Dielectric Properties of Epoxy/Alumina Nanocomposite Influenced by Particle Dispersibility

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***Abstract*- For applications of nanocomposite materials to solid insulator for electric power apparatus, we have to consider various characteristics from mechanical, thermal and electrical points of view. In particular, the agglomerate of nanoparticles would decline the excellent properties of nanocomposite.**

In this paper, we investigated the influence of dispersibility of nanoparticles on electrical properties. Firstly, we fabricated epoxy/alumina nanocomposites with application of ultrasonic wave and centrifugal force. Secondly, we evaluated the dispersibility of nanoparticles by image analysis of agglomerate diameter from scanning electron microscopy (SEM) and the measurement of filler contents. Then, we examined the dielectric properties such as permittivity and dielectric loss of the nanocomposite. Finally, we verified the influence of the dispersibility of nanoparticles on the dielectric characteristics.

I. INTRODUCTION

For the enhancement of insulation reliability and compact design in electric power apparatus, the electrical properties of polymer composites with inorganic fillers in the polymer matrix play an important role. As new composite materials with significant features of insulation performances, functionally graded material (FGM) with grading distribution of filler density and the nanocomposite through the incorporation of nanopartlces have been reported [1-3]. Recently, it has been recognized that nanoparticle fillers have benefit for avoiding the degradation of insulation in long-term characteristics [3,4]. However, the agglomerate of nanoparticles can be more or less essential properties in nanocomposites and might reduce the excellent properties. Therefore, we have to make clear the influence of the dispersibility of nanoparticles on electrical and dielectric properties in nanocomposite materials.

This paper focuses on the influence of the dispersibility of alumina nanoparticles in epoxy resin on dielectric characteristics. Firstly, epoxy/alumina nanocomposites were fabricated with application of ultrasonic wave and centrifugal force. Secondly, the dispersibility of nanoparticles was evaluated by the image analysis of agglomerate diameter from SEM micrographs and the measurement of filler contents. We made sure that the dispersibility of nanoparticles changed with the application condition of ultrasonic wave and centrifugal force. Then, we examined the dielectric permittivity and dielectric loss among various dispersibilities of nanoparticles. Finally, we could verify the difference of dielectric characteristics among dispersibilities of nanoparticles.

II. FABRICATION OF NANOCOMPOSITE

We fabricated the nanocomposite material from epoxy resin with alumina nanoparticles. The epoxy resin was based on bisphenol-A cured with anhydride-type hardener. The alumina nanoparticles are of spherical shape with an average diameter of 31nm. The filler content was 0.5vol%. As coupling agent, 1wt% silane coupling was added in epoxy resin.

For improving the dispersersibility of the nanoparticles in epoxy resin, we asynchronously applied the ultrasonic wave and centrifugal force. Table I shows the application conditions of the ultrasonic wave and the centrifugal force. Furthermore, we made experiments by two procedures of application of the ultrasonic wave and the centrifugal force. The first one is "Proc.1"; we applied the ultrasonic wave and then applied the centrifugal force. The other one is "Proc.2"; we applied the ultrasonic wave, centrifugal force and then applied the ultrasonic wave again.

TABLE I
PROCESS OF IMPROVING DISPERSIBILITY FOR NANOPARTICLE

Process	Characteristics	Duration time [min]
Application of ultrasonic wave	Amplitude : 13.0μm	30
Application of centrifugal force	Centrifugal force : 200 - 20,000G	60

The fabrication process of the nanocomposite specimens consisted of the following steps.

1) Epoxy resin is mixed with hardeners, silane coupling and fillers by a planetary mixer.
2) The ultrasonic wave and the centrifugal force are applied to the mixture. The application conditions are varied as shown in Table II. After application of the centrifugal force, the precipitation is removed from the mixture.
3) The mixture is poured into casts, and is degassed enough for removal of bubbles.
4) The specimen is cured and postcured.

The specimens were sliced as discs with a thickness of 1.0mm for measurement of filler contents and dielectric properties.

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TABLE II
SPECIFICATIONS OF NANOCOMSPOSITE SPESIMENS

Specimen	Ultrasonic wave	Centrifugal force	Application procedure
N_MM	–	1G (gravitation)	–
N_USW	○	1G (gravitation)	–
N_CF1	–	200G	–
N_CF2	–	2,000G	–
N_CF3	–	20,000G	–
N_CB1	○	20,000G	Proc.1
N_CB2	○	20,000G	Proc.2

III. EXPERIMENTAL PROCEDURE

A. Image analysis of agglomerate diameter

The dispersibility of nanoparticles was observed by scanning electron microscopy (SEM). We focused on the diameter of agglomerate as a typical parameter of the dispersibility of nanoparticles. Therefore, we investigated the maximum diameter of agglomerate cross-section on the SEM micrographs by image process and image analysis which was assumed to be the same as actual diameter of agglomerate.

B. Measurement of filler contents

Even if the precipitation was removed after application of the centrifugal force, the filler contents of the specimens were obtained by measurement of the specific gravity of the specimens. We measured the specific gravity of the specimens.

C. Measurement of dielectric properties

Relative permittivity and dielectric loss tangent (tanδ) were estimated from measurement of capacitance of the specimens at the frequency of 1MHz under 30˚C.

IV. RESULTS AND DISCUSSIONS

A. Dispersibility of nanoparticles

Figure 1 shows the example of SEM micrographs of the fabricated specimens. In the micrographs, white colors show alumina particle and black colors show epoxy resin. In the micrographs at the magnification of 1,000, we found that agglomerates were well removed by application of the centrifugal force. Though we can see several small agglomerates less than 1μm size by observation of the micrographs at the magnification of 10,000, the nanoparticles were well dispersed.

Figure 2 shows the centrifugal force characteristics of maximum diameter of the agglomerates. In this figure, the largest 10 data were sequentially plotted from the observed micrographs for each specimen. With the increase of the centrifugal force, the size reduction of agglomerates can be seen. Especially at 20,000G, the diameter of the agglomerates is 500–900nm. Furthermore, the movement of an agglomerate in viscosity liquid at 20˚C under the centrifugal force was calculated with the assumption that the shape of the agglomerate was spherical as the following equation.

$$D_p^{\,2} = 0.18\eta_c R_1 \ln(R_1/R_0)/(F\pi^2 t(\rho_p - \rho_f)) \qquad (1)$$

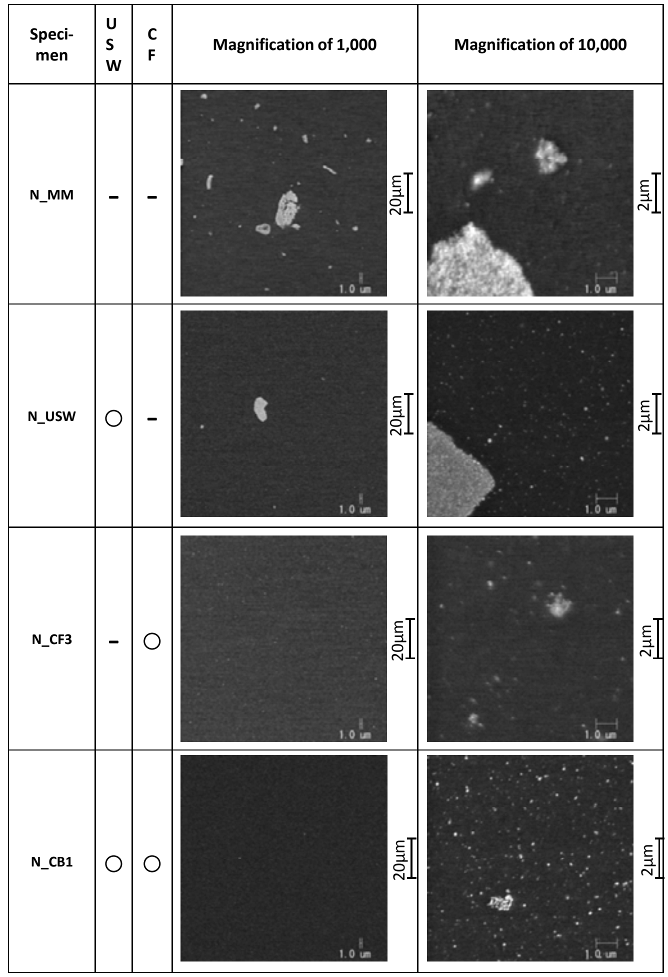

Fig. 1. Scanning electron microscopy (SEM) micrographs of epoxy/alumina nanocomposite with the ultrasonic wave (USW) and centrigfugal force (CF)

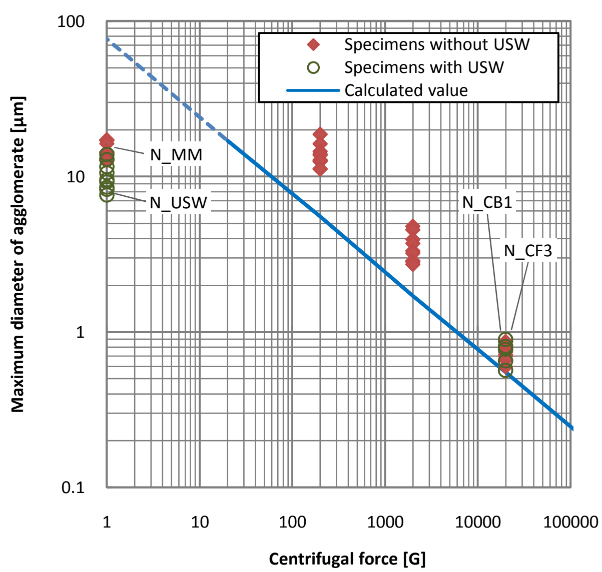

Fig. 2. Centrifugal force characteristics of maximum agglomerate diameter with measument results of specimens and calculated value by particle motion equation

Here, D_p is a particle diameter, η_c is a viscosity, R_1 and R_0 are outer and inner radius of centrifuging respectively, F is a centrifugal force, t is a duration time, ρ_p and ρ_f are a specific gravity of the particle and polymer matrix, respectively. In Figure 2, the measurement results of agglomerate diameter of specimens are close to the calculated value. This result confirms the centrifugal force application can well remove the agglomerates with micrometric diameter. On the other hand, the agglomerate diameter of the specimens at 1G (gravitation) is significant smaller than the calculated value. The result may be explained by the fact that the agglomerate with more than 20μm diameter can precipitate in curing at high temperature.

Figure 3 shows the measurement results of the maximum diameter of agglomerates as the effective procedure by the ultrasonic wave and the centrifugal force. From this figure, the maximum diameter of agglomerates can be reduced to 300-600nm by the application of "Proc.2".

Figure 4 compares the filler contents for all of the tested specimens. The filler contents of specimens without the ultrasonic wave (0.16-0.32vol%) is lower than the entire filler content (0.5vol%) and decreased with the increase of the centrifugal force. The results show that a lot of agglomerates with micrometric diameter can precipitate by the gravitation and the centrifugal force and the precipitation can be removed from the specimens in the fabrication process. On the other hand, the filler contents of specimens with the ultrasonic wave (0.46-0.47vol%) are larger than that without the ultrasonic wave (0.16-0.32vol%) and almost independent with the increase of the centrifugal force. The results show the number of the agglomerate with micrometric diameter can be reduced by application of the ultrasonic wave. Furthermore, specimens of N_USW, N_CB1 and N_CB2 have almost the same filler content but the different diameter of agglomerate as shown in figure 3. Therefore, we obtained the nanocomposite specimens with different dispersibilities with keeping the filler content.

From the viewpoint of particle distribution, we can illustrate the Figure 5 for qualitative explanation of the reducing effect of agglomerate diameter and number by the ultrasonic wave and the centrifugal force application. From this figure, it is understood that the application of the ultrasonic wave can move the particle distribution to smaller diameter, i.e. improvement of dispersibility, thus filler content with the ultrasonic wave does not change even if the centrifugal force is applied.

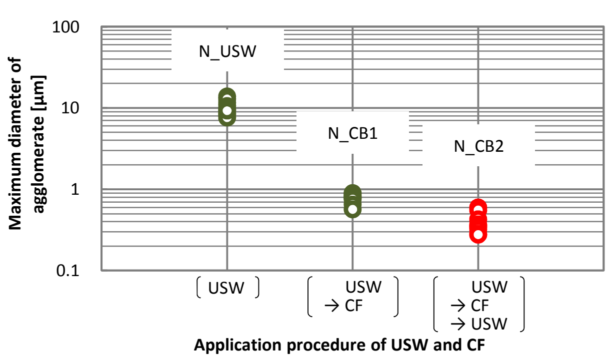

Fig. 3. Maximum diameter of agglomerate for application procedure of ultrasonic wave (USW) and centrifugal force (CF)

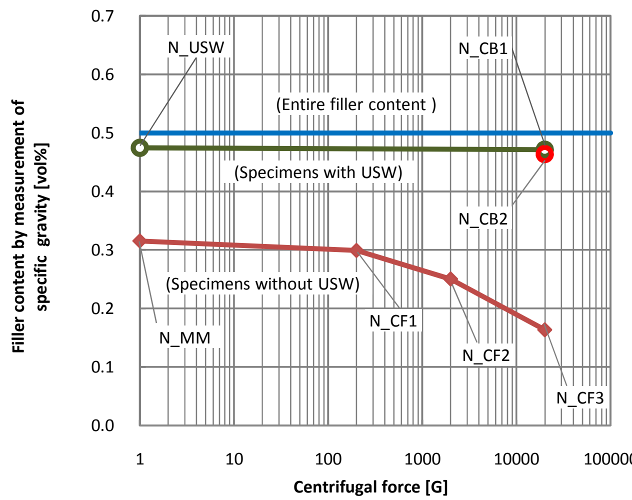

Fig. 4. Centrifugal force characteristics of filler contents for epoxy/alumina nanocomposite

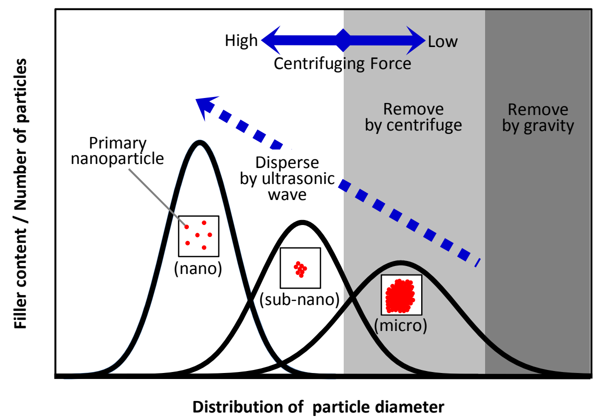

Fig. 5. Illustrations of improving dispersibility by ultrasonic wave and centrifugal force

B. *Dielectric properties*

Figures 6 and 7 show the centrifugal force characteristics of relative permittivity and tanδ. The relative permittivity of specimens without the ultrasonic wave is decreased with the increase of the centrifugal force. This result can be explained by the fact that the filler contents of specimens without ultrasonic wave are decreased with the increase of the centrifugal force as shown in figure 4. It is presumed that the relative permittivity characteristics of the micrometric agglomerate of alumina nanoparticles are close to alumina micrometric particle. On the other hand, the relative permittivity of N_USW is smaller than N_MM even if the filler content of N_USW is larger than N_MM as shown in figure 4. This result shows that the permittivity of nanocomposite in 0.5 vol% filler content at 30℃ with 1MHz can be made lower by improving the dispersibility of nanoparticles.

Figures 8 and 9 show relative permittivity and tanδ as a function of the maximum diameter of agglomerate. This results show almost the same dielectric characteristics among the different maximum diameter of agglomerate in the same filler content.

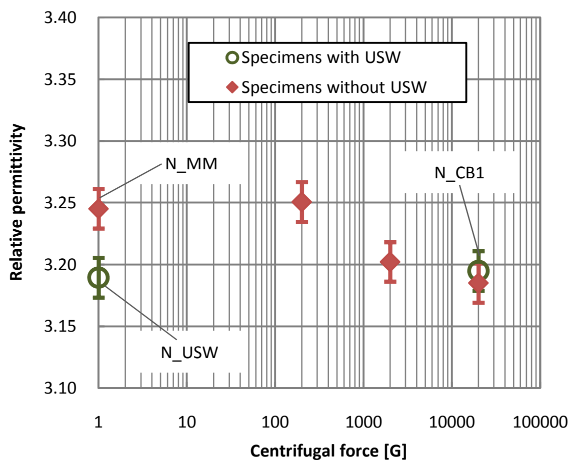

Fig. 6. Centrifugal force characteristics of relative permittivity at 30℃ with 1MHz for epoxy/alumina nanocomposite

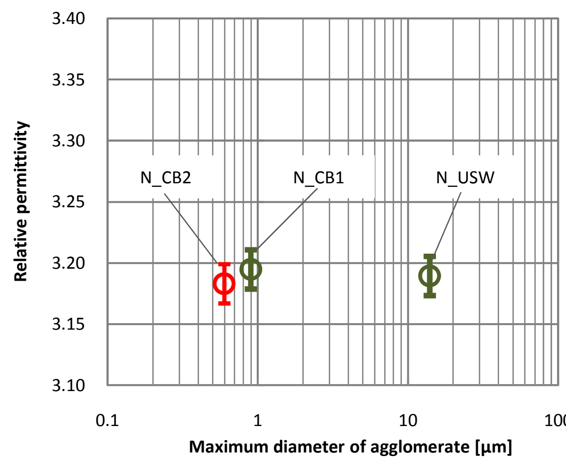

Fig. 8. Relative permittivity as a function of the maximum diameter of agglomerate at 30℃ with 1MHz (filler content: 0.46-0.47vol%)

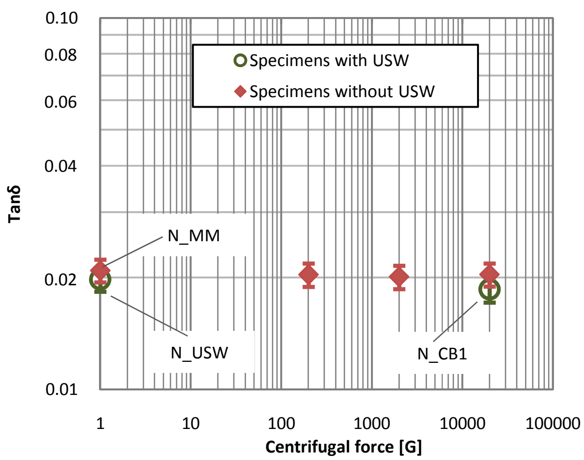

Fig. 7. Centrifugal force characteristics of tan δ at 30℃ with 1MHz for epoxy/alumina nanocomposite

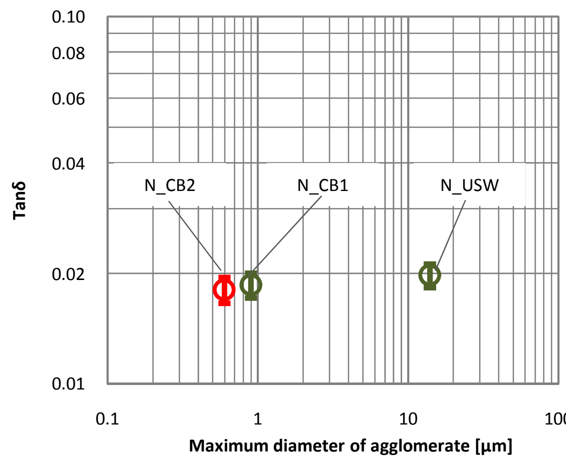

Fig. 9. Tan δ as a function of the maximum diameter of agglomerate at 30℃ with 1MHz (filler content: 0.46-0.47vol%)

V. CONCLUSIONS

In order to make clear the influence of the dispersibility of nanoparticles on electrical property, we investigated the dispersibility and the dielectric properties of alumina nanoparticles in epoxy resin on dielectric properties. Firstly, we fabricated epoxy/alumina nanocomposite and evaluated the agglomerate diameter by an image processing and an image analysis of SEM micrographs and measured filler contents of specimens. Next, we examined the relative permittivity and tanδ in the different application procedures of the ultrasonic wave and the centrifugal force. The main results are summarized as follows.

(1) For improvement of dispersibility of nanoparticles in the fabrication process by direct mixing, the applications of the ultrasonic wave and the centrifugal force were effective.

(2) We found the application of both procedure of the ultrasonic wave and the centrifugal force can reduce the maximum diameter of agglomerate to 300-600nm.

(3) Relative permittivity of nanocomposite at 30℃ with 1MHz in 0.5 vol% filler content could be made lower by improving the dispersibility of nanoparticles.

(4) Dielectric characteristics among the different maximum diameter of agglomerate in the same filler content were almost the same if the nanoparticles were well dispersed by the ultrasonic wave.

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