Electrical Properties of the Epoxy Nano-composites according to Additive

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The use of a filler material in epoxy composite materials is an essential condition for reducing the unit cost of production and reinforcing mechanical strength. However, the dielectric strength of insulators decreases rapidly due to interactions between the epoxy resin and filler particles. In contrast to existing composite materials, nano-composite materials have superior dielectric strength, mechanical strength, and enduring chemical properties due to an increase in the bond strength of the polymer and nano material. It is reported that nano-fillers provide new characteristics different from the properties of the polymer material. This study is to improve the insulation capability of epoxy resins used in the insulation of a power transformer apparatus and many electronic devices mold. To accomplish this, the additional amount of nano-SiO_2 to epoxy resin was changed and the epoxy/SiO_2 nano composite materials were made, and the fundamental electrical properties were investigated using a physical properties and an analysis breakdown test. Using allowable breakdown probability, the optimum breakdown strength for designing an electrical apparatus was determined. The results found that the electrical characteristics of the nano-SiO_2 content specimens were superior to the virgin specimens. The 0.4 wt% specimens showed the highest electrical properties among the specimens examined with an allowable breakdown probability of 20 %, which indicates stable breakdown strength in insulating machinery design.

Keywords: Allowable breakdown probability, Coulomb barrier effect, Dielectric strength, Epoxy resin, Nano-composite, Weibull statistics

1. INTRODUCTION

The increase of the requirement for electrical power with extra high voltage and massive capacity has led to an increased demand for the development of new electric transfer apparatus and electric power cables. Meeting this demand has also resulted many electrical accidents and insulator breakdowns caused by electrical stress[1-3]. Epoxy resin is an insulating material used to insulate power transformer apparatus, as well as in the impregnation of several devices and casting mold treatments. As it is a hardening thermosetting resin at room or elevated temperatures, it has superior mechanical characteristics (such as the ability to withstand high voltage and resist abrasion) as well as excellent chemical characteristics, such as water resistance and chemical resistance[4,5]. Epoxy resin is used widely because it is easy to produce variable characteristics due to the fillers used. Adding filler is indispensable for economical advantages as well as the reinforcement of mechanical strength. However, it is well known that the electrical characteristics of a composite material are reduced because of the interface between the epoxy resin and filler. Currently, micro-sized fillers are used. Recently with the development of nano technology, the addition of various nano composite materials as an insulating material for electric power apparatus has been examined[6,7]. Nano composite materials have superior dielectric strength, mechanical strength, and enduring chemical properties. It has been reported that nano composite materials have superior dielectric strength, mechanical strength, and enduring chemical properties, and they have shown better physical properties than existing insulating materials with the merits as an inorganic matter and polymer[8-10]. In this paper, the allowable breakdown strength was investigated using Weibull statistics, and the electrical properties and insulation breakdown data of the composite material with a nano filler was compared with that of a composite material with micro-sized SiO_2.

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2. EXPERIMEMTS

2.1 Specimens

Figure 1 shows the process for fabricating the specimens. The dimension of the specimens was 25×25×2.5 mm^3. The composite was made from epoxy resin and a hardener with added nano SiO_2 in the following amounts: 0.2, 0.4, 0.6, 0.8, 1.2, and 1.4 wt%. The mixing temperature was maintained at 70 °C for 10 min to mix the epoxy resin, hardener and nano filler. The specimens were extracted in air at 80 °C for 5 min. The specimens were then obtained by first curing at 120 °C for 2 hours followed by a second curing step at 130 °C for 8 hours.

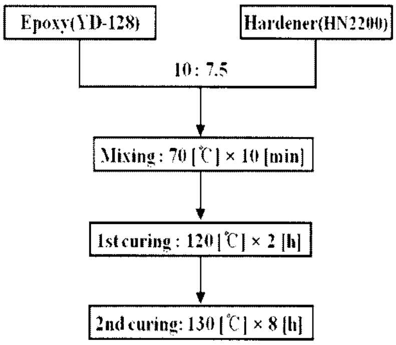


Fig. 1. Fabrication process.

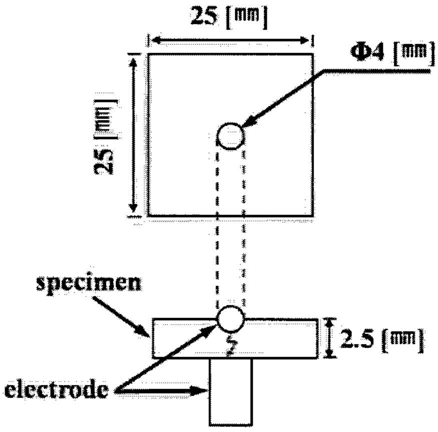


Fig. 2. Shape of the specimen and electrodes.

Figure 2 shows the manufactured shape of the specimen and electrodes. Spherical and flat electrodes were used. Electrical breakdown occurred between the electrodes. The applied voltage was increased using the ramp voltage method until insulation breakdown.

2.2 Physical properties

Scanning electron microscopy was used to investigate the internal structure of the specimens. The specimens were made with dimensions of 20×10×2.5 mm^3. The resulting samples were coated with gold and then imaged.

2.3 Dielectric strength properties

The electrical properties were examined as a function of the amount of nano-filler using a high voltage generating device. By applying a ramp method, an AC voltage with a frequency 60 Hz was increased in 1 kV/s steps until the insulating breakdown voltage was reached. Silicone oil (1,000 cSt) that was vacuum dried for 30 min was used as the surrounding medium in order to suppress the effects of water. The temperature of the silicone oil with the specimens was 25, 50, 80, and 100 °C. The dielectric strength of the specimens was measured 5 min after changing the temperature. The number of measurement specimens was 15 turns under the same conditions, and the average value was used as the breakdown voltage.

2.4 Weibull statistics

The breakdown of the data was examined using Weibull statistics. If the distribution function of the insulating breakdown voltage (accumulative probability) is F(x), the 2-parameter Weibull probability distribution function has the following formula (1) [11,12]:

$$F(x) = 1 - \exp\left[\left(\frac{x}{x_s}\right)^m\right] \quad (1)$$

Where, F(x): accumulative probability
x: measured value of the specimens
x_s: measured parameter(63.2 % value of accumulative probability)
m: shape parameter

3. RESULTS AND DISCUSSION

The physical properties and dielectric strength were examined to investigate the electrical characteristics of the insulating materials according to the nano particle additive; Weibull Statistics were used to investigate the allowable breakdown strength.

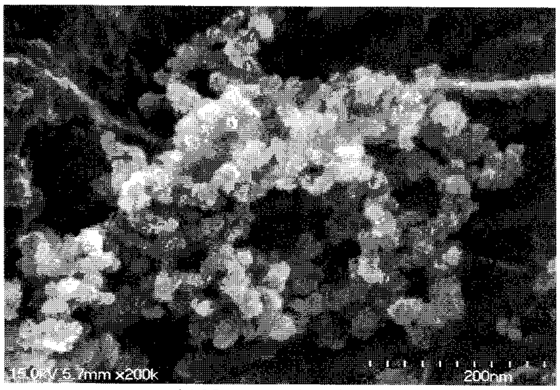
3.1 Physical properties

Photograph 1 shows an SEM image of a nano SiO_2 particle and a cross-section depending on the nano SiO_2 content. Photo. 1(a) shows that the size of nano particle is approximately 12-15 nm. As the amount of additive was increased, the distribution of nano particles was constant until 0.4 wt%. However, as shown in Photo. 1(c), a particle lump was observed when the amount SiO_2 additive was further increased. This deteriorated the dielectric strength due to the increased number of interfaces increase[13]. It is believed that the dielectric strength deteriorated due to the influence of the increasing number of interfaces.

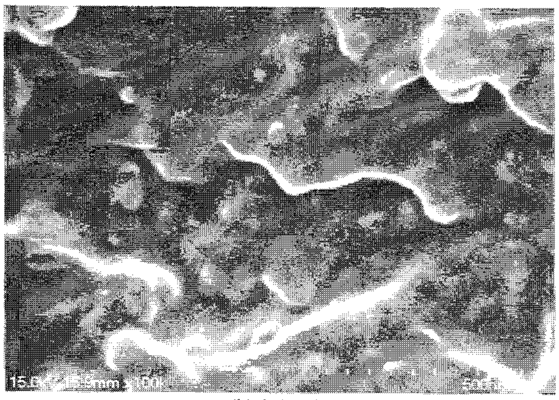
3.2 Dielectric strength

Figure 3 shows the dielectric strength as a function of the amount of additive at 25 °C. It was confirmed that the dielectric strength was increased by 20 % from 2.99 MV/cm to 3.6 MV/cm with increasing additive from 0 to 0.4 wt%. With further increases to 1.2 wt%, the dielectric strength was almost constant. At more than 1.2 wt%, the dielectric strength decreased 12.75 % from 3.45 MV/cm to 3.01 MV/cm. However, it is believed that the dielectric strength of the specimen with 0.4 wt% additives was superior to that of a virgin specimen because the additive interrupted the movement of the carrier.

Figure 4 shows the dielectric strength as a function of the amount of additive at 50 °C. The dielectric strength increased 14 % from 2.24 MV/cm to 2.55 MV/cm as the amount added was increased from 0 to 0.4 wt%. With further increases to 1.2 wt%, the dielectric strength was almost constant. At more than 1.2 wt%, the dielectric strength decreased 5.1 % from 2.35 MV/cm to 2.23 MV/cm. In particular, it was confirmed that the dielectric strength at 50 °C was lower than at 25 °C due to the easier carrier movement by the contribution of thermal energy.



(a) Nano powder of SiO_2



(b) 0.4 wt%

(c) 1.4 wt%

Photo. 1. SEM of specimens.

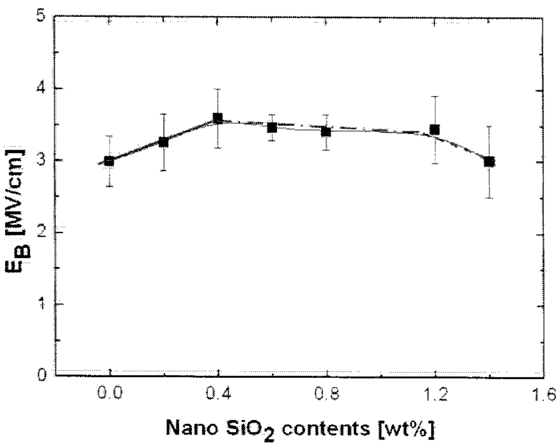


Fig. 3. Dielectric strength as a function of the amount of additive(25 °C).

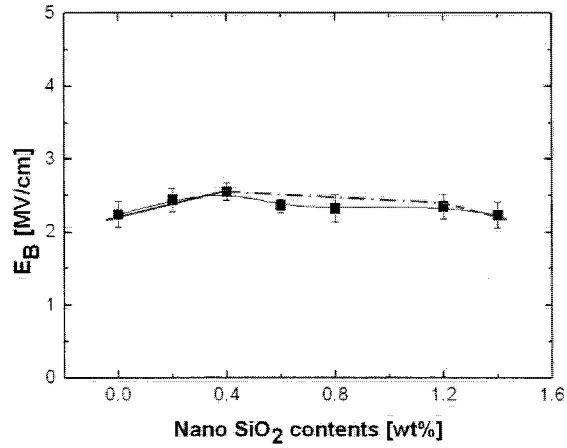


Fig. 4. Dielectric strength as a function of the amount of additive(50 °C).

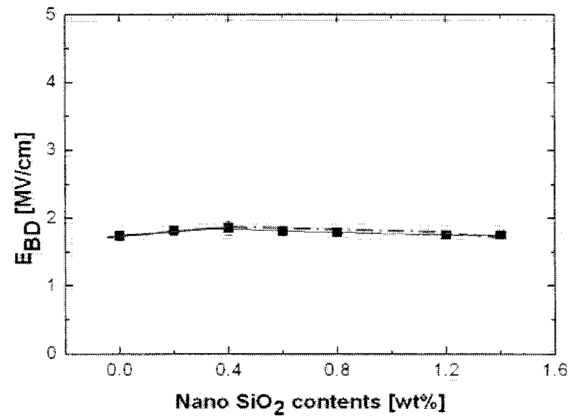


Fig. 5. Dielectric strength as a function of the amount of additive(80 °C).

Figure 5 shows the dielectric strength as a function of the amount of additive at 80 °C. The dielectric strength increased 6.3 % from 1.74 MV/cm to 1.85 MV/cm as the amount added was increased from 0 to 0.4 wt%.

The dielectric strength was almost constant with further increases in nano particle concentration. Generally, the glass transition temperature of an epoxy resin is approximately 80 °C. In this case, the measurement temperature, 80 °C, and was higher than the glass transition temperature and contributed to the phase change, so that the amount of additive had less influence on the dielectric strength.

Figure 6 shows the dielectric strength as a function of the amount added at 100 °C. The dielectric strength increased 5.5 % from 1.62 MV/cm to 1.71 MV/cm as the amount of additive was increased from 0 to 0.4 wt%. The dielectric strength is almost constant with further increases in nanofiller concentration. With an increasing amount of additive to 0.4 wt% the distance between the nano SiO_2 particles decreased. Therefore, the injection of an electron is difficult due to Coulomb's barrier effect, which the decreased dielectric strength for the carrier movement is restricted[14-16].

However as shown in' Photo. 1(c), the further increases in the amount added resulted in a further decrease in dielectric strength due to the increased number of interfaces through the mixing of a nano particle. The decrease in dielectric strength when the temperature was increased to more than 80 °C was attributed to the inner network coherence of the specimens.

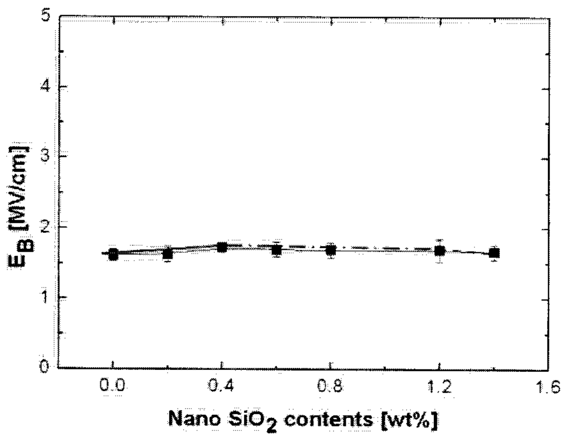


Fig. 6. Dielectric strength as a function of the amount of additive(100 °C).

3.3 Allowable breakdown strength

When designing an electrical apparatus, the allowable breakdown strength is important for improving the insulating ability. The allowable breakdown strength was confirmed using the form of the allowable breakdown probability. Formula (2) shows the allowable breakdown strength using the shape parameter, measure parameter, and allowable breakdown probability[17].

$$E_B = E_{Scale} \cdot \exp\left[\frac{\ln\left(\ln\frac{1}{1-F}\right)}{E_{Shape}}\right] \quad (2)$$

If the breakdown probability is 0.1 %, the allowable breakdown strength is 1.29 MV/cm in the virgin specimen at 25 °C (where, E Scale: 3.17, E Shape: 7.73).

$$E_B = 3.17 \cdot \exp\left[\frac{\ln\left(\ln\frac{1}{1-0.001}\right)}{7.73}\right] = 1.29\,[MV/cm] \quad (3)$$

A breakdown probability of 0.1 % means that breakdown strength is a maximum of 1.29 MV/cm. The allowable breakdown strength is predicted using the Weibull distribution. Tables 1 and 2 show the allowable breakdown strength due to temperature and the breakdown probability was 0.01 %, 0.1 %, 1.0 %, and 20 %. When the breakdown probability was 20 % in all specimens, the allowable breakdown strength was within the error range of the average breakdown strength, which means that the breakdown probability is the stable breakdown strength at 20 %. Problems with stability can appear out of the error range with further increases in the breakdown probability.

Table 1. The allowable breakdown probability depending on the amount of additive(25 °C).

Unit: [MV/cm]

Classification	E_{ave}	σ	Breakdown probability [%]			
			0.01	0.1	1.0	20
Virgin	2.99	0.37	0.96	1.29	1.74	2.61
0.2 [wt%]	3.26	0.40	1.02	1.39	1.88	2.83
0.4 [wt%]	3.60	0.43	1.17	1.58	2.12	3.14
0.6 [wt%]	3.47	0.19	2.06	2.36	2.71	3.26
0.8 [wt%]	3.41	0.25	1.68	2.03	2.44	3.13
1.2 [wt%]	3.45	0.49	0.94	1.32	1.86	2.95
1.4 [wt%]	3.01	0.52	0.56	0.87	1.36	2.45

Table 2. The allowable breakdown probability depending on the amount of additive(100 °C).

Unit: [MV/cm]

Classification	E_{ave}	σ	Breakdown probability [%]			
			0.01	0.1	1.0	20
Virgin	1.62	0.09	0.97	1.15	1.27	1.52
0.2 [wt%]	1.63	0.08	1.05	1.18	1.33	1.56
0.4 [wt%]	1.73	0.07	1.14	1.27	1.42	1.64
0.6 [wt%]	1.70	0.07	1.12	1.26	1.40	1.63
0.8 [wt%]	1.69	0.09	1.00	1.15	1.32	1.59
1.2 [wt%]	1.69	0.08	1.08	1.22	1.37	1.62
1.4 [wt%]	1.60	0.09	0.92	1.07	1.23	1.49

4. CONCLUSION

This study examined the electrical properties of epoxy nano-composites depending on the nano SiO_2 content and measuring temperature. The dielectric strength increased with increasing amount of nano SiO_2 to 0.4 wt%. The rate of increase in the dielectric strength decreased from 20 % to 5.5 % with increasing measurement temperature from room temperature to 100 °C. When the nano SiO_2 content was increased, the Coulomb barrier effect occurred due to the decreased distance between the nano particles in the epoxy resulting in an increase in the dielectric strength.

When the additive exceeded 0.4 wt%, the dielectric strength_decreased due to an increase in conductivity as a result of condensation between particles. When the breakdown probability in the specimens was 20 %, there was an allowable breakdown strength within the error range of the average breakdown strength. It was confirmed that a breakdown probability up to 20 % produces stable breakdown strength in the insulating device design.

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