92.3	91.9	90.7	89.3	87.5
30	98.9	97.2	96.3	95.8	91.8	91.7	90.3	89.6	84.5	84.3	72.2	-

Table 1 lists all data of (+)HVDC insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content, and all the experimental data, t are statistically analyzed by the following two-parameter Weibull probability distribution.

$$f(t) = \frac{\beta}{\eta} \left(\frac{t}{\eta}\right)^{\beta-1} e^{-\left(\frac{t}{\eta}\right)^{\beta}} \quad (1)$$

where β is the shape parameter and η is the scale parameter. By taking the natural logarithm of both sides twice and rearranging, Eq. (1) is transformed into a straight-line equation, $y = a + bx$, as follows:

$$\ln\left(\ln\left(\frac{1}{1-f(t)}\right)\right) = \beta \ln(t) - \beta \ln(\eta) \quad (2)$$

Here, $y = \ln\left(\ln\left(\frac{1}{1-f(t)}\right)\right)$, which is cumulative probability and $x = \ln(t)$ are set, Eq. (2) can be rewritten as follows:

$$y = \beta x - \beta \ln(\eta) \quad (3)$$

This is a straight-line equation with a slope of β and an intercept of $\beta \ln(\eta)$.

Fig. 7 shows Weibull plots of (+)HVDC insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content ((●) 0 phr, (■) 5 phr, (▲) 10 phr, (▼) 20 phr and (◆) 30 phr, respectively) at 25°C insulating oil. Three Weibull parameters (scale, B10, and shape parameters) can be obtained from Weibull plot for each EPDM composite. The shape parameter, β is calculated from the slope, which means the breakdown data distribution. The scale parameter, η is obtained from (+)HVDC insulation breakdown strength at 63.2% cumulative probability, and the B10 is a value at 10% cumulative probability of breakdown (90% would survive) under the applied (+)HVDC. The three statistical parameters for

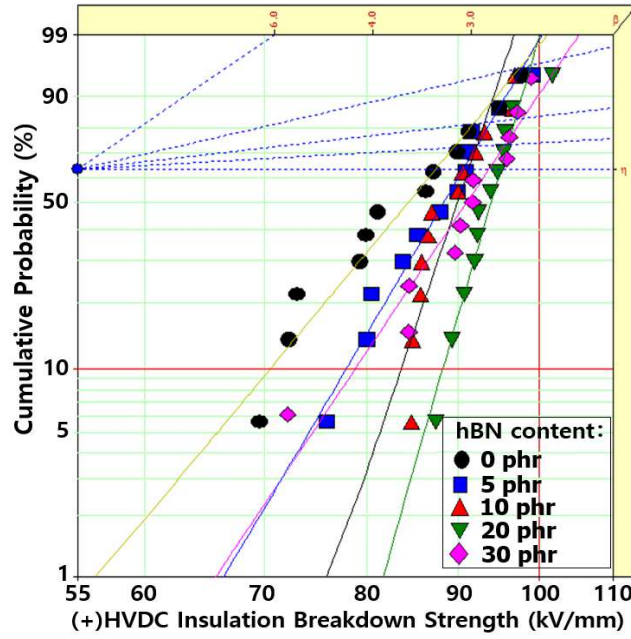


Fig. 7 Weibull statistical analyses of (+)HVDC insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN contents ((●) 0 phr, (■) 5 phr, (▲) 10 phr, (▼) 20 phr and (◆) 30 phr)

Table 2 Weibull parameters of (+)HVDC insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Shape Parameter	Scale Parameter (kV/mm)	B10 Value (kV/mm)
0	10.4	87.3	70.6
5	14.7	90.5	77.9
10	25.7	91.3	83.6
20	29.7	95.4	88.2
30	13.0	93.6	78.8

each EPDM composite are listed on Table 2. (+)HVDC electrical breakdown voltage for EPDM/micro-SiC(25 phr) composite without nano-hBN is 87.3 kV/mm and the maximum value, 95.4 kV/mm is obtained from EPDM/micro-SiC(25 phr) composite with nano-hBN (20 phr), which is 9.3% higher, and the value decreases thereafter. The shape parameter represents the shape of the data distribution indicating how the product failure rate changes over time.²⁵ When $\beta < 1$, the material fails frequently during the initial stages of use or immediately after manufacturing, with the failure rate decreasing over time, and when $\beta = 1$, the material's failure rate

remains constant regardless of time, primarily caused by random external shocks or unpredictable factors. Furthermore, when $\beta > 1$, the failure rate increases rapidly over time due to component aging, fatigue accumulation, and wear, and in this study, the EPDM/micro-SiC(25 phr)/nano-hBN (0-30 phr) composites exhibit β values approximately 10-30 times higher than 1, and it can be seen that the effect is greatest when nano-hBN is used at 20 phr. This implies a need to improve the long-term durability of the EPDM system for HVDC insulator, and incorporation of nano-hBN with high heat dissipation characteristics could be a solution.

Table 3 lists all data of (-)HVDC insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content, and all the experimental data are statistically analyzed by Weibull probability distribution, as shown in Fig. 8. Nano-hBN contents are (●) 0 phr, (■) 5 phr, (▲) 10 phr, (▼) 20 phr and (◆) 30 phr, respectively at 25°C insulating oil. Three Weibull parameters are listed on Table 4. (-)HVDC electrical breakdown voltage for EPDM/micro-SiC(25 phr) composite without nano-hBN is 89.3 kV/mm and the maximum value, 98.2 kV/mm is obtained from EPDM/micro-SiC(25 phr) composite with nano-hBN (20 phr), which is 10.0% higher, and the value decreases thereafter. As is well known, (-)HVDC breakdown strength is slightly higher than the (+)HVDC breakdown strength. This is because the injection rate of charge carriers at the cathode is higher than that at the anode [29]. β values are approximately 14-23 times higher than 1, and it can be seen that the effect is greatest when nano-hBN is used at 20 phr.

Table 3 (-)HVDC insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Specimen Number											
	1	2	3	4	5	6	7	8	9	10	11	12
0	95.0	93.9	92.8	90.5	88.7	86.7	84.6	83.9	83.1	81.8	79.4	71.4
5	98.1	96.8	95.2	94.4	92.2	91.5	89.0	87.9	85.4	83.6	82.4	78.9
10	100.1	98.3	97.4	95.2	94.3	93.7	93.1	92.6	91.9	90.6	88.1	82.3
20	103.6	102.2	100.5	98.8	98.3	97.6	95.7	94.8	93.8	92.7	90.6	88.3
30	102.3	100.6	99.6	97.7	96.6	96.1	94.4	93.9	92.7	90.9	89.6	84.5

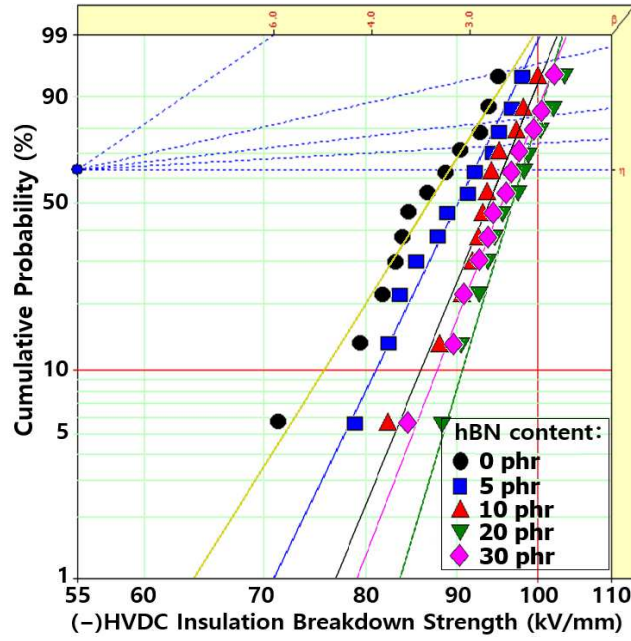


Fig. 8 Weibull statistical analyses of (-)HVDC insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN contents ((●) 0 phr, (■) 5 phr, (▲) 10 phr, (▼) 20 phr and (◆) 30 phr)

Table 4 Weibull parameters of (-)HVDC insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Shape Parameter	Scale Parameter (kV/mm)	B10 Value (kV/mm)
0	14.2	89.3	75.7
5	15.6	92.1	81.1
10	22.0	95.7	85.8
20	23.7	98.2	90.8
30	21.1	96.9	87.9

Table 5 lists all data of HVAC (60 Hz) insulation breakdown strength for EPDM/micro-SiC(25 phr) composites with various nano-hBN contents in 25°C insulating oil, and all data is also estimated by Weibull analysis, as shown in Fig. 9. Nano-hBN contents are (●) 0 phr, (■) 5 phr, (▲) 10 phr, (▼) 20 phr and (◆) 30 phr, respectively, and three statistical parameters for each system are listed on Table 6. The HVAC electrical breakdown voltage for EPDM/micro-SiC(25 phr) composite without nano-hBN is 33.2 kV/mm and the maximum value, 37.9 kV/mm is obtained from EPDM/micro-SiC(25 phr) composite with nano-hBN (20 phr), which is 14.2% higher, and the value decreases thereafter. β values are approximately 13-18 times higher than 1, and it can be seen that the effect is greatest when nano-hBN is used at 20 phr.

Table 5 HVAC (60 Hz) insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Specimen Number											
	1	2	3	4	5	6	7	8	9	10	11	12
0	37.0	35.1	34.3	34.1	32.6	31.6	31.0	30.8	30.0	29.1	28.6	27.4
5	37.7	35.8	35.4	35.1	33.8	33.2	33.0	32.7	30.7	29.8	29.3	28.2
10	38.1	27.8	36.0	35.5	35.0	33.6	32.5	31.6	31.3	30.7	29.8	29.5
20	39.6	39.0	38.5	38.2	37.5	37.2	36.4	35.7	35.2	34.0	33.5	32.6
30	38.8	38.2	37.8	37.1	36.8	35.5	35.2	34.5	34.3	33.4	30.6	29.8

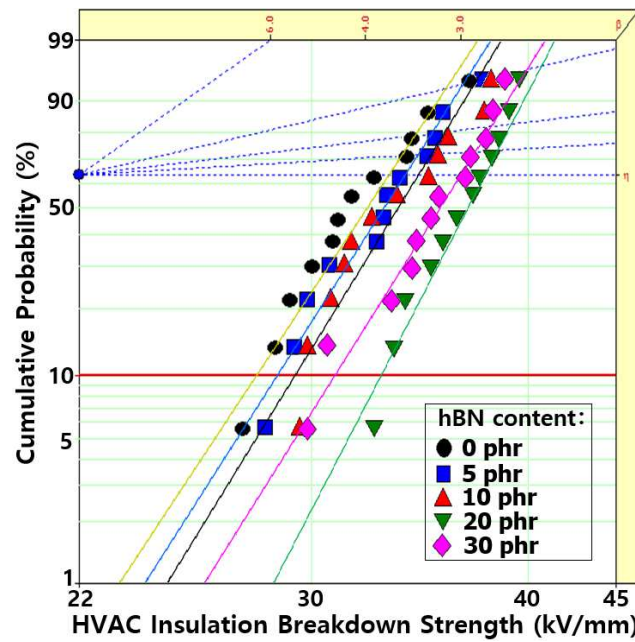


Fig. 9 Weibull statistical analyses of HVAC (60 Hz) insulation breakdown strength for EPDM/micro-SiC (25 phr)/nano-hBN composites according to nano-hBN contents ((●) 0 phr, (■) 5 phr, (▲) 10 phr, (▼) 20 phr and (◆) 30 phr)

Table 6 Weibull parameters of HVAC (60 Hz) insulation breakdown strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Shape Parameter	Scale Parameter (kV/mm)	B10 Value (kV/mm)
0	12.9	33.2	27.9
5	13.0	34.0	28.6
10	14.2	34.6	29.3
20	18.2	37.9	33.0
30	13.8	36.5	30.9

The (-)HVDC breakdown strength is 2.6-2.8 times higher than that of the HVAC breakdown strength. This is because under DC field, the maximum DC voltage equals to the applied DC voltage, whereas under AC condition, the maximum AC voltage (ACV_{peak}) equals to $\sqrt{2} \cdot V_{RMS}$, in which RMS refers to the root mean square [30]. Furthermore, under DC condition, the voltage direction remains constant, so fatigue or additional dielectric heat generation due to polarity reversal does not occur. On the other hand, under AC condition, AC voltage reaches to $+V_{peak}$ and again $-V_{peak}$ in 60 cycles per second. This causes molecules inside the insulator to rapidly change direction, generating frictional heat (dielectric loss) and subjecting the insulating material to greater physical and chemical fatigue (stress) [31]. Therefore, HVDC breakdown strength is far higher than that of HVAC breakdown strength.

Table 7 Tensile strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Specimen Number								
	1	2	3	4	5	6	7	8	9
0	4.41	4.33	4.28	4.26	4.24	4.23	4.20	4.13	3.98
5	4.46	4.35	4.33	4.30	4.28	4.26	4.25	4.18	4.17
10	4.67	4.58	4.55	4.52	4.47	4.46	4.39	4.37	4.24
20	4.75	4.63	4.59	4.54	4.51	4.48	4.39	4.25	-
30	4.81	4.67	4.64	4.59	4.58	4.55	4.52	4.44	4.41

Table 8 Elongation for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Specimen Number								
	1	2	3	4	5	6	7	8	9
0	550	546	540	533	531	527	523	514	496
5	556	536	535	533	530	524	516	508	491
10	534	529	526	520	516	511	509	500	481
20	523	509	506	505	498	487	479	470	-
30	505	500	489	484	480	478	472	468	455

Tables 7 and 8 list all data of tensile strength and elongation for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content, respectively, and all the experimental data are statistically analyzed by Weibull probability distribution, as shown in Fig. 10(a) and 10(b). Nano-hBN contents are (●) 0 phr, (■) 5 phr, (▲) 10 phr, (▼) 20 phr and (◆) 30 phr, respectively. Three Weibull parameters for tensile strength and elongation are obtained from each plot and listed on Table 9 and 10. As nano-hBN content increases in the EPDM/micro-SiC(25 phr)/nano-hBN composites, scale parameter and B10 value for tensile strength increase, while elongation decrease. Tensile strength for EPDM/micro-SiC(25 phr) composite without nano-hBN is 4.30

MPa, and that for EPDM/micro-SiC(25 phr) composite with nano-hBN(20 phr) is 4.58 MPa, which was 6.5% higher.

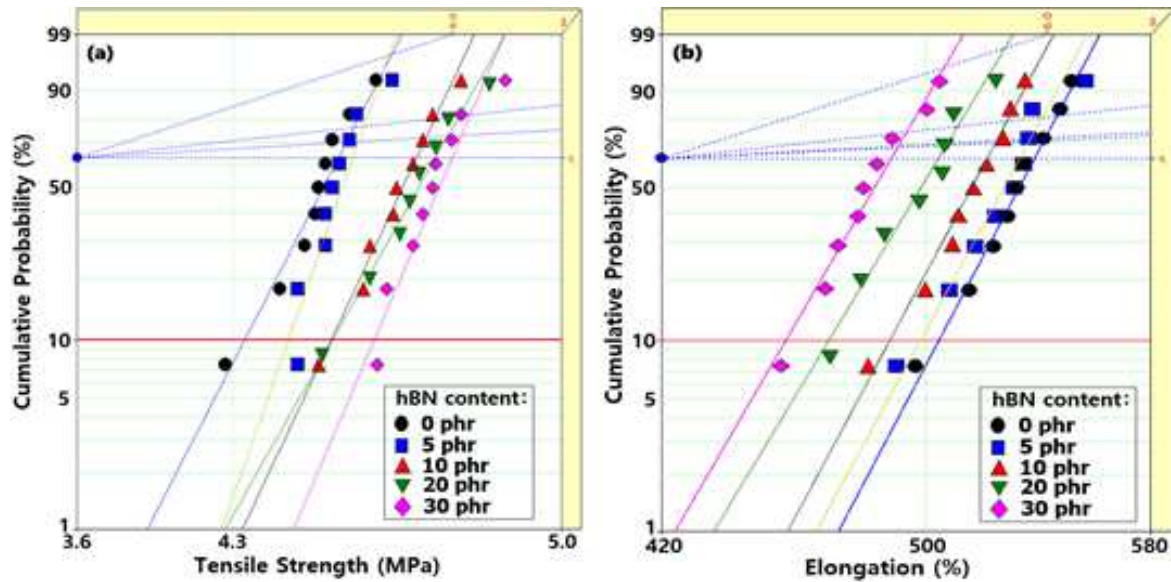


Fig. 10 Weibull statistical analyses of (a) tensile strength and (b) elongation for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN contents (●) 0 phr, (■) 5 phr, (▲) 10 phr, (▼) 20 phr and (◆) 30 phr

Table 9 Weibull parameters of tensile strength for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Shape Parameter	Scale Parameter (MPa)	B10 Value (MPa)
0	38.8	4.30	4.03
5	56.8	4.34	4.15
10	39.0	4.53	4.28
20	32.7	4.58	4.28
30	43.9	4.64	4.40

Table 10 Weibull parameters of elongation for EPDM/micro-SiC(25 phr)/nano-hBN composites according to nano-hBN content

Nano-hBN Content (phr)	Shape Parameter	Scale Parameter (%)	B10 Value (%)
0	35.0	538	504
5	31.5	533	499
10	34.8	521	488
20	31.6	505	469
30	34.7	489	456

Conclusion

Nano-sized hexagonal boron nitride was incorporated into EPDM/micro-SiC(25 phr) composite in order to prepare an HVDC insulation material, and HVDC breakdown strength, HVAC breakdown strength, thermal conductivity and tensile strength were studied. The surface of nano-hBN particle was organized by 2.76 wt% DVTMDS and FT-IR spectrum showed -CH=CH_2 and -CH_3 groups originating from DVTMDS, indicating that the surface of nano-hBN particle was successfully organized with vinyl groups. TEM observation showed that vinyl-organized nano-hBN was evenly dispersed EPDM matrix in exfoliated small state. (+)HVDC breakdown strength for EPDM(100g)/micro-SiC(25g) composite without nano-hBN was 87.3 kV/mm and that for the composite with nano-hBN(20 phr) was 95.4 kV/mm, which was 9.3% higher. (-)HVDC breakdown strength for the composite without nano-hBN was 89.3 kV/mm and that for the composite with nano-hBN (20 phr) was 98.2 kV/mm, which was 10.0% higher. Thermal conductivity for EPDM/micro-SiC(25 phr) system without nano-hBN is 2.68×10^{-1} W/m·K and that for EPDM/micro-SiC(25 phr) system with nano-hBN(30 phr) is 4.16×10^{-1} W/m·K, which was 1.55 times higher. As nano-hBN content increased, tensile strength increased, From these results, it is evaluated that EPDM/micro-SiC(25 phr)/nano-hBN(20 phr) can be used as an HVDC insulating material.

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Ethics declarations

Ethics approval

Not applicable

Consent to participate

Not applicable

Consent for publication

Not applicable

Clinical trial number

Not applicable.

Competing interests

The authors declare that they have no competing interests.

Data availability

The data related to the current study is presented in the paper.