TRANSACTIONS ON ELECTRICAL AND ELECTRONIC MATERIALS
Vol. 14, No. 5, pp. 278-281, October 25, 2013

Regular Paper

pISSN: 1229-7607 eISSN: 2092-7592
DOI: http://dx.doi.org/10.4313/TEEM.2013.14.5.278

Effect of Electric Field Frequency on the AC Electrical Treeing Phenomena in an Epoxy/Layered Silicate Nanocomposite

Jae-Jun Park†
Department of Electrical and Electronic Engineering, Joongbu University, Geumsan 312-702, Korea

Received September 9, 2013; Accepted September 13, 2013

The effects of electric field frequency on the AC electrical treeing phenomena in an epoxy/layered silicate (1.5 wt%) were investigated in a needle-plate electrode arrangement. A layered silicate was exfoliated in an epoxy-base resin with AC electric field apparatus. To measure the treeing initiation and propagation- and the breakdown rate, a constant alternating current (AC) of 10 kV with three different electric field frequencies (60, 500, and 1,000 Hz) was applied to the specimen in the needle-plate electrode specimen in an insulating oil bath at130℃. At 60 Hz, the treeing initiation time was 12 min, the propagation rate was 0.24×10^{-3} mm/min, and the morphology was a dense branch type. As the electric field frequency increased, the treeing initiation time decreased and the propagation rate increased. At 1,000 Hz, the treeing initiation time was 5 min, the propagation rate was 0.30×10^{-3} mm/min, and the morphology was a dense bush type.

Keywords: Electrical treeing, Epoxy nanocomposite, Layered silicate, Treeing phenomena, Electric field frequency

1. INTRODUCTION

Multilayered silicates have been used as nano-sized fillers in polymer nanocomposites, and the most well-known include montmorillonite, saponite, hectorite, talc, and mica [1-3]. Generally, they are naturally synthesized as bulk-material and they are pulverized into fine μm-size particles. To synthesize polymer nanocomposites, the multilayered μm particles should be separated into sheet-like monolayers in the polymer matrix, whose dimensions (20-1,000) × (20-1,000) × 1 nm^3. The driving force to separate them from each other is given by polymer penetration through the interlayers of the silicate monolayers. The polymer chains are in the form of coil-like conformation in the interlayers, which leads to intercalated or exfoliated nanocomposites [1,2]. However, it is very difficult for the polymer chains to penetrate into the hydrophilic interlayers. Therefore, the interlayers should be rendered organophilic by means of cation exchange with alkyl ammonium ions [2-5].

Epoxy resins are well-known materials in the field of insulation systems for heavy electric equipment, because they have good mechanical and thermal properties and excellent electrical properties [6-8]. When multilayered silicate particles are mixed with an epoxy resin, the epoxy resins and curing agents penetrate into the interlayers of the multilayered silicate and separate the sheet-like monolayers from each other in the epoxy matrix, forming an intercalated or exfoliated state.

To study the insulation performance of epoxy/layered silicate nanocomposites, many works have been done by studying the electrical treeing phenomena in order to estimate the insulation characteristics of neat epoxies or their nanocomposites. The treeing phenomena have often been considered to be the most important mechanism for the deterioration of polymeric insulators (such as high voltage polymeric cables) [9-12]. The treeing growth mechanism is divided into three processes: (1) the incubation process, (2) the initiation process, and (3) the propagation process. After electrical treeing is initiated, it propagates

† Author to whom all correspondence should be addressed:
E-mail: jjpark@joongbu.ac.kr

rapidly, and breakdown eventually occurs. Hence, the initiation time should be delayed, and the propagation rate should be retarded in order to obtain excellent insulation polymeric materials.

In this study, an epoxy/layered silicate nanocomposite for the insulation of heavy electric equipment was prepared by an electric field method, and the effect of the electric field frequency on the electrical treeing phenomena were studied with needle-plate electrodes.

2. EXPERIMENTS

2.1 Materials

YD 128 a commercial DGEBA (diglycidyl ether of bisphenol A)--type epoxy resin was used (Kukdo Chem. Co.). The equivalent weight was 184-190, and the viscosity was 11,500-13,500 cps at 25℃. The curing agent was HN-2200-grade Me-THPA (3- or 4-methyl-1,2,3,6-tetrahydrophthalic anhydride. Hitachi Chem. Co.). It is widely used in the field of electric insulation. The accelerator was BDMA (benzyl-dimethyl amine, Kukdo Chem. Co.). Cloisite® 10A (Southern Clay Products, Inc., USA) a natural montmorillonite organically modified with 2MBHT (dimethyl-benzyl-hydrogenated tallow quaternary ammonium salt), was used as a multilayered silicate. It wasdried at 110℃ for 24 h in a vacuum oven and stored in a desiccator before use. A needle-type steel electrode was purchased from Ogura Jewelry Co., Japan. Its diameter and length were 1 mm and 60 mm, respectively, with a tip angle of 30^o and a radius of curvature of 5 μm.

2.2 Specimen preparation for AC treeing test

To prepare epoxy/layered silicate nanocomposite, DGEBA (100 g) and Cloisite 10A (2.74 g) were mixed with ultrasonic homogenizer (20 kHz) for 30 min and then put into a custom AC electric field apparatus [14]. The AC electric field was generated by a high voltage (HV) generator in the following conditions: (1) inter-electrode distance: 50 mm, (2) application voltage: 11 kV, (3) frequency: 1 kHz, and (4) application time: 60 min. During the AC application time, the epoxy resin penetrated into the interlayers, making the layered silicate swollen. Then, the mixture was mixed with THPA (80 g) and BDMA (0.9 g). The content of the nano-silicate in the epoxy nanocomposite was 1.5 wt%.

The mixture was poured into a mold with a 15 × 15-mm^2 cavity with 30-mm height, in which a needle electrode was arranged beforehand to make the distance of the needle-plate electrodes 4.2 mm. Then, it was cured at 120℃ for 2 hr and post-cured at 150℃ for 2 hr, and then cooled slowly at a rate of -0.5℃/min to room temperature to avoid internal stress. Finally, the opposite-side of the needle electrode in the epoxy specimen was coated with conductive silver paste.

2.3 AC treeing test

To measure the treeing initiation and propagation rate, a constant alternating current of 10 kV with three different electric field frequencies (60, 500, and 1,000 Hz) was applied to the specimen in a needle-plate electrode arrangement in an insulating oil bath at 130℃. Then, the specimen was inserted into the insulating oil and maintained sufficiently for 2 hr until the temperature of the needle tip area reached the testing temperature. High voltage (HV) was applied by an AC Endurance Voltage Tester (Haefely, Germany) at a rising speed of 1 kV/s until 10 kV, and the test voltage was kept until electrical breakdown took place. The treeing

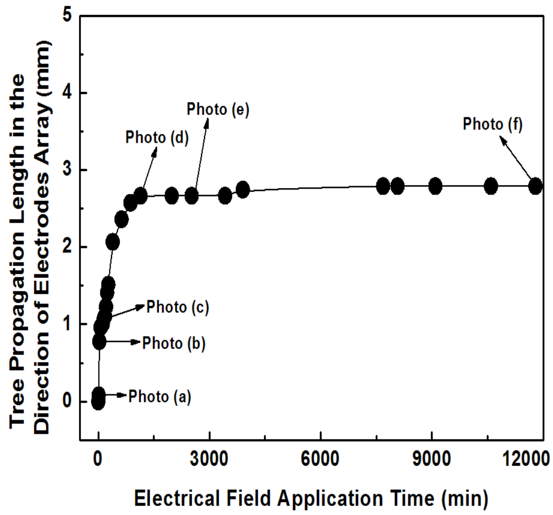

Fig. 1. Treeing growth rate in epoxy/layered silicate (1.5 wt%) system tested in the constant electric field of 10 kV/4.2 mm (60 Hz) at 130℃.

morphology was monitored by a video microscope system (ICS-305B, SOMETECH Inc.), and treeing images were collected every 1 min.

3. RESULTS AND DISCUSSION

In previous work [14], the exfoliated state in the epoxy/layered silicate nanocomposite was confirmed and observed using WAXD (wide-angle X-ray diffraction) analysis and TEM (transmission electron microscopy). Well-dispersed monolayers act as barriers to the treeing propagation. This means that the peeled monolayers were well dispersed in the epoxy matrix, and would act as barriers to treeing propagation.

In order to study the barrier effect of the monolayers on the treeing phenomena, the treeing growth rate curve for the epoxy/layered silicate (1.5 wt%) system was obtained from theAC treeing test at 10 kV with an electric field frequency of 60 Hz. The result are shown in Fig. 1. Electrical treeing was initiated in 12 min and propagated at the speed of 0.24 × 10^{-3} mm/min. The propagation speed until 1,140 min was 2.34 × 10^{-3} mm/min, and it was almost zero after that time without any propagation in the treeing growth. The propagation speed in the initial state was 9.7 times higher than the average speed until 11,790 min.Thiswas because as the treeing length increased with time, the distance between the needle-plate electrodes decreased. If the distance between the needle-plate electrodes became shorter, the electrical field value at a newly generated carbonized conductive tree tip increased. This can be explained by the Masons formula [15], $E_{tip} = 2V/(r \cdot \ln(1+4x/r))$, where E_{tip} is the applied electrical field at the needle tip, V is the applied voltage, r is the needle tip radius, and x is the distance between the needle-plate electrodes. When high voltage was applied to a needle tip, the electrical field value at the needle tip was far higher than the applied voltage. In this study, the values r = 5 μm, V = 10 kV, and x = 4,200 μm were introduced to the Masons formula, resulting in an E_{tip} of 492.6 kV/mm. The electrical field value at a newly generated carbonized conductive tree tip was much higher. Therefore, the tree length grew rapidly in the initial state. However, treeing growth was disturbed after 1,140 min. As explained in Fig. 2, the silicate monomers blocked the treeing growth, so the treeing had to develop a new root to propagate, which is a time-consuming process.

In order to confirm the barrier effect of the silicate monomers,

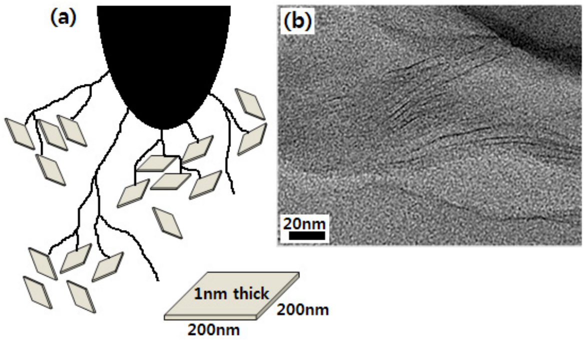

Fig. 2. (a) Barrier effect of silicate monolayers on the electrical treeing growth and (b) cross-sectional TEM image of the exfoliated silicate monolayers.

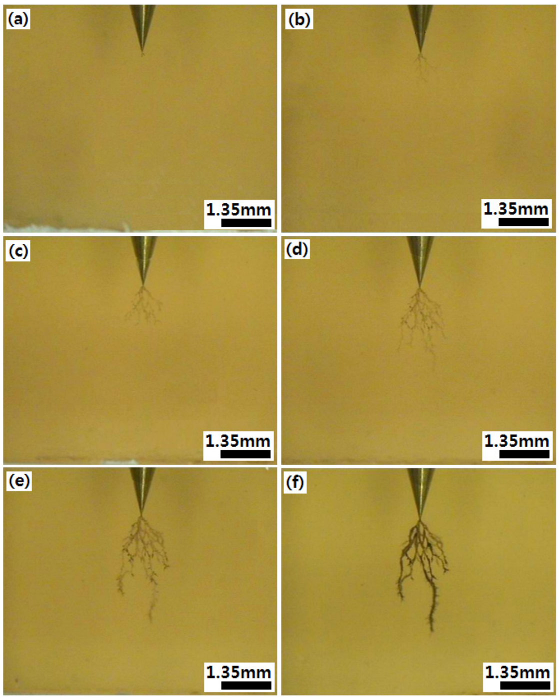

Fig. 3. Morphology of electrical tree corresponding to photos (a)-(f) was collected during HV (10 kV/4.2 mm, 60 Hz) applied at 130℃ for (a) 12 min, (b) 30 min, (c) 180 min, (d) 1,140 min, (e) 2,520 min, and (f) 11,790 min in the epoxy/layered silicate (1.5 wt%) system.

the treeing morphology was observed, and the electrical treeing morphologies corresponding to photos (a)~(f) in Fig. 1 were collected after applying 10 kV (60 Hz) for (a) 12 min, (b) 30 min, (c) 180 min, (d) 1,140 min, (e) 2,520 min, and (f) 11,790 min, and the results are displayed in Fig. 3. In the initial state, typical branch-type electrical treeing was observed in the morphology. Electrons injected and extracted at the needle tip initiated very small electrical treeing from the needle tip, as shown in Fig. 3(a), and then electrons injected and extracted at the newly generated conductive treeing tip were carbonized, so that several new branches appeared (Figs. 3(b)~(d)). The branches grow rapidly in the initial state. However, the treeing shape and the length were almost constant after 1,140 min (Fig. 3(e)), and the treeing became wider and darker without any new branches (Fig. 3(f)). These results meant that the treeing growth was disturbed by the dispersed silicate monolayers.

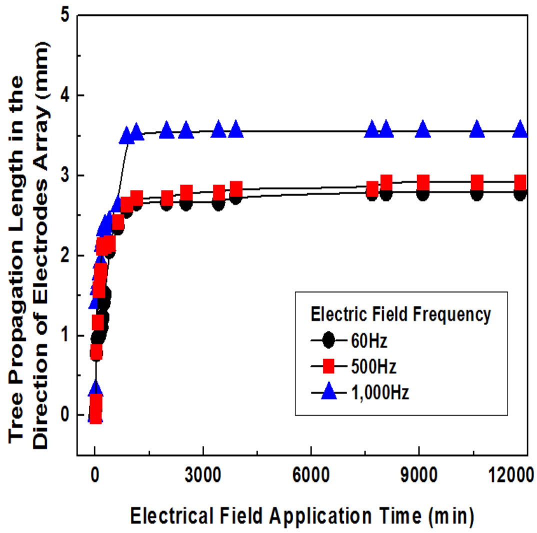

Fig. 4. Treeing growth rate in epoxy/layered silicate (1.5 wt%) system tested at 10 kV/4.2 mm with three different electric field frequencies(60, 500, and 1,000 Hz) at 130℃.

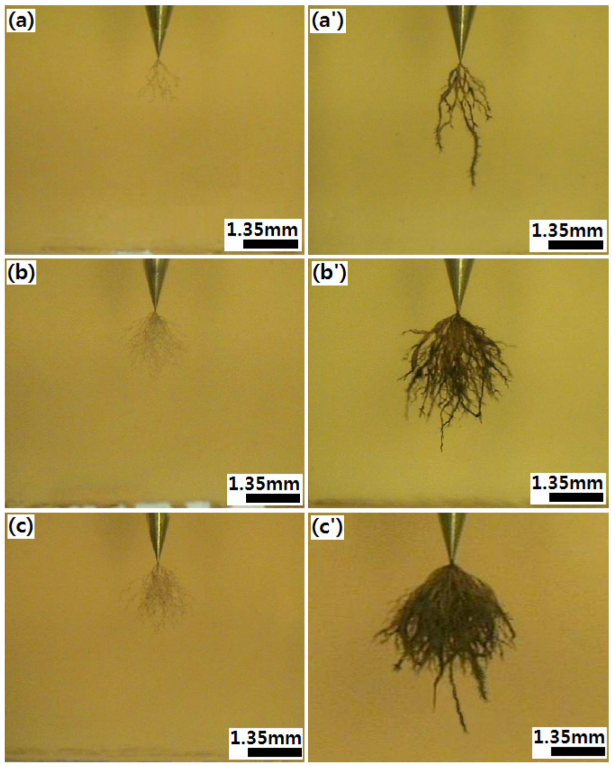

Fig. 5. Morphology of electrical treeing for the epoxy/layered silicate (1.5 wt%) nanocomposite tested in the constant electric field of 10 kV/4.2 mm with different electric field frequency: at 60 Hz for (a) 180 min and (a') 11,790 min; at 500 Hz for (b) 180 min and (b') 11,790 min; and at 1,000 Hz for (c) 180 min and (c') 11,790 min.

In order to study the effect of the electric field frequency on the AC treeing growth in the epoxy/layered silicate (1.5 wt%) nanocomposite, treeing growth curves at three different electric field frequencies are shown in Fig. 4. As the electric field frequency increased, the treeing initiation time decreased and the propagation rate increased. At 1,000 Hz, the treeing initiation time was 5 min, and the propagation rate was 0.30×10^{-3} mm/min. The propagation speed until 1,140 min was 3.09×10^{-3} mm/min, and it was almost zero after that time without any propagation in the treeing growth. The propagation speed in the initial state was 10.3 times higher than the average speed until 11,790 min. This was because as the treeing length increased with time, the distance between the needle-plate electrodes decreased, so that the electrical field strength at the treeing tip became higher.

The effect of the electric field frequency was also found by comparing the treeing growth speed. In the initial state, the speed at 1,000 Hz was 1.32 times higher than that at 60 Hz, and the average speed until 11,790 min at 1,000 Hz was 1.25 times higher than that at 60 Hz. This was due to the collision frequency of electrons at the needle tip or newly generated treeing tips.

As expected, the barrier effect of the silicate monolayers was also confirmed by observation of the treeing morphology, as shown in Fig. 5. At 500 Hz, bush-type treeing was generated in the initial state (Fig. 5(b)), and the treeing became dense, and the thickness became wide and dark (Fig. 5(b'). At 1,000 Hz, the bush-type treeing became denser, and the thickness became wider and darker (Fig. (c) and (c')). At 60 Hz, branch-type treeing was observed, as shown in Figs. 5(a) and (a'). A bush-type tree might occur be because the momentum power of the injected electrons could not pass through the silicate monolayers, so they should develop a new root to avoid the silicate monolayers. This is a time-consuming process. Therefore, it was found that well-dispersed silicate monolayers acted as good barriers to the treeing propagation.

4. CONCLUSIONS

The effects of electric field frequency on the AC electrical treeing phenomena in an epoxy/layered silicate (1.5 wt%) were investigated in a needle-plate electrode geometry. At 60 Hz, the treeing initiation time was 12 min, and the propagation speed until 1,140 min was 2.34×10^{-3} mm/min, and it was almost zero after that time without any propagation in the treeing growth. The propagation speed in the initial state was 9.7 times higher than the average speed until 11,790 min. As the electric field frequency increased, the treeing initiation time decreased and the propagation rate increased. In the initial state, the speed at 1,000 Hz was 1.32 times higher than that at 60 Hz, and the average speed until 11,790 min at 1,000 Hz was 1.25 times higher than that at 60 Hz. This was due to the collision frequency of electrons at the needle tip or newly generated treeing tip. The morphology was a dense branch type at 60 Hz, and the treeing morphology at 1,000 Hz was a dense bush type. It was found that well-dispersed silicate monolayers acted as good barriers to the treeing propagation.

ACKNOWLEDGMENT

This work was supported by Joongbu University (2013).

REFERENCES

[1] D. J. Suh and O. O. Park, J. Appl. Polym. Sci., **83**, 2143 (2002) [DOI: http://dx.doi.org/10.1002/app.10166].

[2] L. Zhang, Y. Z. Wang, Y. Q. Wang, Y. Sui and D. S. Yu, J. Appl. Polym. Sci., **78**, 1873 (2000) [DOI: http://dx.doi.org/10.1002/1097-4628(20001209)].

[3] K. Varlot, E. Reynaud, M. H. Kloppfer, G. Vigler and J. Varlet, J. Polym. Sci.: Part B, **39**, 1360 (2001) [DOI: http://dx.doi.org/10.1002/polb.1108].

[4] N. Artzi, Y. Nir, M. Narkis and A. Siegmann, J. Polym. Sci.: Part B, **40**, 1741 (2002) [DOI: http://dx.doi.org/10.1002/polb.10236].

[5] H. L. Tyan, K. H. Wei and T. E. Hsieh, J. Polym. Sci.: Part B, **38**, 2873 (2000) [DOI: http://dx.doi.org/10.1002/1099-0488(20001115)].

[6] J. Y. Lee, M. J. Shim and S. W. Kim, Polym. Eng. Sci., **39**, 1993 (1999) [DOI: http://dx.doi.org/10.1002/pen.11592].

[7] Y. S. Cho, M. J. Shim and S. W. Kim, Mater. Chem. Phys., **66**, 70 (2000) [DOI: http://dx.doi.org/10.1016/S0254-0584(00)00272-8].

[8] R. Sarathi, R. K. Sahu and P. Rajeshkumar, Mater. Sci. Eng.: A, **445**, 567 (2007) [DOI: http://dx.doi.org/10.1016/j.msea.2006.09.077].

[9] T. Tanaka, G. C. Montanari and R. Mülhaupt, IEEE Trans. Dielectr. Electr. Insul., **11**, 763 (2004) [DOI: http://dx.doi.org/10.1109/ TDEI.2004.1349782].

[10] T. Imai, F. Sawa, T. Yoshimitsu, T. Ozaki, and T. Shimizu, IEEE Annual Report Conference on CEIDP, p.239 (2004).

[11] T. Tanaka, IEEE Transactions on Dielectrics and Electrical Insulation, **9**, 704 (2002) [DOI: http://dx.doi.org/10.1109/TDEI.2002.1038658].

[12] R. Vogelsan, T. Farr, and K. Frohlich, IEEE Transactions on Dielectrics and Electrical Insulation, **13**, 373 (2006) [DOI: http://dx.doi.org/10.1109/TDEI.2006.1624282].

[13] L. A. Dissado, IEEE Transactions on Dielectrics and Electrical Insulation, **9**, 483 (2002) [DOI: http://dx.doi.org/10.1109/TDEI.2002.1024425].

[14] J. J. Park and J. Y. Lee, IEEE Trans. Dielectr. Electr. Insul. **17**, 1516 (2010) [DOI: http://dx.doi.org/10.1109/TDEI.2010.5595553].

[15] K. Theodosiou and I. Gialas, J. Electr. Eng., **59**, 248 (2008).