

## Characteristics of Polymer Insulator Materials - Behavior of Dielectric Breakdown in DGEBA/MDA/Nitrile System -

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### 고분자 절연재료의 특성연구 - DGEBA/MDA/Nitrile계에서 절연파괴 거동 -

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**초 록** 에폭시 수지 DGEBA/MDA/GN 계의 트링 열화에 미치는 전압과 온도의 영향에 대한 고찰을 통하여 고분자 절연 재료의 절연파괴 거동을 연구하였다. 전압 인가시간에 따라 트리의 성장속도는 역S자의 형태로 나타났다. 인가전압과 온도가 증가함에 따라 트리의 성장속도는 증가하였으며 트리형상의 복잡함, 즉 국부적인 트링열화의 정도는 인가전압이 감소하거나 온도가 증가한 경우에 높게 나타났다. 절연파괴 강도는 유리 전이온도 이상에서 급격히 감소하였다.

**Abstract** The effects of voltage and temperature on tree deterioration in DGEBA/MDA/GN system were investigated to understand behavior of dielectric breakdown in polymeric insulating materials. Growth rate of tree with time appeared in the form of a reversed sigmoid shape when voltage was applied to the epoxy resin system. Growth rate of tree was decreased and the tree shape became complicated with the decrement of applied voltage. As temperature increased, growth rate of tree increased, dielectric breakdown strength decreased and tree shape were more complicated. In particular, in the vicinity of  $T_g$ , dielectric breakdown strength decreased rapidly.

### 1. Introduction

Polymeric insulating materials would be broken or deteriorated with using for a long time. These long periodic deterioration and breakdown of electrical insulating materials subjected to AC voltages can be usually associated with electrical tree phenomena<sup>1)</sup>. Electrical trees are produced by partial discharge activities in small defects, such as cracks, conducting particles or cavities inside the insulating materials. These generated trees propagate continuously and then breakdown phenomena occur finally. Therefore, it is very important to understand treeing deterioration as one of the main causes of dielectric breakdown for the development or for the design of good insulating materials<sup>2, 3)</sup>.

In this study, epoxy resin system was used as polymeric materials and GN(glutaronitrile) as a nitrile system was introduced to DGEBA(diglycidyl ether of bisphenol A)/MDA(4,4'-methylene dianiline) system

for the purpose of improving toughness. Of the factors which affected electrical tree shape, voltage and temperature dependences of tree deterioration in modified epoxy resin system were investigated by using needle/plane electrodes.

### 2. Experiment

#### Materials

Epoxy resin was DGEBA type and MDA, one of aromatic group having heat resistance, was used as curing agent. GN, as a reactive additive, was used to extend main chain length of epoxy resin system and to improve toughness. The angle and the radius curvature of needle tip were 30° and 5  $\mu\text{m}$ , respectively.

#### Procedure

A needle was inserted into DGEBA/MDA/GN(10phr) system and then the system was cured at 150 °C for 1.0 hr after being cured at 80 °C for 1.5 hr. The dis-

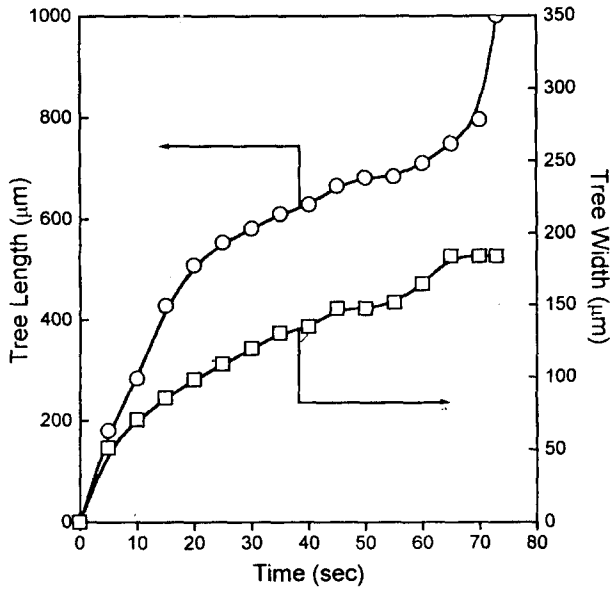


Fig. 1. Leading tree length as a function of applied time under 12.5 kV.

tance between needle electrode and plane electrode kept to be 1 mm. The cured system was annealed at the rate of  $-1\text{ }^{\circ}\text{C}/\text{min}$  until room temperature in order to minimize the formation of voids subjected to different thermal expansion coefficients between the metal needle and epoxy resin system. After cutting and grinding specimens in the size of  $30 \times 30 \times 1\text{ mm}$ , various voltages

were applied to the modified epoxy resin system at different temperatures. The specimens were immersed in silicon oil during test to prevent surface discharge and they removed periodically from the circuit to determine the tree length and its shape with an image analyzer.

### 3. Results and Discussion

Fig. 1 shows the leading tree length as a function of time when the voltage of 12.5 kV was applied to DGEBA/MDA/GN system. Tree length and tree width were expressed as the longest length of tree directed to plane electrode from the needle tip and the vertical length of the longest tree, respectively. If the growth rate of tree was defined as the tree length per unit time, tree length increased rapidly in the beginning and final stage. But, the growth rate of tree was decreased in the middle stage. Thus, the growth rate of tree length presented with a reversed sigmoid shape. The growth rate of tree width was increased and then were decreased gradually. The growth of tree width stopped above 65 sec.

The phenomena of tree propagation, when 12.5 kV was applied to DGEBA/MDA/GN system, are shown in Fig. 2. In the beginning stage, tree propagated linearly toward plane electrode and the branched tree is formed and grown with increasing the voltage applying

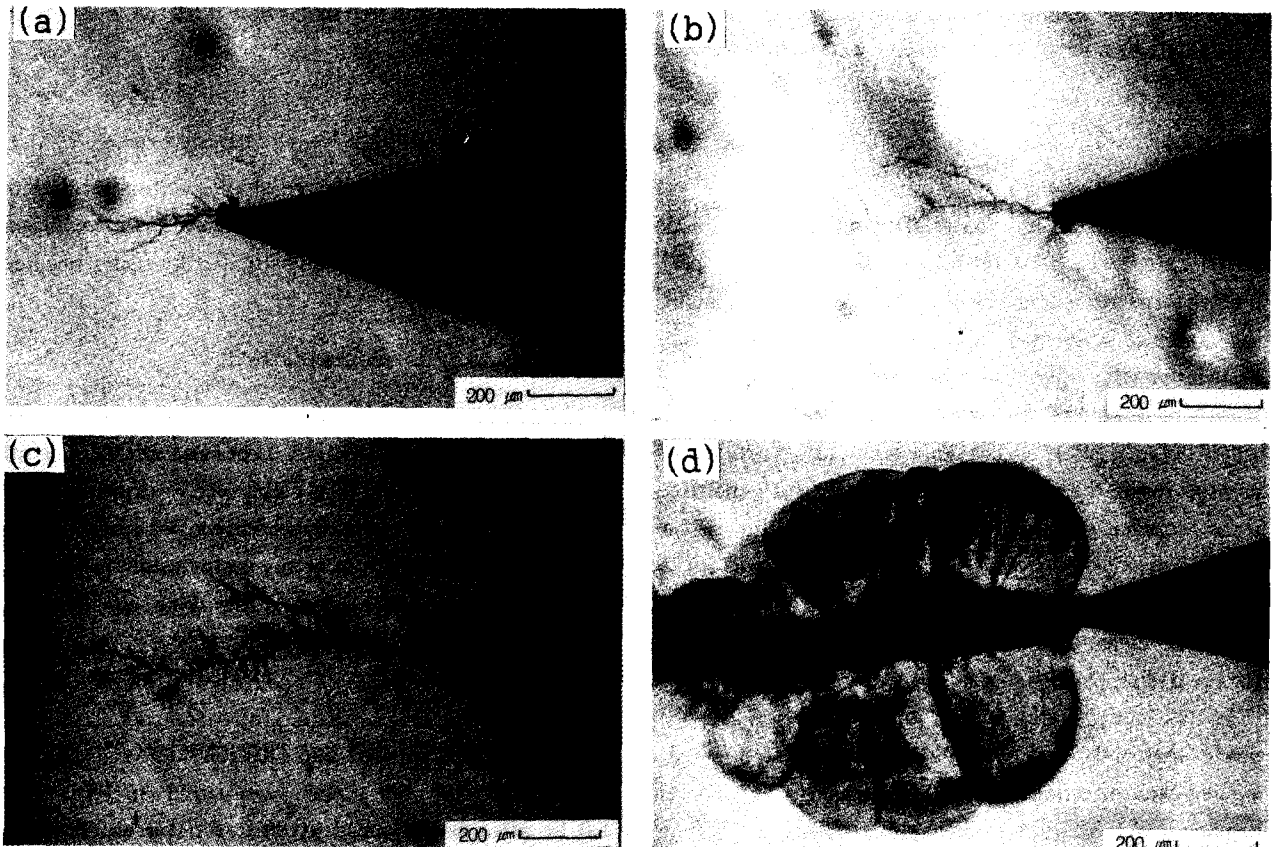


Fig. 2. Phenomena of tree propagation in DGEBA/MDA/GN system. (a) 10 sec, (b) 30 sec (c) 50 sec and (d) 72 sec

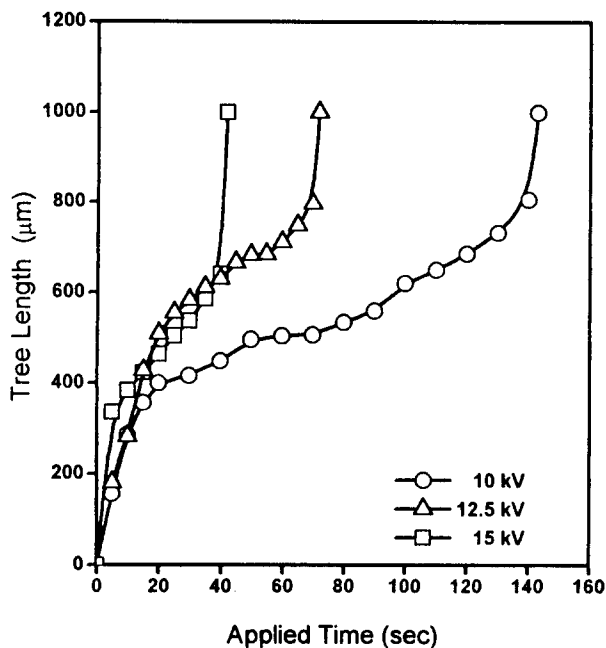


Fig. 3. Leading treeing length as a function of applied time under various voltages.

time. At the voltage applying time of 72 sec, breakdown finally occurred. These results can be explained as follows: After the voltage was applied, a strong electric field near the needle electrode was enhanced by the accumulated charge deteriorating the epoxy resin system easily so that tree was initiated and grew linearly immediately. The equivalent curvature radius of electrical tree tip got larger during the growth of tree since local field around the tree tip was influenced by the curvature radius of the tree tips. Therefore, the growth rate of electrical tree decreased as the local electrical field around the tree tip grew weak. Also, continuously injected electrons were trapped on the walls of the tree channel and new space charges were created so that branched trees were formed<sup>(6)</sup>. In the second step, as voltage applying time increased, many branched trees were formed and the growth rate of tree length decreased. However, as electrical tree grew near to the plane electrode, maximum local field became higher again and growth rate of tree also became faster. Therefore, it is considered that the electrical growth rate of tree showed a fast-slow-fast tendency and the electrical tree propagate with the form of a reversed sigmoid shape as voltage was applying time increased.

The growth rate of tree width was fast in the initial step while it was slow with applying time. Because all electrical trees propagate toward plane electrode as voltage was applied to the epoxy resin system, generated branched tree also propagated toward plane electrode.

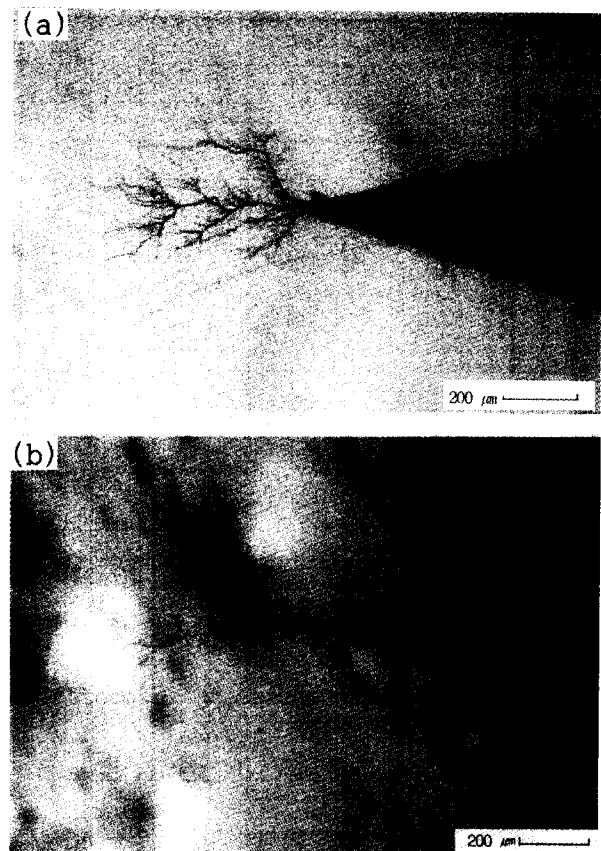


Fig. 4. Phenomena of tree deterioration under different voltages. (a) 10 kV and (b) 12.5 kV.

Thus, the growth rate of tree width decreased and then kept constant.

#### Voltage dependence

Fig. 3 shows the leading treeing length when different voltages were applied to DGEBA/MDA/GN system. When voltages of 10, 12.5 and 15 kV were applied to epoxy resin system, it showed breakdowns occurred at 143, 72 and 42 sec, respectively. All tree propagation displayed the reversed sigmoid shape.

The growth rate of electrical tree increased and the retarded term of tree decreased with the increment of applied voltage. As the higher voltage was applied to epoxy resin system, the inhomogeneous high electric field was formed. Thus, the deterioration of epoxy resin system was accelerated under the higher voltage and then its breakdown happened faster than under the lower voltage.

Fig. 4 shows the electrical tree shape when different voltages were applied to DGEBA/MDA/GN system. The electrical tree shapes got simpler with the increment of applied voltage. Since the electrons were trapped deeply toward the plane electrode under high voltage, the rate of tree propagation was accelerated

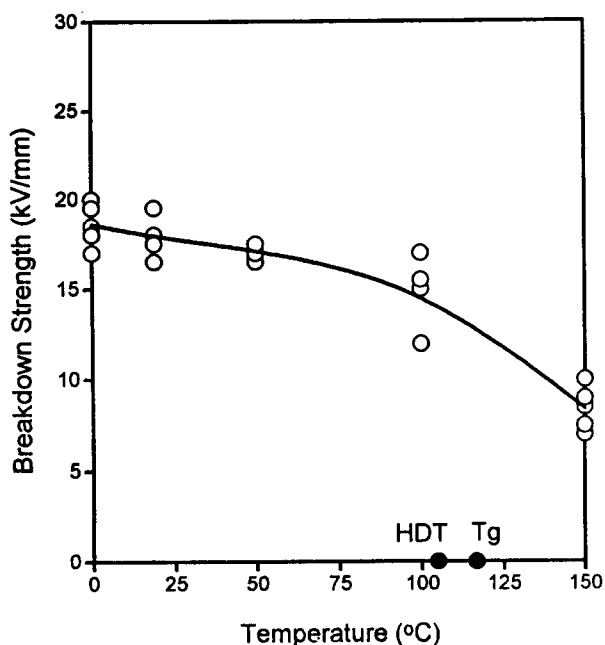


Fig. 5. Dielectric breakdown strength in DGEBA/MDA/GN system as a function of temperature.

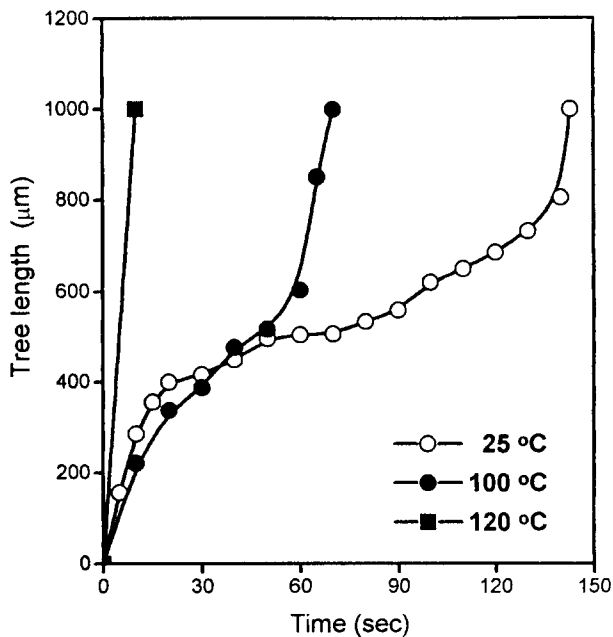


Fig. 6. Growth rate of tree as a function of applied time under 10 kV at different temperatures.

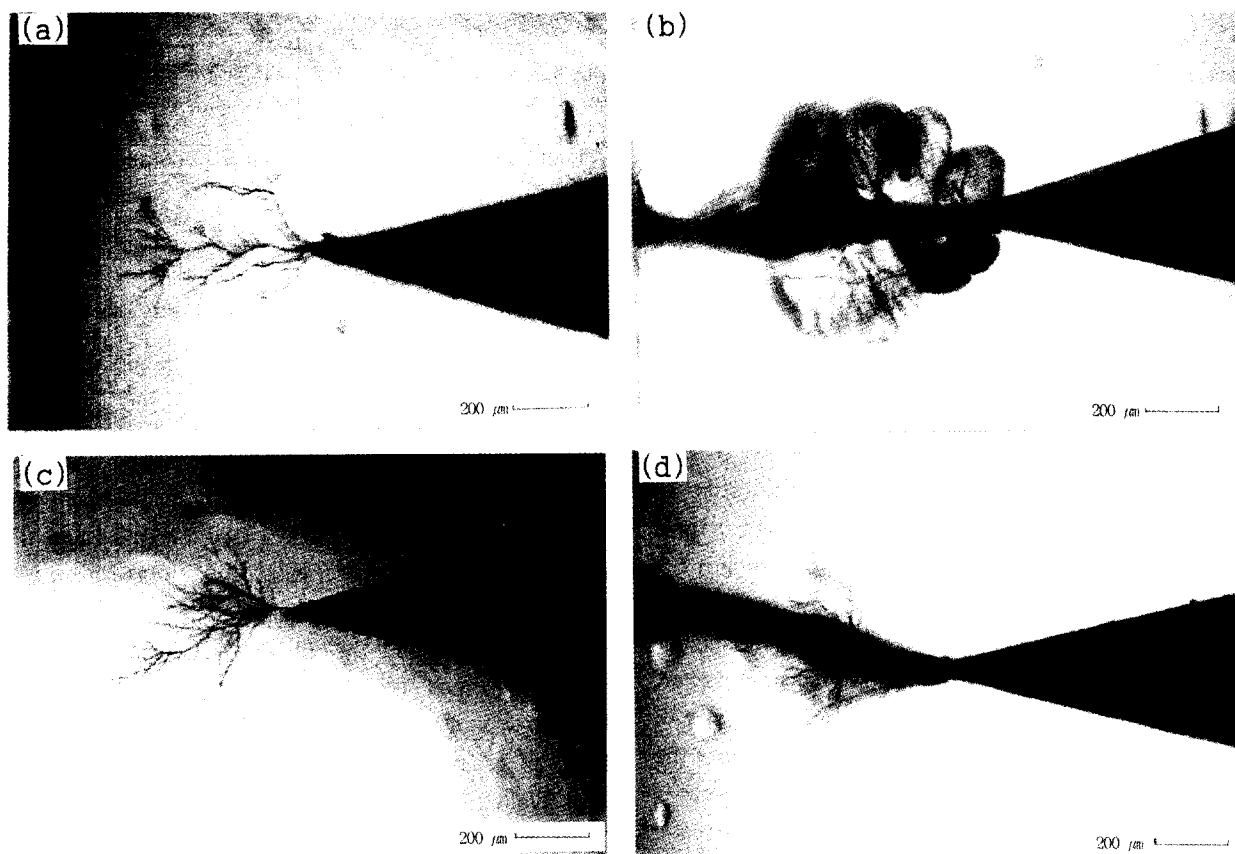


Fig. 7. Tree phenomena at different temperatures. (a) tree grown at 25°C, (b) dielectric breakdown at 25°C, (c) tree grown at 100°C and (d) dielectric breakdown at 100°C.

and the retarded term of tree got lessened. Thus, electrical tree consisted of low-order channels at high vol-

tage. However, under lower voltage, time for generation and propagation of branched tree in the main tree

slow increased because the growth rate of tree was low. Therefore, the tree shape was more dense<sup>9,10</sup>.

#### Temperature dependence

Fig. 5 shows the dielectric breakdown strength in DGEBA/MDA/GN system as a function of temperature. To investigate temperature dependence of dielectric breakdown strength in DGEBA/MDA/GN system, voltage was applied to epoxy resin system at the rate of 500 V/sec at various temperatures. The breakdown strength decreased with the increment of temperature. In particular, dielectric breakdown strength decreased sharply in the vicinity of glass transition temperature (below  $T_g$ , 117°C). With the increment of temperature, molecular chains took microbrown motion easily so that injected electrons from needle tip were trapped deeply inside material and then tree inception voltage got lower and the growth rate of tree grew fast. In the vicinity of  $T_g$ , the rapid decrement of dielectric breakdown strength was in well accordance with theory of  $T_g$  at which the segments of molecular initiated microbrown motion. These results indicated that high temperature is one of the main role of dielectric breakdown<sup>11,12</sup>.

When the voltage of 10 kV was applied to epoxy resin system at different temperatures, the characteristics of tree length are presented in Fig. 6. At 25, 100 and 120°C, breakdown occurred at 143, 62 and 10 sec, respectively. As temperature increased, retarded term of tree propagation decreased and the growth rate of tree increased. Especially, the growth rate of tree was very fast at 120°C. These result is well consistent with the previous result.

When 10 kV was applied to epoxy resin system at different temperatures, the tree shape and dielectric breakdown shape are shown in Fig. 7. The number of branched trees at 100°C are more than those of branched trees at 25°C. These results show the reason why space charges were formed widely as temperature increased. Many fan-type cracks around the breakdown channel at low temperature were presented. However, at 100°C, no fan-type crack could be found and several electrical treeing branches were observed. These reason may be the same with followings. When

breakdown occurred, impact pulse by partial discharge led to produce fan-type cracks around breakdown channel. But, as temperature increased, impact pulse was absorbed more effectively. Thus, high temperature played an important role in the dielectric breakdown characteristics<sup>11</sup>.

#### 4. Conclusions

Tree length propagated with the form of a reversed sigmoid shape. The growth rate of tree width was increased and then were decreased gradually. The growth of tree width stopped above 65 sec. Growth rate of tree increased but tree shapes were more simplified with increasing the applied voltages. As temperature increased, dielectric breakdown strength decreased. In particular, dielectric breakdown strength decreased sharply in the vicinity of  $T_g$ . Growth rate of tree increased and tree shape became complicated with the increment of temperature. Above the temperature of  $T_g$ , the tree shape can not be analyzed because the tree growth rate was too fast.

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