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경화제 함량과 후기경화조건에 따른 DGEBA/MDA/SN계의 절연열화 특성

(Effects of Curing Agent Content and Post-curing Conditions on Dielectric Deterioration Characteristics of DGEBA/MDA/SN System)

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Abstract : The effects of aromatic curing agent of MDA contents and post curing conditions on dielectric deterioration characteristics of DGEBA/MDA/SN system were investigated. The dielectric properties were measured by using needle-plane electrode geometry under the commercial AC high electric field application. As the curing agent content increased, the dielectric breakdown strength increased and then decreased slightly. All the trees initiated from the tip of needle electrode and the shape of the tree in this system was a dendrite type.

1. Introduction

The consumption of electricity increased rapidly with the stable growth of economy and it requires the credibility of electrical power systems for supplying economical and stable electric power. Researchers have tried to develop a new electrical materials to extend the life expectancy and to improve the insulating properties of dielectrics by adding new reactive additives or nonreactive fillers [1]. The epoxy resin system has a good combination of electrical, mechanical, thermal properties and have been processed in types of molding, impregnating, lamella, thin layer, etc [2]. Dielectrics must frequently serve as a structural material as well as an insulating material. But the conventional epoxy resin system was too brittle because of its high cross-linking density. We reported the impact resistance improved new epoxy resin system by adding new reactive additives as a chain extender [1,3]. The long-term dielectric breakdown phenomena by electrical treeing deterioration in polymeric insulator under inhomogeneous high electric fields attributed to electrically conducting inclusions, rough electrode surfaces, voids, cracks etc. are one of the main causes of electric failure. To suppress the treeing phenomena, the initiation mechanism and propagation characteristics have been investigated worldwide. The dielectric breakdown by treeing deterioration is governed by various factors such as electrode material, applied charge type, polymeric structure and environmental condition [2,3]. To design a good electrical insulator, all the factors should be considered. In this study, the

dielectric breakdown strength and deterioration phenomena in needle electrode embedded DGEBA/MDA/SN system at various MDA contents under AC high electric field were observed at various test conditions. The phenomena of treeing at the tip of the needle electrode were photographed through the optical microscope.

2. Experiment

2-1. Materials

The epoxy resin of DGEBA(EPON 828) was supplied by Shell Chemical Co. Its EEW, MW and viscosity were 188, 385 and 11,000-14,000 cps(25℃), respectively. 22, 26 and 30 phr of MDA(methylene dianiline) was added as a curing agent. 15 phr of SN(succinonitrile) was introduced as a reactive additive. The needle electrodes of hard steel with diameter of 1 ㎜, tip curvature radius of 3.0 ㎛, tip angle of 30° were used. The electrodes were carefully washed ultrasonically in acetone and then dried under nitrogen gas to insure the best possible bonding with epoxy resin.

2-2. Specimen Preparation

Needle electrodes were set in the mold with electrode gap of 1.0 ㎜. The mixture of pretreated reactants was poured into the mold and cured at 80℃ for 1.5 hr and then 2nd cured at 150℃ for 1.0 hr. The system cured at the elevated temperature was cooled down slowly inside the oven to minimize the occurrence of microvoids by the difference of thermal expansion coefficient and cut into 30 ㎜ square blocks with 8 ㎜ thickness and observed under the microscope(×100). Specimens without defects such as voids and cracks around needle tip were selected and used. The specimens were post-cured at the same condition with 2nd curing condition.

2-3. Dielectric Breakdown Test

The specimen was immersed in the silicone oil to prevent surface discharge at high electric field. The commercial AC voltage was applied to the specimen until dielectric breakdown at the voltage rising rate of 500 V/sec and the dielectric breakdown voltage was measured. Under a specific voltage the electrical tree propagating phenomena were observed.

3. Results and Discussion

Fig. 1 shows the DSC curves of cured DGEBA/MDA/SN system at various MDA contents. The stoichiometric content of MDA with DGEBA is 26 phr. When the curing agent is 22 phr the thermal properties of glass transition temperature was too low of 53.9℃. All the epoxide functional groups couldn't participate in the curing process. As the curing agent content increased, the glass transition temperature increased and when 30 phr of MDA was introduced the glass transition temperature was 95.1℃. There might be the residual curing agent which is unreacted and

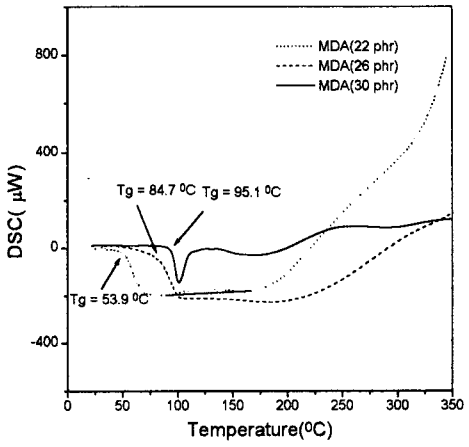

Fig. 1 Effects of MDA curing agent content on DSC thermograms at the heating rate of 10 ℃/min with N_2 gas flow rate of 50 ml/min

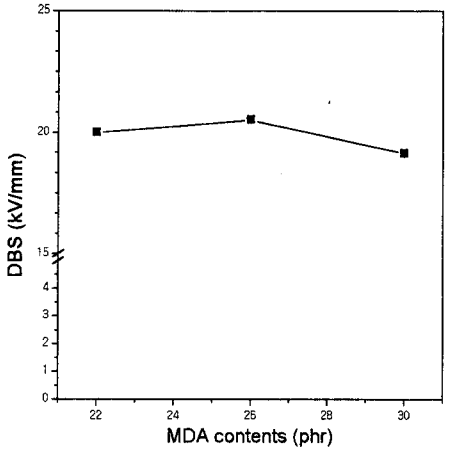

Fig. 2 The delectric breakdown strength dependence on the curing agent content.

may act as the impurities on dielectric characteristics. The effects of MDA contents on dielectric breakdown characteristics of epoxy resin DGEBA/MDA/SN system is the topic of this study.

Fig. 2 shows the effects of MDA contents on dielectric breakdown strength. The dielectric breakdown strength was measured by the dielectric breakdown voltage divided by the electrode separation gap. The magnified electric field when the dielectric breakdown occurs can be approximately estimated by using Mason equation at the tip of needle electrode in plane-needle inhomogeneous electric field. As the MDA content is higher than stochimetric concentration, the dielectric breakdown strength decreased sightly. It is thought that the residual curing agent acted as the impurities, though, the cured resin became more thermally stabilized. As the contents of impurities increases the electrical stress is concentrated on it ealily and the dielectric deterioration is initiated from the weakest point such as impurities and defects of voids, cracks, dust, metallic particles, and so on.

Fig. 3 Fracture surface of treeing deteriorated epoxy resin system

Fig. 3 shows the fractured surface of treeing deteriorated epoxy resin system. As a tree channel reached the counter-plane electrode the dielectric breakdown occurred. The electrical treeing shape was dendrite type. The inner-surface of tree channel was eroded and carbonated. when the electrical tree reached the counter outer electrode, hugh amount of electrons were discharged and the cracks inside polymeric insulator was occurred by the electron pulse shock. The electrical treeing resistance can be improved by changing the curing conditions. Also, the mechanism of space charge formation and distribution inside the dielectrics should be investigated to analyze the dielectric breakdown mechanism precisely[4].

Fig. 4 shows the DSC thermograms of DGEBA/MDA/SN system at various post or second curing time. When the post-curing time increased the DSC curve shifts toward right

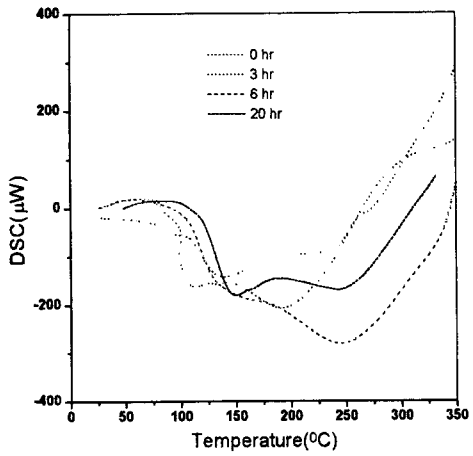

Fig. 4 Effects of post-curing time on DSC thermograms of DGEBA/MDA(30 phr)/SN(15 phr) system.

and the thermal properties of Tg increased. From our previous reports, the thermal properties decreased when the system post cured at high temperature too long[3]. The system is thermally deteriorated and the process is accelerated under the presence of oxygen. Thermal oxidation at low temperature can lessen the dielectric breakdown properties and mechanical properties[5]. All this test results will be presented in the presentation.

4. Conclusions

From the test results of the dielectric breakdown characteristics in DGEBA-MDA-SN system at various MDA contents under high electric field, following conclusions were obtained.

1. As the MDA contents increased the thermal properties of glass transition temperature increased
2. The dielectric breakdown strength was decreased with the increase of MDA contents higher than stochiometry slightly.
3 The tree shape was too complex to characterize and dendrite type.
4. As the post-curing time increased Tg increased.

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