

Physical Factors Determining Thermal Conductivities of Isotropic Conductive Adhesives

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The thermal conductivities of a typical isotropic conductive adhesive (ICA) composed of an epoxy-based binder containing flake-shaped and spherical Ag particles have been studied. The ICA specimens exhibit anisotropy in their thermal conductivities between the vertical and in-plane directions. In addition, their thermal conductivities apparently depend on their thermal history during the curing and subsequent annealing processes, even if they are fully cured during the preliminary curing stage. The thermal conductivities tend to increase with decreasing electrical resistivities. The experimental values for the thermal conductivities of the ICA specimens were apparently higher than those predicted using Bruggemann's equation. Analysis using the Wiedemann–Franz (W–F) law indicates that their thermal conductivities increase with increasing contribution from electrical conduction. In order to discuss the thermal conductivities of ICA specimens that exhibit relatively low electrical resistivities (below $10^{-3} \Omega \text{ cm}$), the contribution of the conducting electrons must be included.

Key words: Electrically conductive adhesives, thermal conductivity, curing, annealing, percolation

INTRODUCTION

Electrically conductive adhesives, including isotropic and anisotropic conductive adhesives (ICAs), are expected to become one of the prime candidates for interconnecting materials in novel electronics packaging systems.^{1–3} In order to expand the practical applications of these adhesives, their interconnect properties and reliability under severe conditions need to be clarified.^{4,5} Although various properties of the adhesives need to be evaluated in order to study interconnect reliability, these properties have not always been understood accurately.

The present authors have investigated the effects of curing conditions on the electrical properties of ICAs composed of an epoxy-based binder.^{6,7} As a

result, the onset of electrical conductivity in these adhesives (which exists due to percolation networks formed by the filler particles) has been clarified as originating during the curing and cooling processes, which are accompanied by shrinkage of the adhesive binder. Furthermore, the curing temperature has an influence on the electrical resistivity of the adhesives. The resistivity decreases with increasing curing temperature, even when the adhesives exhibit similar degrees of conversion. In order to discuss the electrical properties of the adhesives, the effect of process conditions should be considered, as well as other material factors.

The present work investigates the thermal conductivity of a typical ICA. Thermal conductivity is a key property in the thermal management of electronic products; for example, the thermal conductivity of ICAs becomes important when they are

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employed in the die-attach applications. The present authors have previously analyzed the thermal conductivity of ICAs using composite theories (rules of mixtures).^{8,9} The previous works show that the thermal conductivity of model adhesives comprised of an epoxy-based binder containing Cu filler particles can be analyzed using a generalized equation based on Bruggemann's theory.

On the other hand, Agari et al.¹⁰ proposed their original equation containing a "coefficient of ease" in forming conductive chains of fillers, which is related to the critical volume fraction (CVF) in electrical conductivity (percolation threshold),^{11,12} for analyzing the thermal conductivity of polymer matrix composites. Because this equation can directly describe the effect of differences in the states of the percolation networks, it is worth also considering this equation for analyzing the thermal conductivity of ICAs, in addition to the use of Bruggemann's theory. These theories are often employed for analyzing the thermal conductivity of polymer matrix composites although the state of adhesion between the matrix and fillers can slightly affect the thermal conductivity.¹³

However, the physics of thermal conductivities of conductive adhesives has not been clarified precisely¹⁴; for example, some ICAs exhibit extremely high thermal conductivity¹⁵ that is inconsistent with the composite theories. In the present work, the thermal conductivity of a typical ICA comprised of an epoxy-based binder containing flake-shaped and spherical Ag particles is examined in order to clarify the factors that are influential in determining the thermal conductivities of electrically conductive adhesives.

EXPERIMENTAL PROCEDURE

A typical ICA (Namics Co. Ltd., Niigata, Japan) composed of an epoxy-based binder containing Ag filler particles (flake-shaped and spherical particles) was used in the present work. The total Ag loading was ~85 wt.%. The as-received ICA paste also appeared to contain some additives, such as nonreactive diluents. As recommended by the manufacturer, a suitable curing condition for the ICA was 150°C for 1.8×10^3 s. In the present work, the ICA was cured in an isothermal furnace under various conditions in the temperature range between 100°C and 180°C for 6.0×10^3 s. In addition, some specimens were post-annealed at 200°C for 10.8×10^3 s after the curing process.

The degree of conversion (α) of the ICA was estimated based on differential scanning calorimetry (DSC) measurements¹⁰ with a heating rate of $8.33 \times 10^{-2} \text{ }^\circ\text{C s}^{-1}$. The DSC equipment was also utilized to estimate the specific heat (C_p) of the cured and annealed ICA specimens. Their densities (d) were measured using the water immersion method. In order to measure their electrical resistivities (ρ) using the four-point probe method,

specimens printed on glass substrates with dimensions of $3.3 \text{ mm} \times 76 \text{ mm} \times 50 \text{ }\mu\text{m}$ were also cured and annealed under various conditions.

The thermal diffusivity (α_{therm}) of the ICA specimens was measured at ambient temperature using two different techniques, the laser flash (TC-7000, ULVAC Co., Ltd.) and the laser-heating Ångström (Laser PIT, ULVAC Co., Ltd.) methods,^{15–17} which were used to evaluate thermal diffusivity in the vertical and in-plane directions, respectively. The specimens for the thermal diffusivity measurements were formed on a polytetrafluoroethylene (PTFE) substrate. The specimens were removed from the substrate after the curing and post-annealing processes. The dimensions of the specimens were $10 \text{ mm} \times 10 \text{ mm}$ by $100 \text{ }\mu\text{m}$ to $400 \text{ }\mu\text{m}$ for the laser flash method, and $3 \text{ mm} \times 20 \text{ mm}$ by $20 \text{ }\mu\text{m}$ to $40 \text{ }\mu\text{m}$ for the laser-heating Ångström method. The thermal conductivities (λ) of the specimens were estimated using the following equation:

$$\lambda = \alpha_{\text{therm}} C_p d. \quad (1)$$

The thermal diffusivities of each sample were measured three times. Then, the average of the measured values was used for estimating thermal conductivities.

RESULTS AND DISCUSSION

Electrical Resistivities of ICA Specimens Cured at Various Temperatures

Specimens of the ICA paste were cured at 100°C, 150°C, and 180°C for 6.0×10^3 s in order to achieve full conversion. Figure 1 shows the electrical resistivity of these specimens measured at ambient

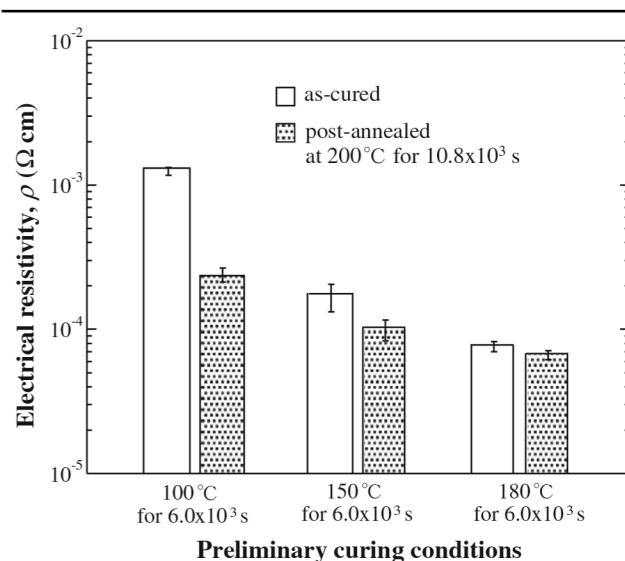


Fig. 1. Electrical resistivities (at ambient temperature) of ICA specimens cured at 100°C, 150°C, and 180°C for 6.0×10^3 s before and after annealing at 200°C for 10.8×10^3 s. These specimens already exhibited full conversion after preliminary curing processes at 100°C, 150°C, and 180°C.

temperature. Although the specimens were fully cured, their electrical resistivities were apparently different depending on the curing temperature. The electrical resistivities tend to decrease with increasing curing temperature. The curing temperature dependency of the electrical resistivity is considered to relate to variations in the interparticle (electrical) resistances due to the generation of internal stress during the curing and cooling processes.^{6,7}

Furthermore, the electrical resistivities of these specimens were significantly decreased by a post-annealing treatment at 200°C for 10.8×10^3 s, as also shown in Fig. 1. The electrical resistivities obtained after post-annealing clearly depend on the preliminary curing temperature. These experimental results indicate that the electrical resistivities that could be attained for the specimens after post-annealing depended on the curing state before the annealing process, even if the post-annealing temperature was much higher than the preliminary curing temperature.

Orientation of Filler Particles

Figure 2 shows a cross-sectional scanning electron microscopy (SEM) micrograph of a specimen prepared for estimating the thermal conductivity, in which the flake-shaped Ag particles are found to be orientated parallel to the squeegee direction of the ICA paste when it was printed on a PTFE substrate (in-plane direction). Because the orientation of the flake-shaped filler particles is expected to result in some anisotropy in the thermal conductivity of the specimens,¹⁵ the thermal conductivities in the vertical and in the in-plane directions were estimated separately using the laser flash and the laser-heating Angstrom methods, respectively.

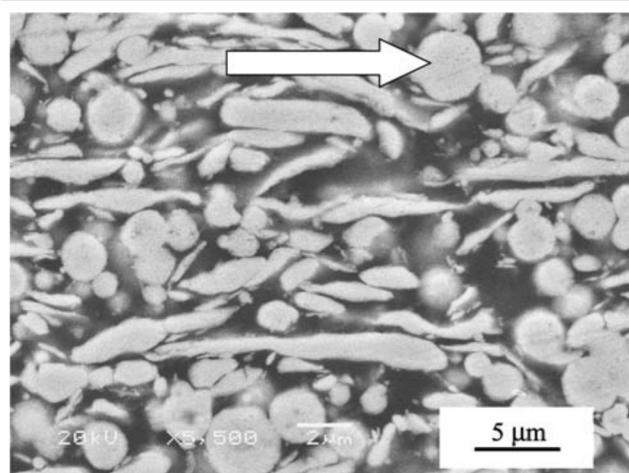


Fig. 2. Cross-sectional SEM micrograph of the cured ICA specimen. Ag flake particles tend to be orientated parallel to the squeegee directions of the ICA paste when it was printed on a PTFE substrate. The arrow indicates the squeegee directions.

Thermal Conductivities of ICA Specimens Cured at Various Temperatures

The thermal conductivities in the vertical direction of the ICA specimens cured at 100°C, 150°C, and 180°C for 6.0×10^3 s were estimated by the laser flash method. Figure 3 shows the relationship between the thickness of the specimens and their thermal conductivity at ambient temperature. In some specimens, numerous blisters originating from the vaporization of volatile components were formed when the specimen thickness was above 300 μm . Those specimens that exhibited blister formation were not used for the thermal conductivity measurements, since the uniformity of their microstructure is not guaranteed. When the specimen thickness is 200 μm to 400 μm , thermal conductivity can be measured sufficiently well because the peaks due to noise associated with the laser irradiation and to thermal conduction through the specimens are clearly separated in the temperature profiles. By contrast, when the specimen thickness was below 200 μm , these peaks were overlapped. Because of the overlapping of the peaks in the temperature profiles, the thermal conductivities of specimens ~ 100 μm in thickness cannot be evaluated when the curing temperature is 180°C. Although the thermal conductivities of specimens ~ 100 μm in thickness can be estimated when the curing temperature is 100°C or 150°C, the credibility of these data needs more consideration. At least, the thermal conductivities of the specimens do not exhibit a meaningful dependence on their thickness in the range between 200 μm and 400 μm . In this paper, experimental results from the specimens with a thickness of ~ 280 μm were used for discussions concerning the thermal conductivities in the vertical direction.

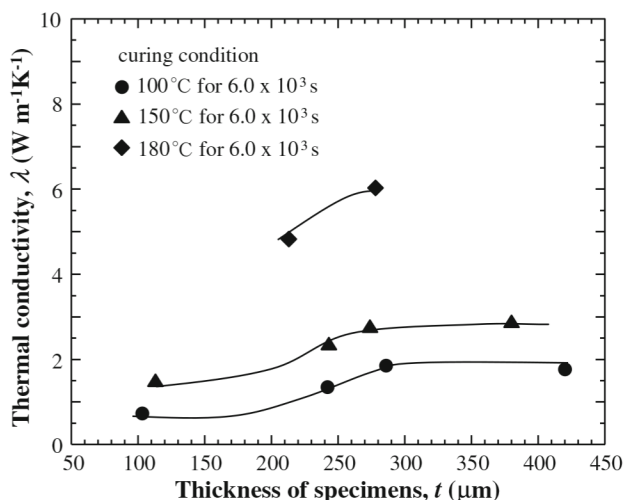


Fig. 3. Relationship between the specimen thickness and thermal conductivity measured using the laser flash method (in the vertical direction) at ambient temperature for ICA specimens cured at 100°C, 150°C, and 180°C for 6.0×10^3 s.

On the other hand, the thermal conductivities of specimens with thicknesses of 20 μm to 40 μm (which is a similar thickness to that of ICAs used in practical applications) in the in-plane direction were estimated by the laser-heating Ångström method. The measurement length in the horizontal direction of the specimens was set to 2 mm to 4 mm when the thermal diffusivity was evaluated using this method. Figure 4 shows the thermal conductivities that were measured using the laser-heating Ångström method for specimens cured at various temperatures for 6.0×10^3 s. The values of thermal conductivities measured using this method are apparently larger than those measured using the laser flash method. Some of the papers¹⁵ that cover the laser-heating Ångström method describe how this method can provide a similar value of thermal conductivity to that estimated using the laser flash method when the same specimen is used. According to our preliminary examinations in which we used another type of ICA specimen composed of an epoxy-based binder containing only spherical Ag particles, there were also no significant differences between the thermal conductivities when measured using the two different methods. These methods are considered to intrinsically provide any differences in the values of the thermal conductivities for ICA specimens. Therefore, the specimens are considered to exhibit anisotropy in thermal conductivities between the vertical and in-plane directions. The thermal conductivities in the in-plane direction are much higher than those in the vertical direction. Because Campbell et al. reported similar anisotropy in thermal conductivity of ICA specimens sandwiched between Si wafers,¹⁵ the anisotropy is considered to be often realized in the die-attach applications.

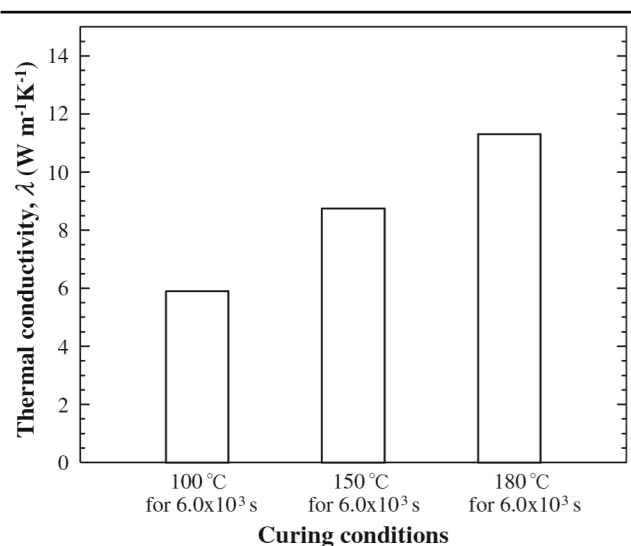


Fig. 4. Thermal conductivities of specimens measured using the laser-heating Ångström method (in the in-plane direction) at ambient temperature.

From the results indicated in Figs. 3 and 4, the thermal conductivities in both the vertical and in-plane directions increase with increasing curing temperature, even when the specimens were fully cured. The electrical resistivities of these specimens decrease with increasing curing temperature, as shown in Fig. 1. This dependence of electrical resistivity on curing temperature means that different states are induced in the percolation network in terms of variations in the interparticle electrical resistance by changing the curing temperature, which results in some variations in the curing states of the adhesive binder. The variation in state of the percolation network due to differences in curing temperature is also considered to influence the thermal conductivities of these specimens, as well as their electrical conductivities.^{6,7}

Theoretical Analysis of Thermal Conductivities of ICA Specimens

The thermal conductivities of polymer matrix composites containing metal particles are usually analyzed using a composite theory, for example, Bruggemann's theory. In fact, the present authors had clarified that the thermal conductivity of an epoxy-based composite containing Cu particles (electrical resistivity $6 \times 10^{-1} \Omega \text{ cm}$) can be analyzed using Bruggemann's equation^{8,9}:

$$1 - V_f = \frac{\lambda_c - \lambda_f}{\lambda_m - \lambda_f} \left(\frac{\lambda_m}{\lambda_c} \right)^{\frac{1}{n}}, \quad (2)$$

$$n = \frac{3}{\Psi}, \quad (3)$$

where V_f , λ_c , λ_m , and λ_f are the volume fraction of the fillers, and the thermal conductivity of the composites, the matrix, and the fillers, respectively. Ψ is the sphericity of the filler particles, which is defined as the ratio of the surface area of a sphere with a volume equal to that of the particle to the surface area of the particle.¹⁸

Here, we predict the thermal conductivities of epoxy-based composites containing Ag particles based on the composite theory. Figure 5 shows the predicted thermal conductivities of epoxy/Ag composites. In order to calculate thermal conductivity, the Ag fillers were assumed to be either spherical ($\Psi = 1$) or flake ($\Psi = 0.67$) shaped. Because the volume fraction of the Ag particles for the ICA that was used in this work was estimated to be ~ 0.4 , the values of thermal conductivity measured with the laser flash and the laser-heating Ångström methods (Figs. 3 and 4) are apparently higher than those predicted using the Bruggemann's equation. The differences in thermal conductivity between the experimental and predicted values become larger as the curing temperature of the specimens

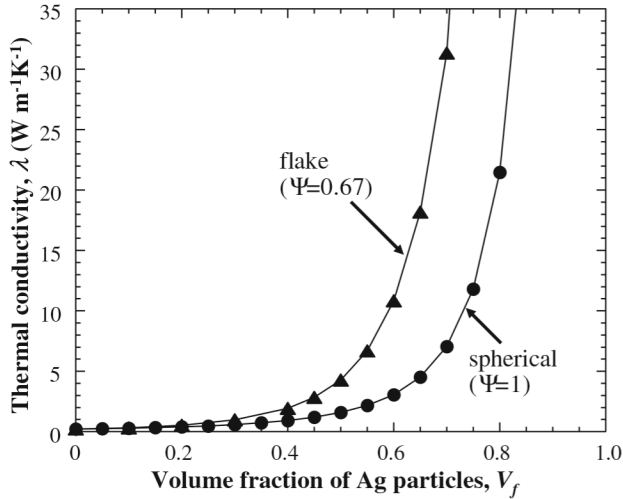


Fig. 5. Prediction of the dependence of thermal conductivity on the content of filler for Ag/epoxy systems using the Bruggemann's equation. The Ag fillers were assumed to have spherical ($\Psi = 1$) and flake ($\Psi = 0.67$) shapes.

increases, which results in a decrease in the electrical resistivity.

The discrepancy between the experimental results and predicted values is likely to originate from the following two factors. One is the effect of the orientation of the flake-shaped fillers in the ICA specimens. Bruggemann's theory assumes a random dispersion of filler particles. However, the flake-shaped fillers tend to orientate along the in-plane direction in the ICA specimens. Hence, the thermal conductivity of the specimens possibly exhibits anisotropy.

The discrepancy is also considered to be related to the contribution from the conducting electrons in the ICA specimens. Here, we consider the thermal current that originates from the electrical current in metallic materials. In this case, the relationship between the electrical conductivity ($\sigma = \rho^{-1}$) and the thermal conductivity (λ) is described by the Wiedemann–Franz (W–F) law^{19,20} as Eq. 4:

$$\frac{\lambda_{el}}{\sigma} = LT \quad (4)$$

where L and T are the Lorenz number and temperature, respectively. The Lorenz number of free electrons is $2.45 \times 10^{-8} \text{ W } \Omega \text{ K}^{-2}$. The W–F law is introduced based on the assumption that the effectiveness of each scattering process in degrading the electrical current is equivalent to the effect in degrading the thermal current. The thermal conductivities of metals are believed to be primarily governed by the contribution of the conducting electrons (free electrons) since this can be analyzed practically using the W–F law.

The essence of the W–F law can be understood by using the semiclassical model of electron dynamics based on the relaxation-time approximation. However, the relaxation-time approximation is

not always valid in the case of real materials. When the relaxation-time approximation is not made, the W–F law is only valid when the energy of each electron is conserved in each scattering event (elastic scattering).¹⁹ In the case of electron conduction in polycrystalline metals, the scattering processes that affect the conducting electrons can be briefly categorized into intragrain and intergrain (grain-boundary) scattering processes. However, the effects of grain-boundary scattering (such as tunneling) are often negligible. Hence, the validity of the W–F law can be analyzed by only considering intragrain scattering processes, such as impurity scattering and phonon scattering. However, the W–F law is not always valid even for metals.^{19,20} The W–F law is generally well obeyed at both high (above the Debye temperature) and extremely low temperatures. In the intermediate temperature range, the W–F law fails due to the effects of inelastic phonon scattering processes.

By contrast, interparticle scattering processes need to be considered along with intraparticle scattering processes in the case of electron conduction through percolation networks of filler (metallic) particles in ICAs. The W–F law is valid for intraparticle conduction in the temperature range above ambient temperature. Interparticle conduction is believed to mainly occur by a tunneling mechanism.^{1,21,22} Tunneling can be regarded as an elastic scattering process when there are no impurity levels that can interact with the conducting electrons. Consequently, the relationship between bias and tunneling current will be ohmic. In fact, it had been reported that nonohmic behavior is not detectable in practical ICAs.^{1,21} Therefore, the W–F law is considered to apply adequately to the conducting processes in ICAs, including both intraparticle and interparticle conduction mechanisms. The contribution (partial thermal conductivity) of conducting electrons to the thermal conductivity of ICAs can be estimated using the W–F law.

The magnitude of the contribution from conducting electrons to the thermal conductivity depends on the electrical resistivity of the ICAs, as described in Eq. 4. Figure 6 shows the relationship between the contribution from conducting electrons to the thermal conductivity and the electrical resistivity of ICAs as calculated using the W–F law. This figure suggests that the contribution from the conducting electrons is negligible when the electrical resistivity of the ICAs is above $10^{-3} \Omega \text{ cm}$. The thermal conductivity of ICAs with such high electrical resistivities can be analyzed effectively using composite theories, including Bruggemann's theory. However, the effect of conducting electrons increases significantly in the resistivity range below $10^{-3} \Omega \text{ cm}$. The contribution from conducting electrons needs to be taken into account in order to analyze the thermal conductivity of advanced ICAs that have a low electrical resistivity, below $10^{-3} \Omega \text{ cm}$.

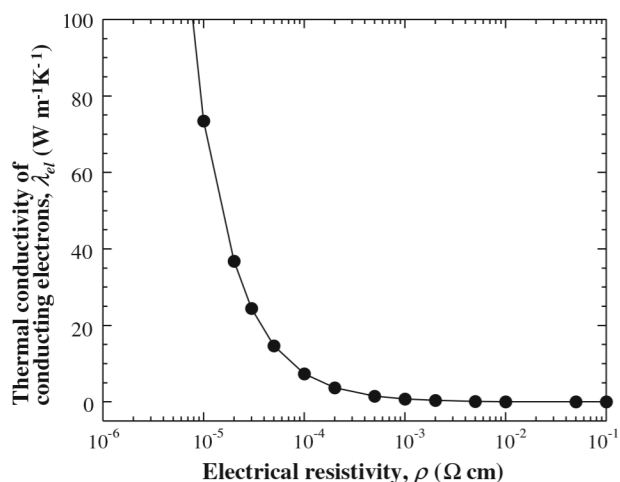


Fig. 6. Relationship between the contribution of the conducting electrons to the thermal conductivity and the electrical resistivity of ICA specimens as predicted by the W-F law.

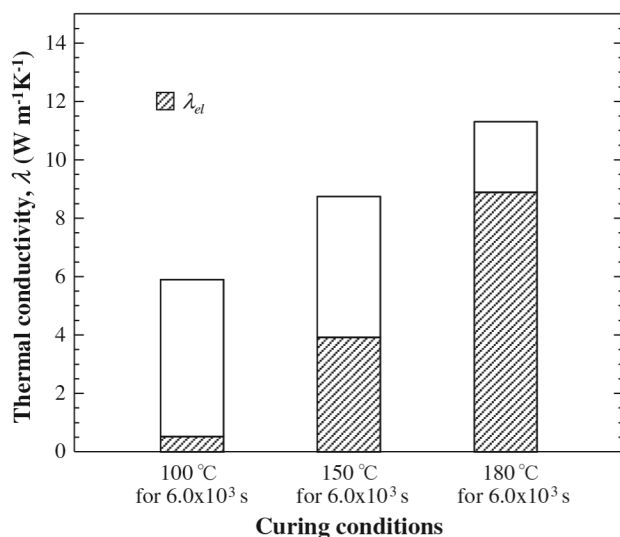


Fig. 7. Total and partial thermal conductivities due to the effect of conducting electrons (λ_{el}) in the horizontal direction of ICA specimens cured at 100°C, 150°C, and 180°C for 6.0×10^3 s.

The contribution from conducting electrons to the total thermal conductivity of the ICA specimens is derived from the experimental values for electrical resistivity using the W-F law. Here, we used the thermal conductivities along the in-plane direction that were measured using the laser-heating Ångström method, since the electrical resistivities as measured using the four-point probe method are not the values along the vertical direction. Figure 7 shows the total thermal conductivity and the contribution from the conducting electrons (λ_{el}) of the ICA specimens at ambient temperature. In the case of as-cured specimens, the contribution of the conducting electrons to the total thermal conductivity increases with increasing curing temperature.

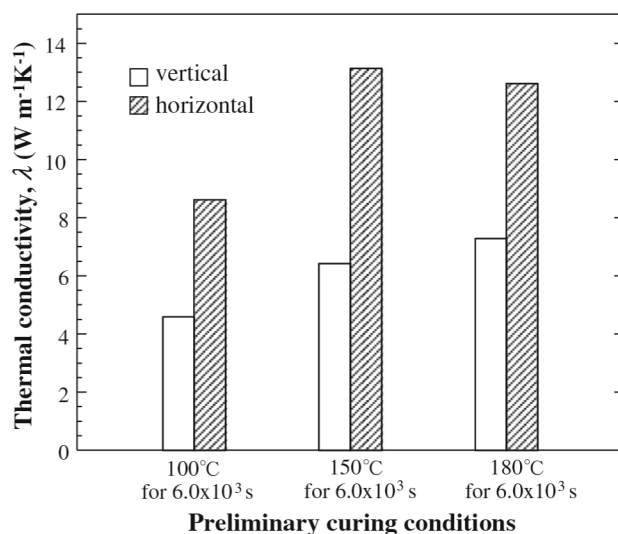


Fig. 8. Thermal conductivities in the vertical and in-plane directions of ICA specimens after annealing at 200°C for 10.8×10^3 s.

The increase in the contribution of the electrical conduction seems to result in an increase in the thermal conductivity of these specimens.

Post-annealing Effect on Thermal Conductivities of ICA Specimens

The electrical resistivities of the ICA specimens were decreased by post-annealing at 200°C for 10.8×10^3 s, as described in the section “Electrical Resistivities of ICA Specimens Cured at Various Temperatures”. Figure 8 shows the thermal conductivities of specimens that were measured using the laser flash and the laser-heating Ångström methods after post-annealing at 200°C for 10.8×10^3 s. The thermal conductivities of these specimens were increased significantly by post-annealing concurrent with decreases in their electrical resistivities (increases in electrical conductivities). The values of thermal conductivity achieved after post-annealing clearly depend on the preliminary curing temperature in a similar manner to the post-annealing effect on electrical conductivities.

The contributions to the thermal conductivities from the effects of the conducting electrons for these specimens apparently increase due to the post-annealing treatment. Here, the effect of annealing on the thermal conductivity can also be analyzed using the W-F law. By assuming that the variation in electrical conductivity due to post-annealing is $\Delta\sigma$, the variation in thermal conductivity relating to variations in electrical conductivity ($\Delta\lambda_{el}$) is indicated as Eq. 5.

$$\frac{\Delta\lambda_{el}}{\Delta\sigma} = LT. \quad (5)$$

From the experimental results for the decreases in electrical resistivity due to the post-annealing

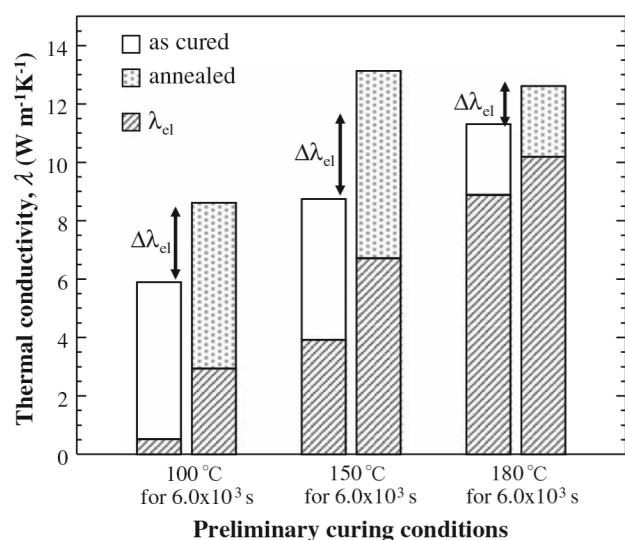


Fig. 9. Comparison of thermal conductivities in the in-plane direction before and after annealing at 200°C for 10.8×10^3 s. The difference in the contribution of conducting electrons to the thermal conductivity due to post-annealing ($\Delta\lambda_{el}$) was estimated using the W-F law.

treatment at 200°C shown in Fig. 1, we calculated the values of $\Delta\lambda_{el}$ for the specimens that underwent preliminary curing at 100°C, 150°C, and 180°C. Figure 9 shows the thermal conductivities of the specimens before and after post-annealing at 200°C as measured using the laser-heating Ångström method by comparing with the values of $\Delta\lambda_{el}$. The summation of the values of thermal conductivity before post-annealing and $\Delta\lambda_{el}$ almost agree with the thermal conductivities after post-annealing for these specimens. Although the specimens cured at 150°C exhibits relatively high thermal conductivity after post-annealing, this measured value possibly contains significant error due to reasons such as variation in thickness (unevenness) of the sample. At least, the increases in thermal conductivity due to post-annealing can be interpreted as almost entirely due to effect of the increase in electrical conductivity.

Fundamental Factors Determining Thermal Conductivities of ICAs

From our previous^{8,9} and present works, the determining factors for the thermal conductivities of ICAs are found to be divided into two categories. One is the effect of the composite structure, which is composed of an adhesive binder containing conductive filler particles. The composite-structural factor, which is related to the shape, size, distribution (orientation), and volume fraction of the filler particles, can be analyzed using a composite theory based on an appropriate model.^{8,9,23} When significant void formation occurs in ICAs, we can analyze the thermal conductivities using a model for a composite structure including voids.^{8,9} The other factor is the contribution from the conducting

electrons (the thermal current that originates from the electrical current). This factor becomes important when the electrical resistivities of the ICAs are below $10^{-3} \Omega \text{ cm}$, as shown in Fig. 6. Therefore, decrease in the electrical resistivities is effective in order to increase the thermal conductivities of ICAs.

In order to introduce a theory for analyzing the thermal conductivities of ICAs including both of these factors, the state (quality) of the percolation network in terms of variations in the interparticle electrical resistances should be taken into account. Conventional rules of mixtures (such as Bruggemann's theory) do not take the state of the percolation network sufficiently into account when considering the thermal conductivity. Hence, these theories are only applicable for analyzing the thermal conductivities of ICAs that exhibit relatively high electrical resistivities (above $10^{-3} \Omega \text{ cm}$) because of the low contribution of the electrical conduction. Agari's equation¹⁰ is currently the only theory that considers the state of the percolation network. In Agari's equation, a percolation factor is defined using the CVF in electrical conductivity (percolation threshold).

However, the electrical resistivities of ICAs can be varied by changing the curing and post-annealing conditions. The dependence on the thermal history of the electrical resistivities indicates that the electrical conductivities along the percolation network vary depending on the curing states of the adhesive binder. Therefore, in order to interpret the dependence on thermal history, the percolation factor is difficult to represent in practical terms by using a CVF, such as is used in Agari's scheme. A novel analytical method including a reasonable definition of the percolation factor needs to be developed in order to analyze the thermal conductivity of advanced conductive adhesives.

CONCLUSIONS

In this paper, we analyzed the thermal conductivities of ICA specimens composed of an epoxy-based binder that were cured and post-annealed at various conditions, in order to discuss the physical factors relevant in determining the thermal conductivity. The main results obtained in the present work are summarized below:

- (1) ICA specimens with an orientated dispersion of flake filler particles exhibit anisotropy in their thermal conductivities.
- (2) The thermal conductivities of ICAs are influenced by a contribution from conducting electrons as well as by composite-structural factors.
- (3) The conventional composite theories are only applicable for analyzing the thermal conductivities of ICAs that exhibit relatively high electrical resistivities (above $10^{-3} \Omega \text{ cm}$) due to the low contribution of electrical conduction.

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