



Article Inverse Proportionality of Thermal Conductivity and Complex Permittivity to Filler-Diameter in Epoxy Resin Composites with Silica

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Abstract: We prepared six kinds of epoxy resin nanocomposites with silica and an epoxy resin with no silica. The nanocomposites contain silica with different diameters (10, 50, and 100 nm) while their silica contents are 1, 5, 10, and 20 vol%. At 25 and 100 °C, the thermal conductivity has a nearly proportional dependence on the silica content and exhibits an almost reciprocal proportionality to the diameter of the silica. The latter result indicates that the interaction at filler-resin interfaces plays a significant role in heat transfer. However, this view contradicts an easy-to-understand thought that the filler-resin interfaces should work as a barrier for heat transfer. This in turn indicates that the interaction at filler-resin interfaces controls the bulk properties of the resin when the filler is in a nm size. Although the dielectric constant increases with the addition of the silica filler, its increment from the resin with no silica is the smallest in the resin with the 10-nm silica. Therefore, the addition of the 10-nm silica is adequate for electrical insulation purposes.

Keywords: polymeric insulation; machine insulation; heat conduction; filler-resin interface; interaction

1. Introduction

Organic polymers, into which nanofillers (inorganic in most cases and metallic in some cases [1,2]) are dispersed uniformly, are called polymer nanocomposites (NCs) [1–18]. They have been attracting much attention in various fields recently since their good potential abilities have been reported. In Japan, two research projects are currently being funded by a public organization for the applications of NCs for the electrical insulation of power apparatuses. More specifically, the common goal of the two projects is to develop innovative electrical insulations that can reduce the insulation thickness of the coil windings used in power generators. One of the two projects is for generators up to the 1.5-kVA class [17,18], while the other is for larger generators [13–16]. For contributing to the project for larger generators, we have been investigating the best nanofiller to add to the epoxy resin. Up to the present, NCs of epoxy resin with silica (SiO₂), rutile (TiO₂), magnesia (MgO), or magnesium hydroxide (Mg(OH)₂), added as a nanofiller, were selected as candidate materials. Therefore, various characteristics important for electrical insulation, such as impulse breakdown strength [13], insulation lifetime [14], electroluminescence [15], complex permittivity, and elasticity [16], have been investigated in such NCs.

To our knowledge, several NCs have already been applied in the industry. As far as we know, the applications of NCs have been realized fast in the electrical insulation of magnet wires and power cables in comparison to many other pieces of apparatuses. Examples of such industrial applications realized in several pieces of power apparatus were reviewed, together with general aspects of NCs (in the books where [1,2] appear and reference [4]). One of the authors of the present paper (YO) also reported the details of



Citation: Ohki, Y.; Hirai, N.; Umemoto, T.; Muto, H. Inverse Proportionality of Thermal Conductivity and Complex Permittivity to Filler-Diameter in Epoxy Resin Composites with Silica. *J. Compos. Sci.* **2021**, *5*, 266. https:// doi.org/10.3390/jcs5100266

Academic Editor: Francesco Tornabene

Received: 16 September 2021 Accepted: 4 October 2021 Published: 11 October 2021

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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). the applications of NCs to solid-insulated DC power cables and coil windings [19–23]. In addition, a solid-state switchgear insulated with nano-microcomposite, the development of which was reported in [24], has already been installed widely in the world market [25].

For industrial applications of polymer NCs as electrical insulating materials, not only their electrical insulating properties but also their thermal properties are important. From this view, in this research their thermal conductivity was measured in six kinds of epoxy resin NCs with silica as well as epoxy resin with no silica. Our results show that their thermal conductivity depends almost reciprocally on the diameter of the silica filler. This result is strange if we assume that the interfaces between the resin and the filler should prevent the heat from being transferred in each direction. Instead, this result indicates that the resin/filler interfaces exert significant effects on various properties of the bulk resin.

2. Materials and Methods

The host resin used in this experiment is bisphenol F epoxy resin. Bisphenol F has a low viscosity and less often crystallizes than bisphenol A and, thus, is suitable for our project. The curing agent used is acid anhydride. The nanofiller we added is almost spherical amorphous silica with various diameters (listed in Table 1).

No.	Symbol	Diameter (nm)	Content (vol%)
1	•	Neat	
2		100	10
3	٠	50	10
4	۲	10	10
5	٠	100	1
6	A	100	5
7	▼	100	20

Table 1. Numbers and symbols of NC samples along with the diameters and contents of silica fillers.

The color symbols do not apply to Figures 4 and 5.

No surface treatments were given to the silica. The silica was added to the epoxy resin with a content of 1, 5, 10, or 20 vol% by high-pressure shearing using a high-pressure wet-type pulverization machine called Nanovater (Yoshida Kikai). A high shearing force emerges and cavitation is consequently induced when the resin with fillers passes through a nozzle of the machine. This force and cavitation assist the pulverization of aggregates of fillers and their uniform dispersion. Then, each resin with the filler was cured at 120 °C for 2 h in air and then cured secondly at 180 °C for 4 h. Finally, sheets of NC samples, 0.5–0.2 mm thick, were prepared.

Figure 1 shows energy-dispersive X-ray (EDX) images taken on fracture surfaces of samples No. 5 and No. 6 using a scanning electron microscope (SEM). The color bars beside the images represent the intensities of the characteristic X-rays that correspond to the abundance of Si atoms. Here, the black and the white bars mean the lowest and highest intensity levels, respectively, while the green and red ones, if they are present, show the intermediates.

Figure 1 indicates that Si atoms of silica are distributed uniformly inside the two samples. We confirmed similar EDX images for the other samples. In addition, although not shown here, we observed all the samples' SEM images, which are probably more popularly used in general to check the dispersion of fillers in NCs. Therefore, we believe that the silica nanofillers were dispersed uniformly in all the NC samples.

The thermal conductivity λ in W m⁻¹ K⁻¹ was measured at 25 and 100 °C twice for each NC sample. Here, λ is the product of the density ρ in kg m⁻³, the specific heat C_p in J kg⁻¹ K⁻¹, and thermal diffusivity α in m² s⁻¹. In this regard, ρ was measured by the water displacement (Archimedes') method using an electronic balance (Shimadzu AUW120D). Since the changes in ρ and thickness induced by the sample's thermal expansion are negligible, ρ was measured only at 25 °C. In addition, C_p was measured in an N₂ atmosphere at 25 and 100 °C by dynamic scanning calorimetry with a PerkinElmer DSC8500 spectrometer using α -Al₂O₃ single crystal (sapphire) as a standard substance. Furthermore, α was measured in an Ar atmosphere at 25 and 100 °C by the flash method. For the flash method, a light flash apparatus (LFA 467 HT HyperFlash, Netzsch) with a xenon flash tube and an InSb infrared sensor was used. Before the measurement of α , graphite was sprayed on the sample surface to make it black.



Figure 1. SEM-EDX images taken on fracture surfaces of samples No. 5 and No. 6. (**a**) Sample No. 5. (**b**) Sample No. 6.

We also measured the complex permittivity of the NCs in vacuum at frequencies from 10^{-2} to 10^5 Hz by applying ac 3 Vrms using an impedance analyzer (SI126096, Solartron) in a temperature range from 20 to 200 °C.

3. Results and Discussion

Using the values measured at 25 and 100 °C for samples 1, 2, 5, 6, and 7 or the NCs with silica fillers with the average diameter of 100 nm, the relation between the thermal conductivity and the volume content of fillers is shown in Figure 2.



Figure 2. Thermal conductivity as a function of filler content, measured in epoxy NC samples with 100-nm silica. For the sample symbols, refer to Table 1. Several data overlap each other. (**a**) 25 °C, (**b**) 100 °C.

As is clear from Figure 2, the thermal conductivity increases with the increase in the content of fillers, showing a positive correlation with one another. Although in the literature

the value of the thermal conductivity of silica ranges from 1.4 to 20 W m⁻¹ K⁻¹ [26–28], it is much higher than the value of the epoxy resin sample 1 shown on the ordinate of Figure 2a or Figure 2b. Regarding this, the above result is quite understandable. Furthermore, as shown by the curves in Figure 2, the thermal conductivity of the NC samples can be well approximated by the following Bruggeman equation [29,30] based on the effective medium approximation theory:

$$\frac{\lambda_r - \lambda_c}{\lambda_r + 2\lambda_c} (1 - f) + \frac{\lambda_f - \lambda_c}{\lambda_f + 2\lambda_c} f = 0.$$
(1)

Here, λ_r , λ_f , and λ_c are the thermal conductivity of the base material, the filler, and the composite, and f is the volume fraction of the filler. In the calculations, since the thermal conductivity of silica at 100 °C is unknown, 2.0 W m⁻¹ K⁻¹ [27], reported as the value at room temperature, was used at both 25 and 100 °C.

Next, using the measured values of samples 1, 2, 3, and 4, or the NC samples with silica at the same content of 10 vol%, the thermal conductivity is shown in Figure 3 as a function of reciprocal filler diameter. Here, in Figure 3, the values of thermal conductivity are normalized by the averages of the values measured two times for the epoxy resin sample with no silica fillers, namely 0.16 and 0.18 W m⁻¹ K⁻¹ at 25 and 100 °C, respectively.



Figure 3. Thermal conductivity as a function of reciprocal filler diameter, measured in epoxy NC samples containing 10-vol% silica. For the sample symbols, refer to Table 1. Several data overlap each other. (a) 25 $^{\circ}$ C, (b) 100 $^{\circ}$ C.

As mentioned above, we have been carrying out this research as part of a project, and one of the goals of the project is to improve thermal conductivity 1.2-fold by replacing the epoxy resin with its NC. The horizontal lines drawn in Figure 3 are meant to guide the eyes. As shown in Figure 3, when we added the silica filler by 10 vol%, we can achieve the target if the filler diameter is 10 or 50 nm at 25 and 100 °C. However, the filler diameter of 100 nm has almost no margin. Taking Figure 2 into account, for the silica with the diameter of 100 nm the filler content of 5 vol% or less cannot exceed the target value. From Figures 2 and 3, the boundary for silica seems to be a diameter of 100 nm and a filler content of 10 vol%.

As shown in Figure 3, the thermal conductivity correlates almost linearly and positively with the reciprocal filler diameter. When the same volume of fillers is added to a polymer resin, the total area of the filler surfaces or that of the filler-resin interfaces increases as the filler size decreases. The ratio of the total surface area of the fillers to their unit volume is called the specific surface area. If we assume simply that all the fillers are spherical and with the same diameter, their specific surface area is inversely proportional to the filler diameter. In this regard, the thermal conductivity increases proportionally to the total area of the interfaces between the filler and the resin, regardless of whether the measurement temperature is 25 or 100 $^{\circ}$ C.

Concerning the above, the large specific surface area of small-sized fillers (in other words, some interaction at the large filler-resin interfaces) should influence various proper-

ties of the polymer bulk. Many researchers would have had such an idea that the above is the cause of the excellent insulating properties of polymer NCs from the early days of research on polymer NCs. As one of the models assuming the interaction at the filler-resin interfaces, T. Tanaka proposed a multi-core model [31], together with one of the authors (YO) of this paper. This model has widely been accepted worldwide, partly due to its easy-to-understand and easy-to-quote naming. However, it would be true that neither a clear explanation of the actual interaction at the filler-resin interfaces nor its direct evidence has been given.

As mentioned above, the filler size should be reduced if we want to increase the thermal conductivity of the NCs. If we assume the situation that the heat generated inside the fillers is transferred to the resin through the filler-resin interfaces, the heat is transferred more easily if the interface area becomes larger, making the thermal conductivity of the NC higher. However, the situation discussed here is quite different. If the thermal motion of molecules and atoms is transmitted to surrounding molecules and atoms, heat is transferred. This is the principal mechanism of heat conduction, although heat in metals is also transferred by free electrons. In an insulator, of course, heat conduction by free electrons can be ignored, and heat is transmitted in a substance, taking the form of the aforementioned lattice vibrations or phonons. If we use a slightly rough analogy, the filler-resin interface is assumed to work as a resistance to the transmission of phonons, just as in the case where sound is reflected at the interface of substances with different acoustic impedances. This common-sense approach is the opposite of the results shown in Figure 3, wherein the larger filler-resin interfacial area brings about a higher thermal conductivity when the filler content is the same. Regarding heat conduction, the nanofillers seem to behave out of the ordinary, and their interfaces with the resin act as layers assisting the heat transfer by phonons.

Such out-of-the-ordinary behavior seems to be indirect evidence of the concept that some interaction at the large filler-resin interfaces is the cause of the excellent insulating properties of polymer NCs. In other words, some interaction at the large filler-resin interfaces in a NC really alters various properties of the bulk of resin significantly. We have published several papers [8,16,32–36], which describe examples of experimental results that indicate variations of bulk properties induced by the addition of nanofillers to polymeric insulating materials.

Figure 2 shows that the thermal conductivity becomes higher at 100 °C than at 25 °C. In general, the thermal conductivity of nonmetals is insensitive to the temperature compared to metals [37]. Although we cannot determine how the thermal conductivity of bisphenol F epoxy resin changes as a function of temperature at room temperature and above, it is reported that the thermal conductivity of a kind of polyethylene increases slightly with an increase in temperature around room temperature [38]. Therefore, there is a possibility that a similar mechanism to the above polyethylene occurs. However, the thermal activation of the filler-resin interaction we are proposing or its wider expansion to the bulk of the epoxy resin seems possible as another mechanism.

Of course, complex relative permittivity or its real part (i.e., relative dielectric constant ε_r') and its imaginary part (i.e., dielectric loss factor ε_r'') are important for the goals of our project. We measured ε_r' and ε_r'' in the above-mentioned wide frequency and temperature ranges since broadband frequency data of complex permittivity at different temperatures contain a lot of important information concerning the dielectric behavior of solid dielectrics. We have conducted, in our previous paper [16], such analysis on dielectric behavior of various NCs similar to the present ones, including ones with silica. Therefore, we deal with only the data of ε_r' and ε_r'' measured under the industrially important 50 Hz at 20, 100, and 140 °C in this paper. Figure 4 shows ε_r' and ε_r'' as a function of filler content, measured in the epoxy resin sample 1 and the NC samples 2, 5, 6, and 7 with 100-nm silica.

At 20 °C, ε_r' becomes higher monotonically with the increase in the content of silica filler as shown in Figure 4a. This result reflects simply that ε_r' of silica is 4.41–4.60 at room temperature [39], which is higher than ε_r' of sample 1 or the pristine epoxy

resin. However, ε_r' does not exhibit such monotonic filler-content dependence at 100 and 140 °C. The dominant charge carrier species in epoxy resin is ions, especially at high temperatures [16,40,41]. In this regard, the value of ε_r' receives significant influences from the accumulation of hetero space charge in front of electrodes. This phenomenon is often called electrode polarization [10]. Various complicated bulk processes such as ionization, recombination, and transport of charge carriers, as well as various electrode processes such as charge exchange, are involved in the electrode polarization. These phenomena seem to be the cause of the complicated dependence of ε_r' on the filler content at 100 and 140 °C.



Figure 4. $\varepsilon_r'(\mathbf{a})$ and $\varepsilon_r''(\mathbf{b})$ as a function of filler content, measured in epoxy NC samples with 100-nm silica at 20 (**I**), 100 (**•**), and 140 (**•**) °C.

As shown in Figure 4b, ε_r " is always lower in the four NC samples than in the epoxy resin sample if we ignore two slight exceptions. If the transport of charge carriers occurs, the Joule heat emerges, which contributes to ε_r ". Therefore, the decrease in ε_r " is very often caused by the suppression of the charge transport [16]. A phenomenon similar to the present one, namely the suppression of the charge transport by the addition of silica nanofiller to epoxy resin, was also confirmed in our previous paper [40].

Figure 5 shows ε_r' and ε_r'' as a function of reciprocal filler diameter, measured in the epoxy resin sample 1 and the NC samples 2, 3, and 4 with the filler content of 10 vol%. Neither ε_r' nor ε_r'' shows simple dependence on the reciprocal filler diameter. There is a possibility that the interaction at filler-resin interfaces exerts different influences on the two physical parameters, namely the thermal conductivity and the complex permittivity ε_r' and ε_r'' .



Figure 5. ε_r' (**a**) and ε_r'' (**b**) as a function of reciprocal filler diameter, measured in the epoxy resin sample and the NC samples with the filler content of 10 vol% at 20 (**I**), 100 (**•**), and 140 (**A**) °C. Several data overlap each other.

As mentioned above, the objective of our research is to improve the insulation ability of solid insulation in coil windings in power generators. For this objective, the occurrence of partial discharges in air or gaseous insulation around the solid insulation must be prevented as much as possible. In that sense, it is desirable to lower ε_r' of the solid insulation since the electric field intensity in air or gaseous insulation around the solid insulation is enhanced if ε_r' of the solid insulation becomes high. When we watch Figure 5a while taking the above view into account, the silica with the smallest diameter of 10 nm is the most adequate for the present epoxy resin insulation. As for ε_r'' , a low value is also desirable since it is a parameter governing the dielectric loss. As shown in Figure 5b, the values of ε_r'' of the NC sample with the 10-nm-diameter silica are below those of the epoxy sample with no filler and acceptable at all temperatures. Therefore, making NC insulation layers using silica with diameters of around 10 nm would be a good method to achieve our goal.

Lastly, we forecast the economic and environmental aspects of this industrial application of NCs. It might be true that the use of nanofiller increases the price of a new power generator slightly if we compare it unilaterally to that of conventional ones. However, the reduction of the coil insulation thickness helps the cooling performance of coils, which in turn contributes to the miniaturization of the products and environmental burdens. According to the plan we submitted to the funding organizer, we will utilize this technology in manufacturing new generators in several years.

4. Conclusions

The effects of the addition of silica nanofillers to epoxy resin on their thermal conductivity and complex permittivity were examined, aiming at developing a new insulating material adequate for solid insulation around coil windings in power generators. Our results show that their thermal conductivity increases with the increase in the content of silica in a way that can be approximated by Bruggeman's equation. As for the relationship with the nanofiller size, the thermal conductivity correlates almost linearly with the reciprocal filler diameter (that is, the thermal conductivity becomes higher as the filler becomes smaller). This finding runs counter to the notion that the filler-resin interfaces are barriers for heat transfer. Instead, the finding indicates that the effects of the addition of nanofillers reach broad areas in the bulk of resin and change various bulk properties such as thermal conduction. Although the direct mechanism is unknown, some filler-resin interactions, expanding widely to the bulk of the epoxy resin, favorably affects the heat transfer in the resin when the filler is in the nm size.

The NC sample with the smallest-diameter silica has the lowest dielectric constant. Its dielectric loss factor is also low. The electric field strength in a void or air layer around a solid substance is enhanced if the dielectric constant of the substance is high. In this regard, a low dielectric constant is favorable for a filler to prevent the occurrence of partial discharges in the case of the appearance of voids. A low dielectric loss factor is also beneficial since we can expect a low power loss. Therefore, silica with a small diameter can be a good candidate as a filler to add to epoxy resin for achieving good electrical insulation from both the thermal and dielectric viewpoints.

Author Contributions: Conceptualization, Y.O. and H.M.; methodology, N.H. and T.U.; software, N.H.; validation, N.H. and Y.O.; formal analysis, Y.O. and N.H.; investigation, N.H.; resources, T.U.; data curation, N.H.; writing—original draft preparation, N.H. and Y.O.; writing—review and editing, Y.O.; visualization, N.H.; supervision, Y.O. and H.M.; project administration, N.H. and T.U.; funding acquisition, Y.O. and H.M. All authors have read and agreed to the published version of the manuscript.

Funding: This work was partly supported by a project, Innovation Program for Energy Conservation Technologies (JPNP12004), of the New Energy and Industrial Technology Development Organization (NEDO).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: Karen Kawada and Tomoki Tsujino, former students supervised by Y.O., conducted parts of the experiments. The authors acknowledge them in Acknowledgments. Several other data were taken by an outsourced company. The authors would like to thank Karen Kawada and Tomoki Tsujino for sharing their experimental results.

Conflicts of Interest: The authors declare no conflict of interest.

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