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# Evaluation of the electrical and thermal characteristics of Ag/CuO – epoxy composites

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**This paper presents the manufacture and testing of nano/micro epoxy resins and their properties. In order to prepare the specimens, a main resin, a hardener, an accelerator and a nano/micro filler were used. Varying amounts of CuO and Ag nano fillers were added to the epoxy mixture along with a fixed amount of micro silica. The thermomechanical and electrical properties of these mixtures were then determined by DC breakdown (DCBD) test and TMA analysis. The DCBD strength value of the composites can be described by a Weibull distribution. Totally, the epoxy resin exclusively filled with the nano particles had better electrical properties than micro composite in which there isn't nano filler. However, according to the quantities and type of filler, the properties of the epoxy composites were not found to be linear or proportional. There were optimal combinations and ratios. This results can be explained by Nordheim's rule. And it was confirmed that thermal expansion coefficients (TEC) of nano-epoxy composites have similar value with Al or Cu.**

**Key words: Epoxy resin, Nano filler, Thermal expansion coefficient, Dielectric breakdown.**

## Introduction

Epoxy resin is widely used as an insulating material for electrical power devices. With the continued development of industry, a requirement for the improvement of the electrical properties of materials has become apparent. For this reason, research on materials that have superior strength and characteristics has been conducted. Recently, a new field known as nano science and technology has emerged through notable researches, advancements that reveal new physical phenomena in the nano-sized ultra-micro region, and improvements of material properties.

Recent advances intended to improve certain characteristics through the decrease of cure shrinkage, thermal response, TEC and increasing the electrical properties, mechanical strength, the breakdown voltage, wear-resistance, and thermal conductivity by applying composite materials to epoxy resins.

This work presents the DCBD property and thermomechanical analysis (TMA) of Ag/CuO composites. In order to calculate the temperature dependence of DCBD, the dielectric breakdown tests were done at 50 °C and 100 °C. And Weibull statistics is introduced to analyze the DCBD property.

## Experimental

### The preparation of specimens

The experimental process is based on the method used to manufacture resin for molded transfers. Epoxy resin, nano/micro fillers, hardener and an accelerator are mixed together to form nano/micro composites. Table 1 presents the formulation ratios of the specimens. One micro composite and six nano/micro composites were prepared. Ag and CuO nano fillers were mixed at 3, 5 and 7 phr. After the epoxy resin and nano/micro fillers were mixed, a hardener and accelerator were mixed using a high speed emulsifier for 15 and 30 minutes, respectively, at 60 °C. The composite was then mixed using a Planetary Centrifugal Mixer for 20 minutes. In order to make the specimens 1 mm thick, an epoxy molder was used, as shown in Fig. 1. The manufactured mold was underwent casting, curing, and cooling in a vacuum oven, at 80 °C, 130 °C, and room temperature over 4, 16 and 6 hours, respectively.

When making the specimens, carbon black was painted onto the face of the molder in order to avoid cracks and cavities in the epoxy resin. A mold release agent, chlorinated ethylene, was applied between the

**Table 1.** The specimen formulation ratios. [unit : phr]

| | MC | A3 | A5 | A7 | C3 | C5 | C7 |
|---|---|---|---|---|---|---|---|
| Resin | 100 | 100 | 100 | 100 | 100 | 100 | 100 |
| Hardener | 82 | 82 | 82 | 82 | 82 | 82 | 82 |
| Accelerator | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 |
| Micro-silica | 335 | 335 | 335 | 335 | 335 | 335 | 335 |
| Nano-Ag | X | 3 | 5 | 7 | X | X | X |
| Nano-CuO | X | X | X | X | 3 | 5 | 7 |

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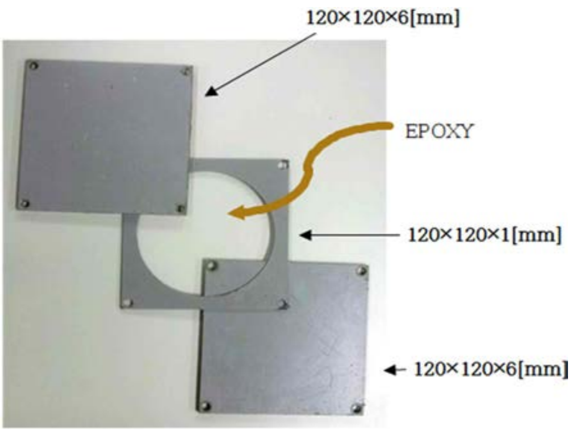


**Fig. 1.** The epoxy molder.

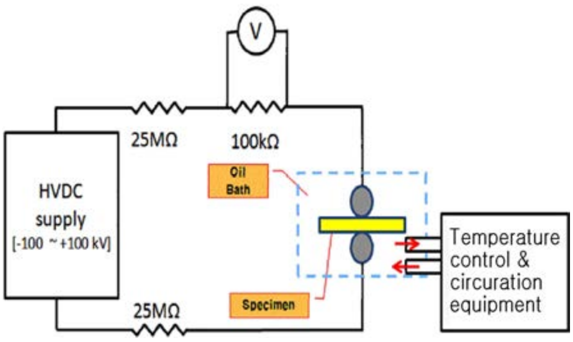


**Fig. 2.** The dielectric breakdown test schematic diagram.

molder and the epoxy resin in order to facilitate the removal of the epoxy resin from the molder.

### The weibull distribution

Weibull distributions are mainly used to assess the degradation of machines. Generally, in the case where there are n data $X_i$ (i = 1,2,3,…,n), the average X and standard deviation σ are:

$$X = \frac{(X_1 + X_2 + \cdots + X_n)}{n}$$

$$\sigma = \sqrt{\frac{(X_1 - X)^2 + (X_2 - X)^2 + \cdots + (X_n - X)^2}{n}} \quad (1)$$

A Weibull distribution has an effective value x for the random variable X:

$$F(x) = 1 - exp\left[-\left(\frac{x - x_l}{\eta}\right)^{\beta}\right] \quad (x \geq x_l)$$

$$= 0 \quad (x < x_l) \quad (2)$$

Where $x_l$, $\eta$, $\beta$ are the location parameter, scale parameter and shape parameter, respectively. $x_l$ is the minimum value in regards to the dielectric breakdown. $\eta$ corresponds to an average breakdown failure ratio of 63.2%. Since â determines the distribution shape of the accumulated probability of failure, $\beta$ and the degree of diffusion are inversely proportional. For example, if $\beta$ is 1 or 4, the Weibull distribution respectively follows either an exponential distribution or a normal distribution.

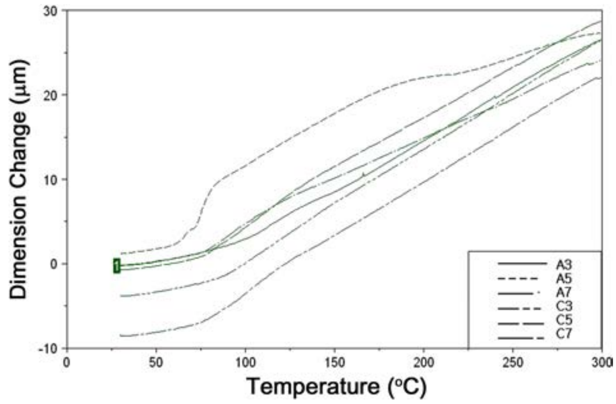


**Fig. 3.** The TMA analysis results.

**Table 2.** Thermal expansion coefficients.

| Specimen | TEC × $10^{-6}$/K |
|---|---|
| A3 | 40.5 |
| A5 | 34.2 |
| A7 | 28.4 |
| C3 | 32.7 |
| C5 | 30.4 |
| C7 | 30.4 |

### The dielectric breakdown test

The apparatus used to determine the dielectric breakdown is shown in Fig. 2. Two 25 MΩ resistors and a 100 MΩ resistor are used in the circuit. An oil bath is filled with non-aerated oil in order to prevent creeping discharges. There are two temperature conditions. One is below the glass transition temperature (Tg) of 50 °C and the other is over Tg at 100 °C. Temperature control and circulation equipment is used to maintain the oil temperature. The voltage rise was 1 kV/s; the voltage was measured until the insulation of the dielectric materials fails. Ten measurements per specimen type were performed. The average breakdown voltage was derived by Weibull distribution. It is recommended that the diameter of the electrode be 10 times larger than the gap. Therefore, 15 mm diameter stainless steel sphere-sphere electrodes were adopted.

## Results and discussion

### The TMA analysis results

Fig. 3 shows the results of the TMA analysis. Because epoxy resins have glassy and rubbery states at the glass transition temperature (Tg), the epoxy resin properties greatly change at Tg. In Fig. 3, all of the

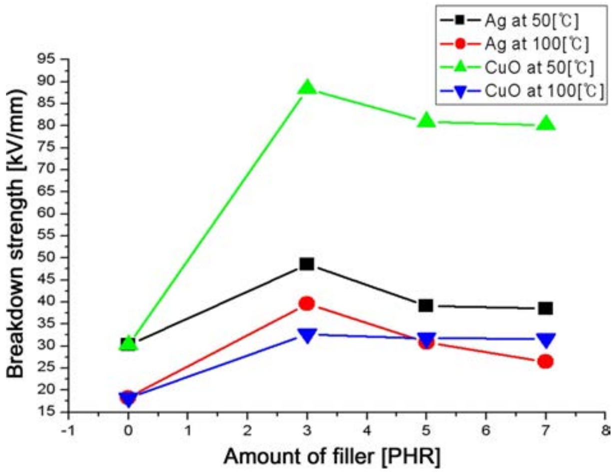


**Fig. 4.** The breakdown strength of the nano filled specimens.

specimens have a steep gradient from 60 °C to 90 °C. Therefore the dielectric breakdown tests were done at 50 °C and 100 °C. Table 2 shows the TEC. The temperature range for this test was 30 °C ~ 300 °C. Even though the epoxy resin is thermosetting, because of its TEC, specimen MC has an unstable value over the range of 30 °C ~ 300 °C, this makes the TEC of specimen MC unmeasurable. Otherwise, all of the other specimens filled with nano filler had measureable TECs. It is thought that the nano filler makes the epoxy resin resistant to heat. The epoxy resins filled with CuO have a uniform value of $30.3 \times 10^{-6}$/K ~ $32.7 \times 10^{-6}$/K, however, the epoxy resins filled with Ag have widely ranging TECs. Generally, the TEC of Cu is $17 \times 10^{-6}$/K and the TEC of Al is $23 \times 10^{-6}$/K. Therefore, it is supposed that the epoxy resin using the Ag filler is good for Al devices. Since the TEC of epoxy can be controlled by the nano filler content, it is thought that Ag fillers can be used for various materials that have different TECs. However, CuO is good when other characteristics are required with its fixed TEC value.

### The dielectric breakdown analysis

Because the glass transition temperature had a range of 60 °C ~ 90 °C, as shown in Fig. 3, the dielectric breakdown tests were done at 50 °C and 100 °C. And the results of the dielectric breakdown were arranged by weibull distribution. Fig. 4 shows the results of breakdown strength of specimens filled with the nano filler. Table 3 shows the breakdown strength, shape parameter, and temperature dependence coefficient (TDC). In order to compare the breakdown strength as it relates to changes in temperature, the TDC is defined below. In the equation (3) T is temperature and $E_b$ is the breakdown voltage.

$$TDC = \frac{1}{Eb(at\ 50°C)} \times \frac{\partial Eb}{\partial T} \times 100 \tag{3}$$

**Table 3.** The scale, shape parameters and TDC of the epoxy composites.

| Sample | 50[°C] | | 100[°C] | | TDC |
|---|---|---|---|---|---|
| | η[kV/mm] | β | η[kV/mm] | β | |
| MC | 30.19 | 21.00 | 18.29 | 19.33 | 0.79 |
| A3 | 48.57 | 8.99 | 39.57 | 13.07 | 0.37 |
| A5 | 39.04 | 15.22 | 30.79 | 9.04 | 0.42 |
| A7 | 38.51 | 9.58 | 26.47 | 10.80 | 0.63 |
| C3 | 88.40 | 9.86 | 32.71 | 7.23 | 1.26 |
| C5 | 80.90 | 6.81 | 31.74 | 9.52 | 1.22 |
| C7 | 80.20 | 5.92 | 31.65 | 6.53 | 1.21 |

The breakdown strength of the specimens filled with nano/micro filler is stronger than that found for the micro composite. According to the kinds and amounts of the filler, the breakdown strength was found not to be proportional. In the case where the same filler is used, the specimens that have the 3 phr nano filler have the strongest breakdown strength. And as the amount of the nano filler increases the breakdown strength decreases. This results can be explained by Nordiem's rule.

$$\rho_l = CX(1-X) \tag{4}$$

Equation (4) presents Nordiem's rule. C is coeffcient and X is a ratio of composites. When amount of nano filler is small ($X << 1$), resistivity is CX ($\rho_l = CX$) because nano filler act like impurity in composition. The mobility of composite is decrease by imputiry and then resistivity increase. After X is bigger than 1 ($X > 1$), resistivity $\rho_l$ decrease. It is supposed that nano-filler is no more impurity after X is bigger than 1.

The TDC is dependent on of the type of nano filler. A3, A5 and A7 have lower TDCs than the MC. Conversely, C3, C5 and C7 have higher TDCs than the MC. When the specimen is filled with Ag, the TDC is proportional to the nano filler amount. Conversely, in the CuO case, the TDC is inversely proportional to the amount of nano filler.

## Conclusions

This work aimed to increase the electrical and thermomechanical characteristics of epoxy resin. The electrical characteristics of nano filled epoxy composites have not been previously known because the reason that affects nano fillers in epoxy composite has not been turned out. Therefore, nano Ag and CuO were added in epoxy resin. The results can be summarized as follows:

1) Epoxy composite filled with nano fillers have different TECs. In the case of CuO epoxy composites, the TEC value was uniform. However, Ag composites have varied TEC values.

2) Nano CuO epoxy composites have higher breakdown strength than that found for micro composite. It is thought that the nano-CuO acts like impurity in composite. Therefore, there are optimistic combination and ratios.

3) The TDC of the Ag epoxy composite had the lowest value. Therefore Ag composite can be used for a stable insulation material.

It was confirmed that there are effects from the nano filler that make the electrical properties good but the degree of this effect is unpredictable. The filler amount is not proportional or inversely proportional. There are optimal ratios between filler and resin. Therefore, a study regarding the optimal ratios between nano filler and other materials and the theory of the phenomena should be conducted.

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