Effects of Temperature on Dielectric Breakdown Strength of Epoxy Compounds Filled with Natural Zeolite

You-Jeong Kim*, Hyeong-Ki Choi**, Sang-Wook Kim*

Abstract

To develop the better insulants, the zeolite particle, which is aluminosilicate mineral, was filled in the DGEBA/MDA/SN epoxy resin system. Dynamic DSC curves of cured specimens with various contents of zeolite were observed and the glass transition temperature(T_g) was obtained. According to this result, we could carry out the experiment concerned with the dielectric breakdown strength around T_g and find the limit temperature for the application of the DGEBA/MDA/SN system filled with natural zeolite. T_g increased with the content of zeolite. As the temperature increased, the dielectric breakdown strength decreased rapidly, in the region of T_g. At the high temperature, the deterioration by electrical stress was activated. The diameter of puncture at the high temperature was larger than that at the room temperature.

Key Words : Epoxy resin, Zeolite, Compounds, Electrical insulation, Temperature effect

1. Introduction

Cured epoxy resins are extensively used for the electrical insulation in high-voltage equipments. The bisphenol A-based epoxy resins cured with amine show, especially, good thermal properties and resistances. For technical and economic reasons, inorganic fillers are added in varying amounts to endow the required special properties. Although the influences of single component on the thermal and mechanical properties of the cured insulants are widely known, there is little information about the potential influence of the used resins and fillers on the electrical properties. So, in this case, we filled the natural zeolite in the DGEBA/MDA/SN compound and investigated the temperature effect on the dielectric breakdown voltage[1,2].

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Treeing in polymeric insulator is one of the main causes of dielectric breakdown and the tree inception and propagation mechanism have been extensively investigated to prohibit electric failure. But the treeing pattern is too complex and varies with several factors such as applied voltage, frequency, temperature and so on, so the developments of trees have not been described, analytically. Until now, tree shapes have been roughly characterized as branch, bush, dendrite type and so on[3,4]. To investigate the complex figure of real electrical trees in the specimen with the needle-plane electrode subjected to 60 Hz alternating current, we used the image analyser.

2. Experiment

2-1. Materials

The epoxy resin used in this study was DGEBA(Epon 828 grade) of Shell Co. and its EEW, MW and viscosity were 188 g/mol, 385 g/mol and 11,000~14,000 cps, respectively. 30 phr (parts per one hundred resin by weight) of MDA was used for curing the resin. SN(10 phr) as a new reactive additive was supplied from Fluka

Chemie AG. Natural zeolite(clinoptilolite type) from Kampo area in Korea[5] was used in various contents after sieving with #325 mesh and drying in 120 ℃ vacuum oven for 5 hrs. The average diameter of zeolite particle was 20.03 ㎛. Hard steel needle with tip curvature radius 3 ㎛ and tip angle 30° was used as high voltage electrode. The aluminum layer was used as counter plane electrode.

2-2. DSC analysis

DGEBA and SN(10 phr) were well mixed with dried natural zeolite(0, 20, 40, 60 and 80 phr) at 80 ℃, then MDA(30 phr) was added to the mixture. The mixture was cured at 150℃ for 1 hr after curing at 80 ℃ for 1.5 hr. The weighed sample(5~7 mg) was tested by DSC. The temperature rising rate was 10 ℃/min and nitrogen gas was flowed into the furnace at 80 ml/min. T_g was detected from the base-line shift on DSC curve.

2-3. Specimen preparation and test

The ultrasonically washed needle electrode with methyl alcohol and acetone were set in the mold with 1 mm electrode gap and the mold was preheated at 80 ℃. The mixture of DGEBA, SN and 40 phr of the dried zeolite was stirred and degassed at 80 ℃ for 1 hr to make zeolite evenly dispersed. And then the curing agent MDA was added to the mixture. The mixture was degassed and stirred until gel state. It was poured into the preheated mold at 80 ℃. The overall curing condition was 1st cure at 80 ℃ for 1.5 hr and then 2nd cure at 150 ℃ for 1.0 hr. The cured system was slowly cooled to the room temperature inside oven to prevent the formation of the microvoids and cracks by shrinking stress and sliced to 8 mm(w)×50 mm(H)×35 mm(L). The specimen was immersed in silicone oil at a constant temperature for 20 mins to suppress surface flashover. High voltage of 60 Hz was applied to the specimen until breakdown at the voltage rising rate of 500 V/sec and the voltage was measured as dielectric breakdown voltage. The dielectric breakdown step was detected by the breakdown sound or the flashover at the tip of needle electrode. Under the specific voltage, the electrical tree phenomena were observed by using image analyzer(Image Pro Plus).

3. Results and Discussion

3-1. DSC analysis

Fig. 1 shows the dynamic DSC curves for DGEBA/MDA/SN with zeolite contents(0, 20, 40, 60 and 80 phr) at the heating rate of 10 ℃/min. All diagrams do not show the exothermic curve above T_g and it means that the system was completely cured. With the increment of zeolite content, T_g increased by 16 ℃. The dielectric breakdown strength and mechanical properties strongly influenced by the thermal stability. The epoxy composite is vulnerable to the electrical stress above T_g, because it is a characteristic temperature determining the motion of end chain segments. The dielectric breakdown strength decreases at T_g. So that the increase in T_g of composite by adding zeolite may broaden the range of tolerance limit temperature to the electrical stress of this system[6,7].

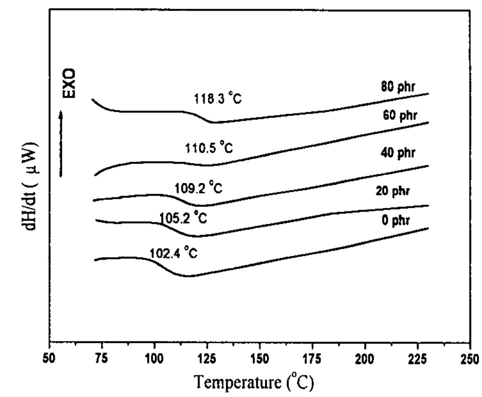

Fig. 1. Dynamic DSC curves at the heating rate of 10 ℃/min in the DGEBA/MDA/SN system filled with various zeolite contents (0, 20, 40, 60 and 80 phr).

3-2. Dielectric breakdown phenomena and dielectric breakdown strength

Fig. 2 shows the dielectric breakdown puncture in the specimen of DGEBA/MDA/SN/zeolite(40 phr) composite at 150 ℃. In this figure, we can see that the neighborhood of puncture and surface of the puncture became carbonized by the heat originated by high electrical stress. Fig. 3 shows also the dielectric breakdown puncture in the specimen of DGEBA/MDA/SN/zeolite(5 phr) composite at the room temperature. In this specimen, the carbonized region is less than the Fig. 2 and the diameter of the puncture is also less than the Fig. 2. In the comparison of these figures, we can deduce the following facts. At the high temperature, the deterioration of specimen is accelerated by the heat of surroundings and the heat originated by the collisions between the hot electrons emitted from the tip of needle electrode and the chains of molecular bond. At the high temperature, the molecular chain will be more mobile so that the specimen become more soften and vulnerable to the electrical stress than at the room temperature. By this reason, the puncture occurred very easily and become larger than at the room temperature. Cross-linked epoxy resin is brittle under the temperature lower than T_g, so that fan type crack was occured by impact waves from discharged energy. But at higher temperature(150 ℃) than T_g(109 ℃), crack did not occur and growing tree

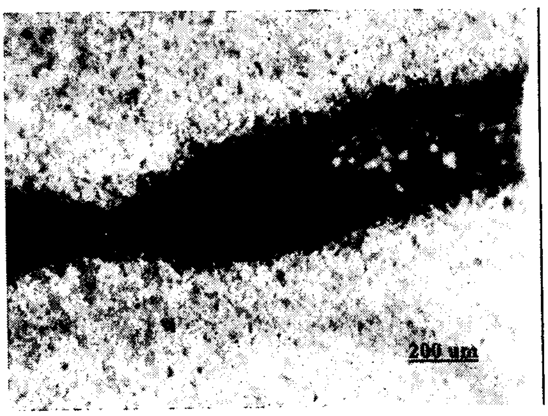

Fig. 2. Dielectric breakdown puncture of DGEBA/MDA/SN/zeolite(40 phr) at 150 ℃.

Fig. 3. Dielectric breakdown puncture of DGEBA/MDA/SN/zeolite(5 phr) at the room temperature.

around the main breakdown channel was not observed. It can be inferred that the impact resistance reinforced DGEBA/MDA/SN/zeolite composite could absorb the impact waves more effectively at higher temperature than T_g.

In the case of the tree growth, the deterioration by partial discharge will become easier in the specimen of high temperature by the same reason. So that the generated tree is more complex than in the room temperature. Fig. 4 shows the tree generated in DGEBA/MDA/SN/zeolite(20 phr) at the room temperature. The path of tree is complex like the dielectric breakdown pattern in Fig. 3.

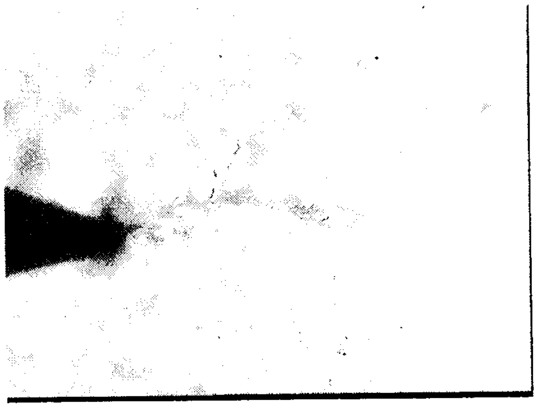

Fig. 4. The tree growth in the specimen of DGEBA/MDA/SN/zeolite(20 phr) at the room temperature.

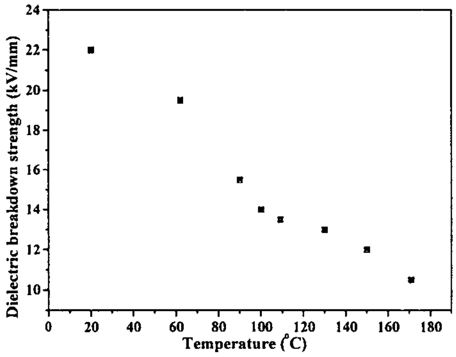

Fig. 5. The dielectric breakdown strength with various temperature of DGEBA/MDA/SN/zeolite(40 phr) composite.

Fig. 5 shows the dielectric breakdown strength (DBS) with various temperature in the specimen of DGEBA/MDA/SN/zeolite(40 phr) at the scanning rate of 500 V/sec until breakdown was measured. Rapid decrement of DBS at the temperature lower than the glass transition temperature(T_g : 109℃) was observed. The dielectric breakdown characteristics depend on the temperature where the movement of the molecular chains become active and the electron diffusion region from the needle electrode is enlarged. When the temperature increases, discharged electrons from the needle tip are trapped deeply inside and forms space charge region widely and electrical stress becomes relaxed. If the electric field at the tip of the space charge region reaches tree initiation voltage, pits occur and grow. Tree initiates when a pit grows and propagates toward the counter-electrode and then dielectric breakdown occurs. So the abrupt decrement of DBS was observed at the temperature lower than T_g[8,9].

4. Conclusions

From the study on the effects of temperature on dielectric breakdown strength of epoxy compounds filled with natural zeolite, the following conclusions were obtained. In the region of T_g, the dielectric breakdown strength decreased by the activated movement of molecular chain. T_g increased with the content of zeolite. So the tolerance limit temperature was increased. The diameter of the puncture in high temperature was larger than in the room temperature. The dielectric breakdown phenomenon was more vigorous in the high ambient temperature.

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