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Spatial spreading behavior of transmitted light from glass particle-dispersed epoxy matrix composites

T. NAGANUMA, Y. KAGAWA*
Institute of Industrial Science, The University of Tokyo, 4-6-1, Komaba, Meguro-ku, Tokyo 153-8505, Japan
E-mail: kagawa@iis.u-takyo.ac.jp

The spatial spreading behavior of glass particle-dispersed epoxy matrix optical composites has been observed, and the composites used have light transmittance in the visible wavelength region. The effect of (i) particle volume fraction, (ii) particle size and (iii) refractive index difference between the particle and matrix on the spreading behavior are discussed. The spatial spreading behavior of transmitted light from a back surface of the composites is found to be strongly correlated with light scattering behavior of the composite. The increase of spatial light spreading also increases spatial light transmittance of the composite. © *2003 Kluwer Academic Publishers*

1. Introduction

Composites with light transmission potential have been developed and are termed "optical composites." Potential of optical composites have been reported in various materials systems, such as short SiCaAlON-fiber glass matrix [1, 2], glass fiber-reinforced polymer matrix [3, 4], and glass particle-dispersed polymer matrix [5–11]. The glass particle-dispersed polymer matrix composites have received special attention for their application of electrical packaging materials to opto-electric devices because they are expected to satisfy both high light transmittance and low thermal expansion coefficient, which is difficult to achieve in pure polymers [9].

Most of the studies on the glass particle-dispersed epoxy matrix composites have been focused on the achievement of high light transmittance of the composites. The effect of particle volume fraction [6, 10], refractive index difference [7], and size and shape of glass particle [8, 10] on light transmittance have been reported. The effect of the light scattering process on the transmittance and the correlation between light transmittance and transparency of the composite were evaluated using the change of pico-second order pulse profile of transmitted light [12, 13] and the phase shift of transmitted light [14]. Although these reports demonstrate the effect of light scattering behavior on the light transmittance of the composites, the major attention is focused light scattering behaviors inside the composites.

For the applications of the composite as light transmitting package materials, the spatial spreading behavior of light transmitted from the back surface of composites is also important. This behavior, however, has not yet been reported. The purpose of this study was to identify the spatial spreading behavior of transmitted light and to discuss the effect of light scattering in the composite on the light spreading behavior of transmitted light in glass particle-dispersed epoxy matrix optical composites.

2. Experimental procedure

2.1. Fabrication of glass particle-dispersed epoxy composites

A glass particle-dispersed epoxy matrix composite was used because this system allowed close refractive index matching at a temperature range of $T = 298$–373 K and a wavelength range of $\lambda = 600$–800 nm. In addition, light spreading behavior in this composite was relatively well known [10, 13]. The chemical composition of the glass particles was SiO_2 (60.0 wt%), Al_2O_3 (17.4 wt%), B_2O_3 (3.8 wt%), MgO (7.6 wt%), and CaO (11.2 wt%) [15]. The glass particles are irregular in shape [8, 10] and average particle diameter is defined as the median value (50% of the particle size distribution) measured by a laser diffraction type particle size analyzer (SALD-2000, Shimadzu Corp., Kyoto, Japan). The average particle diameter is hereafter referred to as the particle diameter, d_p. The average particle diameters were $d_p = 26$ and 85 μm [8, 10], which were much larger than the wavelength of light in the visible wavelength region (450–750 nm).

The matrix material was a clear grade epoxy resin (Epikote 828, Yuka-shell Epoxy Corp., Tokyo, Japan). The resin was cured using 46 wt% of curing agent

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* Author to whom all correspondence should be addressed.

TABLE I Optical properties of a glass particle and the epoxy matrix in a bulk form

	Constants			Refractive index,
	a_1	$a_2(\times 10^3)$	$(\Delta n/\Delta T)_D$	n ($\lambda = 633$ nm, $T = 298$ K)
Glass particle	0.7515	7.256	-1.607×10^{-5}	1.537
Epoxy matrix	0.7521	11.72	-12.67×10^{-5}	1.544

(MH-700E, Shin-Nihon-Rika Corp., Tokyo) and 0.27 wt% of accelerator (DBU, Sun-Apro Corp., Tokyo). These three base components were mixed together in ambient air and poured into a completely dry cylindrical soda-borosilicate glass mold. After degassing, the mixture was cured at 373 K for 4 h.

Refractive index of the pure epoxy was measured using an Abbe refractometer (2T, Atago Co., Ltd., Tokyo) at a wavelength from 480 to 700 nm. The measurement was done at temperatures of 298–373 K using a temperature control unit (Labothermo LH-2020, Advance Corp., Tokyo). The refractive index of glass particle was measured by a maximum transmission method [16, 17] using an immersion liquid (refractive index standard liquid, Cargille Laboratories Inc., USA). The temperature dependence of the refractive index of the particle was obtained by changing the temperature and refractive index of the liquid, respectively. The wavelength dependence of refractive index of the glass and the pure epoxy matrix are approximated using the single-term Sellmeier dispersion formula [18],

$$\frac{1}{n^2(\lambda) - 1} \approx a_1 - \frac{a_2}{\lambda^2} \qquad (1)$$

where n is the refractive index of raw material, λ is wavelength of incident light, and a_1 and a_2 are constants. Table I lists optical values for the pure epoxy matrix and the glass particle; constants of a_1 and a_2, temperature dependence of refractive index at $\lambda = 589$ nm, $(\Delta n/\Delta T)_D$, and refractive index at $\lambda = 633$ nm and $T = 298$ K. Other properties of the same glass particle and the epoxy matrix are reported elsewhere [8–10].

The glass particle was incorporated into the mixture of epoxy matrix before curing. The fabrication process of the composite was detailed earlier [8–10]. The particle volume fractions of the composite, f_p, were 0.1 and 0.4, and where controlled by measuring the weight of the particle before mixing, knowing the density of the epoxy matrix after mixing. The glass particle distribution in the epoxy matrix was observed by an optical microscope (BX-10, Olympus Corp., Tokyo, Japan) using the polished surface of the composites.

2.2. Direct observation of spatial spreading behavior of transmitted light

The composites were cut into a rectangular specimen of $10 \times 20 \times 1.5$ mm with a thickness of 10 ± 0.2 mm for direct observation of the spatial spreading behavior of light transmitted from their back surface. The flat surfaces of the specimens were polished up to 1 μm with a diamond paste finish to achieve a mirror surface. To eliminate stresses introduced on the composite surface during the cutting and polishing processes, they were annealed at a temperature of 373 K for 3 h.

Fig. 1 shows a schematic drawing of the experimental setup for direct observation of the transmitted light. A He-Ne laser (05-LHP-928, minimum output power ~35 mW, Melles Griot KK, Tokyo) with a wavelength of $\lambda = 633$ nm was used as a light source. The incident beam direction was perpendicular to the specimen surface. The beam had a Gaussian profile with a deviation angle of less than 1 degree. The beam spot diameter, ϕ, defined as the incident beam size on the specimen surface, was $\phi \sim 1.0$ mm. A white paper screen was placed parallel to the beam direction (cf. Fig. 1a), and the light scattering image projected on the screen ($L \sim 40$ mm, $W \sim 15$ mm) was recorded by a digital camera (Camedia C-2002, Olympus Optical Co., Ltd., Tokyo). The offset angle of the image plane of the camera from the screen plane was set to $\theta = 90 \pm 2$ degrees (angle θ is defined in Fig. 1b). The laser light was incident to the boundary between the composite and the screen (cf. Fig. 1c), and the recording of the images

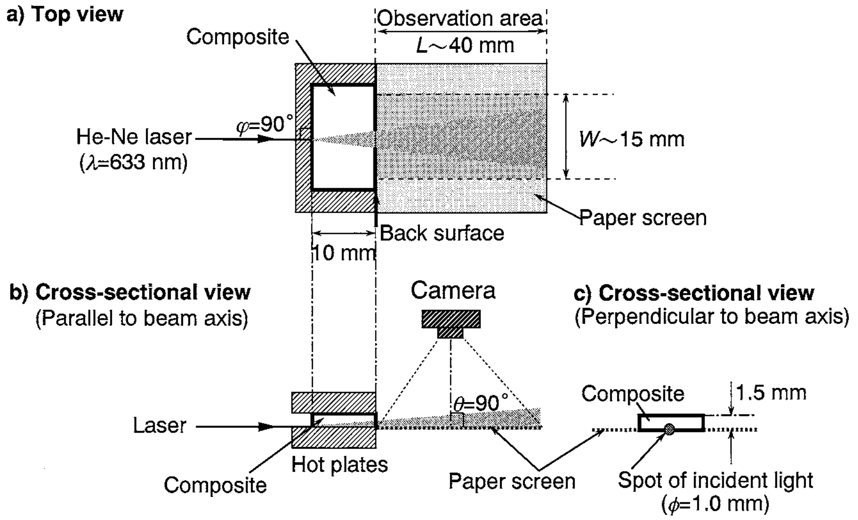
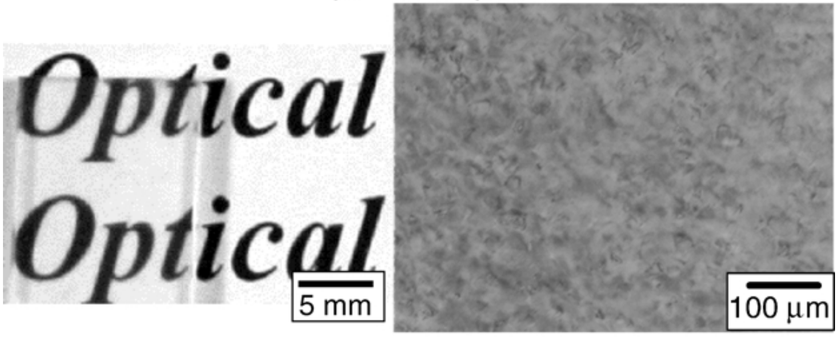


Figure 1 Schematic diagram for observation of light scattering profile in (a) top view, (b) cross-sectional view (parallel to beam axis), and (c) cross-sectional view (perpendicular to beam axis).

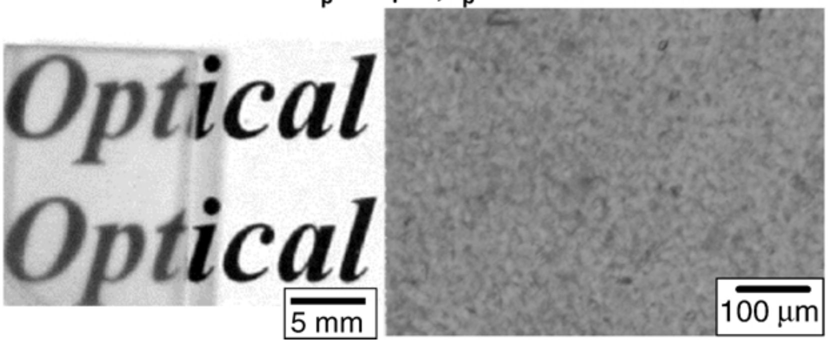
(a) Matrix

(b) Composite

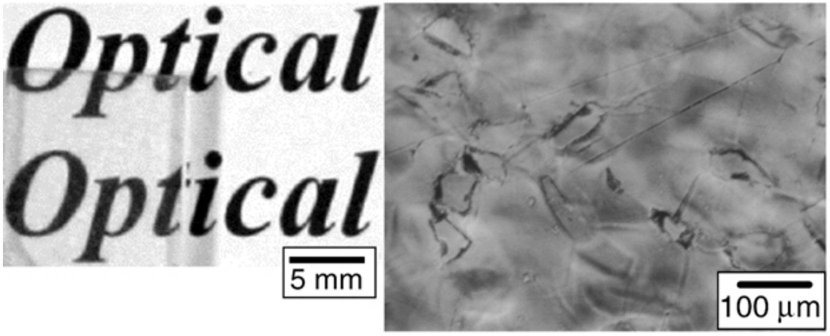
d_p=26 μm, f_p=0.1



d_p=26 μm, f_p=0.4



d_p=85 μm, f_p=0.1



d_p=85μm, f_p=0.4

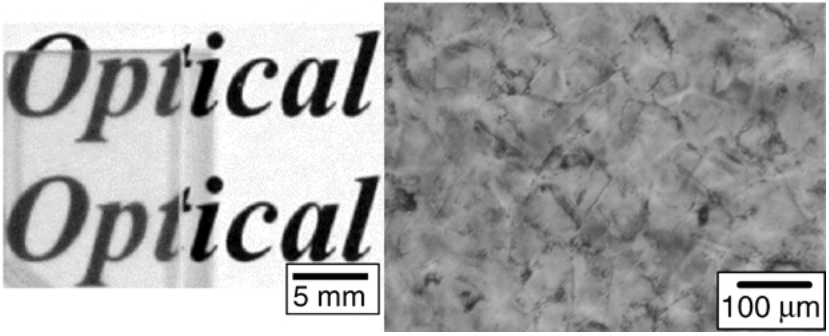


Figure 2 Typical appearance and microstructure of epoxy matrix (a) and composites (b).

parallel to the incident light direction were recorded within a cross-section of the spreading light reflected on the screen. This was the same method used in the previous papers [10, 13].

The refractive index difference between the glass particle and epoxy matrix in the composite was achieved by a changing the temperature in the composite [7]. The change in the measured temperature creates different refractive index between the glass particle and matrix because of the change in their refractive indices (Table I) and thermal stress [7]. The composite was heated up to a given set-point temperature from both sides of the specimen using two hot plates, in which temperature was controlled by a temperature controller (Labothermo LH-2020, Advantec MFS, Inc., CA, USA). The spreading light on the screen was observed after 15 min of heating at the set-point temperature to allow uniform temperature distribution in the specimen.

2.3. Light transmittance of the composites

In-line light transmittance of the composites with a thickness of $t = 1.5 \pm 0.2$ mm was measured at a wavelength range of 200–1100 nm using a transmittance spectrometer (V-530 type, resolution <2 nm, Jasco Corp., Tokyo). The composite was put into a square-shaped transparent glass cell with inner dimensions of $10 \times 10 \times 40$ mm. The surface finish and annealing schedule of the specimen was the same as that by direct observation. The distance between the back surface of the composite to the detector was set at $L \sim 70$ mm. The temperature of the specimen in the glass cell was varied using a temperature controller (ETC-505, Nihon Bunko Corp., Tokyo) at a range of $T = 298$–373 K. Heating of the composite was done through the glass cell and accuracy of the temperature in the used range was less than ±0.5 K. The measurement was made after 10 min at a given temperature to achieve uniform temperature distribution in the composite specimen.

3. Results and discussion

3.1. Appearance and microstructure of composites

Fig. 2 shows the appearances of the pure epoxy matrix (a) and the composites (b) 1.5 ± 0.2 mm thick and typical microstructures on the polished surface of the composites (b). The glass particles are randomly distributed in the epoxy matrix without noticeable segregation. This distribution is observed independent of the particle size and volume fraction, respectively. Characters underneath the pure epoxy matrix (a) and the composites (b) are legible for all of the specimens, indicating that the fabricated composites have light transmittance in the visible wavelength region (λ =450–750 nm). However, the light transmitting level of the composites obviously decreases from that of the pure epoxy matrix, suggesting the existence of a second phase reduces the light transmittance. Increase of the particle volume fraction, f_p, and decrease of the size, d_p, respectively, decreases visible light transmittance of the composite. This appearance and tendency are identical to that reported in similar kinds of composites [5–14].

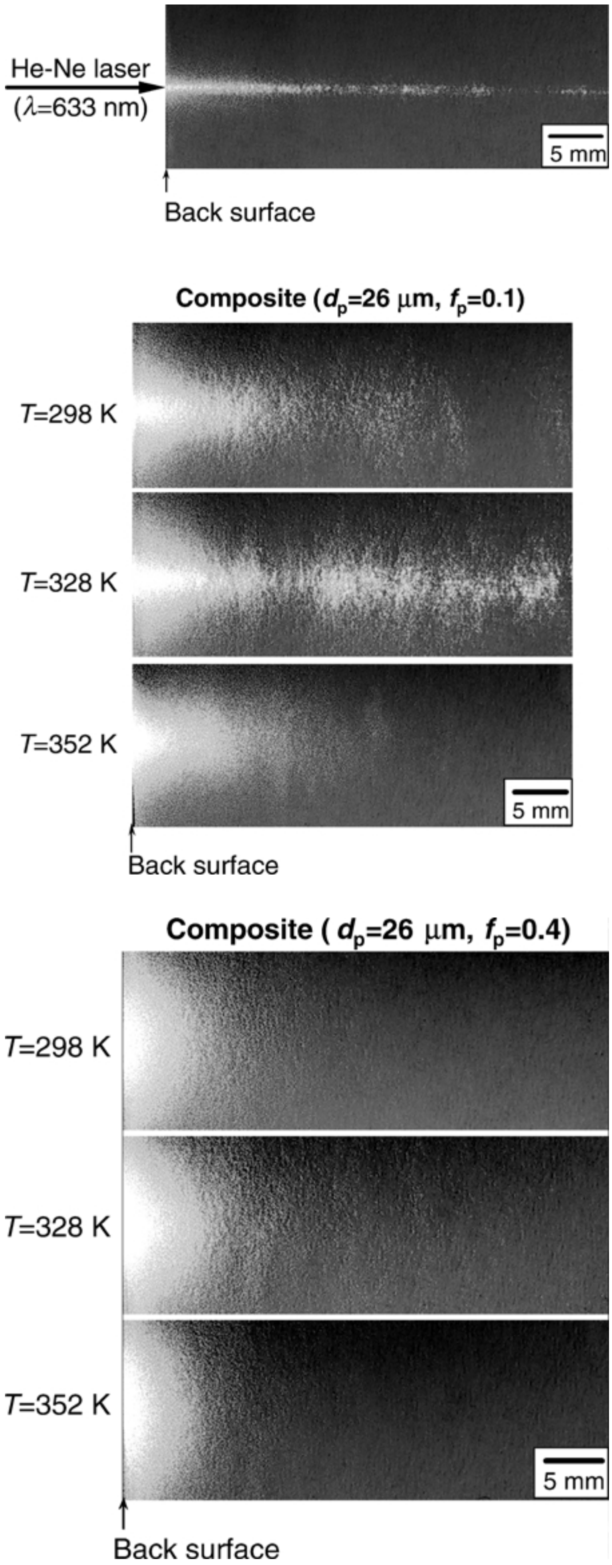


Figure 3 Temperature dependence of spatial spreading behavior of transmitted light for pure epoxy and composites with $d_p = 26\ \mu$m and $f_p = 0.1$ and 0.4.

3.2. Effect of particle size and volume fraction on spatial spreading of transmitted light

Spatial spreading behavior of transmitted light from the pure epoxy and the composites are shown in Figs 3 and 4. In the pure epoxy matrix, the transmitted light from the specimen is straight without noticeable spreading in the transverse direction of the incident beam axis. A

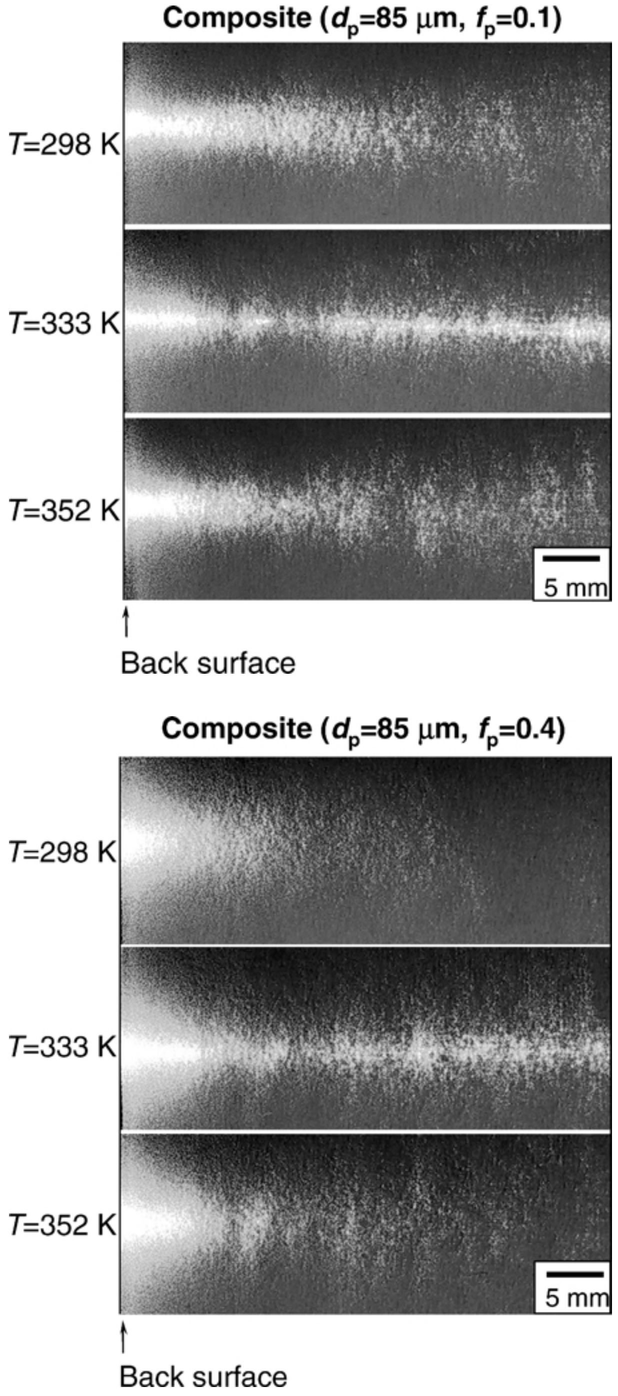


Figure 4 Temperature dependence of spatial spreading behavior of transmitted light for the composites with $d_p = 85\ \mu$m and $f_p = 0.1$ and 0.4.

slight diffusion behavior near the back surface of the specimen is probably caused by the effect of Rayleigh scattering by molecular fluctuation [19, 20]. On the other hand, the light apart from the back surface of the composites shows extensive spatial spreading behavior, which is observed for all composite specimens.

The spatial spreading behavior differs with the particle size, d_p, and particle volume fraction, f_p. The effects of these and the refractive index difference of raw materials, Δn, are understood from the transmitted light spreading patterns. Comparison of the photographs with the same particle volume fraction, f_p, and temperature, T, shows that the smaller particle size has extensive light spreading behavior. The spatial spreading of light among the same particle size, d_p, and temperature, T, becomes clear with higher particle volume fraction for both particle diameters. The increase of particle volume fraction, f_p, causes broadening of the light. These tendencies coincide with the increase of light scattering in the same kinds of composites. In the next section, the effect of refractive index will be discussed.

3.3. Effect of refractive index difference on spatial spreading of transmitted light

Light transmission spectra of the bulk glass and the bulk epoxy matrix at $T = 298$ K are shown in Fig. 5. These spectra of each material have nearly the same profile at a measured temperature range from 298 to 373 K. The raw materials have non-absorption behavior in a wavelength range of $\lambda = 500$–1100 nm because the in-line light transmittance of both materials is close to 90%. In this case, it can be assumed that light transmittance of the composite, I, is roughly proportional to the refractive index difference of the glass particle and matrix, Δn, i.e., $I \propto \Delta n$.

Temperature dependence of in-line light spectra of the composite with $d_p = 26$ and 85 μm and $f_p = 0.1$ is shown in Figs 6 and 7. The figures clearly demonstrate that the light transmission spectra of the composite show temperature dependence as expected from

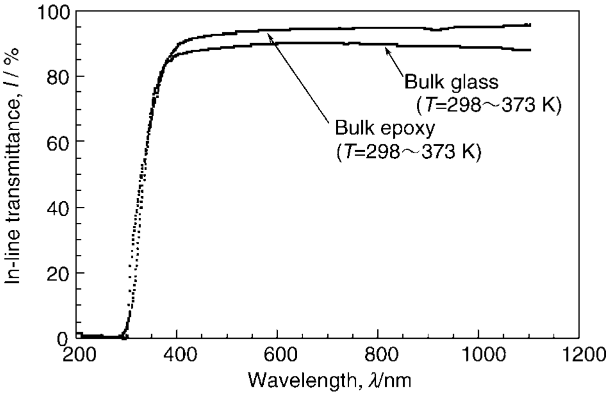


Figure 5 In-line light transmittance of the composite versus wavelength for bulk pure epoxy and bulk glass in a temperature range from $T = 298$ to 373 K.

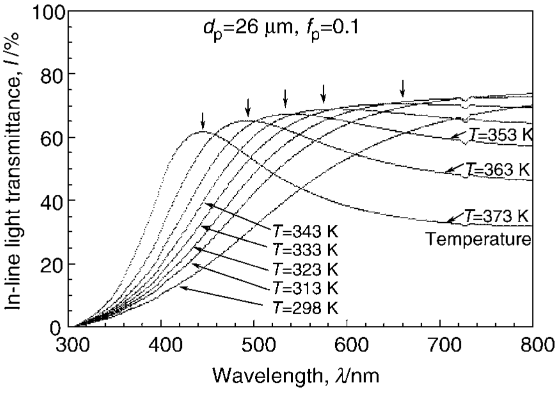


Figure 6 Effect of wavelength on in-line light transmittance of the composites ($d_p = 26\ \mu$m, $f_p = 0.1$) in a temperature range from $T = 298$ to 373 K.

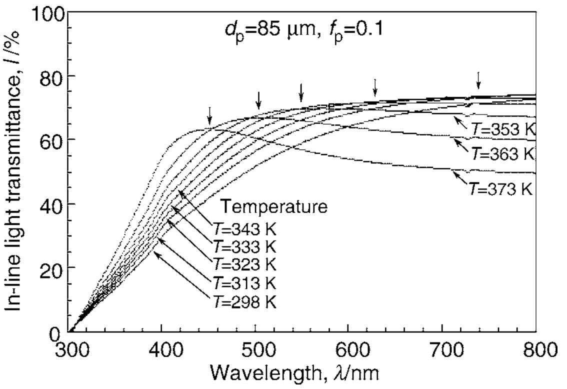


Figure 7 Effect of wavelength on in-line light transmittance of the composites ($d_p = 85$ μm, $f_p = 0.1$) in a temperature range from $T = 298$ to 373 K.

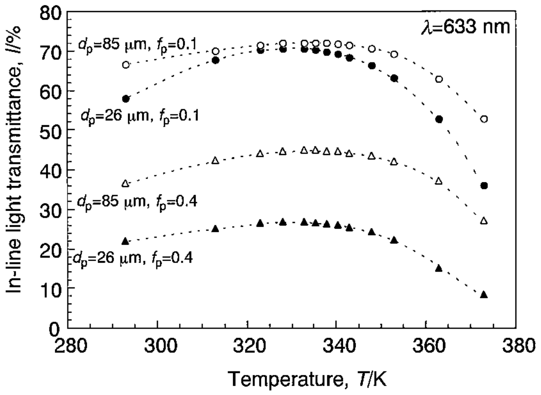


Figure 8 Temperature dependence of in-line light transmittance of the composite for a different d_p and f_p at a wavelength of $\lambda = 633$ nm.

the previous result [7]. At temperatures below 313 K, the light transmittance increases monotonically with increasing wavelength, while the spectra observed at temperature 323 K and above have a maximum light transmittance within the measured wavelength range (indicated by arrows). The value of in-line light transmittance and the wavelength at the maximum, respectively, decrease with increasing temperature. The previous result has shown that the refractive index difference between the glass particle and epoxy matrix changes with temperature and thermally-induced stress in the composite [7]. This temperature dependence of in-line transmittance spectra originates from the change in refractive index difference between the glass particle and the matrix.[1]

Fig. 8 shows plots of the in-line light transmittance versus temperature for the composite with $d_p = 26$ and 85 μm for $f_p = 0.1$ and 0.4. This wavelength is chosen because light-broadening behavior is obtained using a He-Na laser ($\lambda = 633$ nm). The plots indicate that the light transmittance is highly dependent on the temperature and has a maximum around a temperature of ~330 K. Although the absolute value of light transmittance differs, the temperature at the maximum transmittance is roughly independent of the particle size, d_p, and volume fraction, f_p. This result suggests that the refractive index difference between the glass particle and matrix in the composites, Δn, becomes minimum at a temperature around ~330 K. Both increase and decrease in temperature increase the difference of the refractive index difference, Δn. Therefore, the direct observation result at different temperatures reflects the effect of refractive index difference in the composite.

The spatial spreading of transmitted light (Figs 3 and 4) well correlates with the temperature dependence of light transmittance of the composites (Fig. 8). The spreading behavior of transmitted light within the same particle diameter, d_p, and volume fraction, f_p, becomes minimum at a temperature of 328 K for $d_p = 26$ μm and 333 K for $d_p = 85$ μm. The maximum light transmittance temperature (~330 K) is nearly the same as the temperature for the minimum spatial spreading of transmitted light. These tendencies well coincide with the direct observation of the spatial spreading of light traveling in the glass particle-dispersed epoxy matrix composites [10, 13]. Consequently, the transmitted light from the back surface of the composite depends on the spatial spreading behavior of light in the composites, because spatial spreading of light in the composite under the same particle size, d_p, and volume fraction, f_p, increases with increase in the refractive index difference, Δn [10, 13]. Summarizing the results, it is clear that the spatial spreading behavior of transmitted light from the back surface of the composite is predictable by the spatial spreading behavior of light in the composite. The spatial spreading behavior is controlled by the light scattering process in the composite.

4. Conclusion

The effect of light scattering behavior in glass particle-dispersed epoxy matrix composites on spatial spreading behavior of transmitted light from the composite has been studied. The spatial spreading of transmitted light increases with (i) increase of the particle volume fraction, f_p, (ii) decrease of particle size, d_p, and (iii) increase of refractive index difference of raw materials, Δn. These factors are the same as those that increase in light scattering behavior when light passes in the composite. That is, the spatial spreading behavior of transmitted light from the composite well correlated with the light scattering behavior in the composites. The same selection procedure is acceptable to control spatial spreading behavior of light emitted from the composites.

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[1] It is known that in-line light transmittance of glass particle-dispersed epoxy matrix composites reaches a peak when the refractive indices of the glass particle and the epoxy matrix completely match in the composite [7]. In the present case, because of light absorption in a shorter wavelength region, it is difficult to judge whether the maximum value originates from the Christiansen-like effect. However, it is clear in this study that the wavelength at the maximum reflects the closest match between the glass particle and matrix in the composite.

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