Dental Materials Journal 26(1): 38−44, 2007

Effect of Filler Properties in Composite Resins on Light Transmittance Characteristics and Color

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Recieved June 28, 2006/Accepted September 28, 2006

The purpose of this investigation was to examine the effect of filler particle size and shape as well as filler content on light transmittance characteristics and color of experimental composite resins. A mixture of 30 mol% Bis-GMA and 70 mol% TEGDMA was prepared as a base monomer and to which a photoinitiator (camphorquinone) and a co-initiator (N,N-dimethylaminoethyl methacrylate) were added. Four different irregular- and spherical-shaped filler types with an average particle size of 1.9-11.1 μm were added to the mixture in three different filler contents of 20, 30, and 40 vol%. Light transmittance characteristics including light diffusion characteristics of the materials were evaluated. Color values and color differences among filler contents of the materials were also determined. Materials containing smaller and irregular-shaped fillers showed higher light transmittance and diffusion angle distribution with a sharper peak, as compared with those containing larger and spherical-shape fillers. It was also found that there was a significant correlation between the specific surface area of fillers and the color difference of the materials containing the fillers.

Our results indicated that the shape of filler particles, as well as particle size and filler content, significantly affected the light transmittance characteristics−including light diffusion characteristics−and color of composite resins.

Keywords: Filler, Light transmittance, Color

INTRODUCTION

Visible light-cured composite resins have been widely used as direct filling and aesthetic restorative materials for anterior and posterior teeth. However, the problems of matching color to natural teeth are also widely known. Although several factors have been implicated in the failure of achieving an accurate color match to the surrounding teeth, the chief factor might be the difference in optical properties between the resin and natural teeth[1-4].

Composite resins are an optically translucent medium due to their structure of highly transparent base resin, small filler particles, and other additives. When a white incident light transmits through the material, the light would be multiply scattered by the small-size filler particles within the material before it emerges and reaches the eye of the observer−with the specific optical and color information of the material. Therefore, the color appearance of a material is a complex combination of its optical properties.

In particular, light transmittance characteristics are important optical properties to be considered for the color appearance of composite resins[3,5-7]. Some studies−with a view to improving the limited depth of cure of composite resins−have reported on the effects of filler properties, such as filler size and filler content, on the light transmittance characteristics of composite resins[8-12]. In practice, different commercial composite resins incorporate various filler particles with different shapes and sizes[13], and the difference in filler properties among products should result in different optical properties and color appearance of the restorations. However, a limited amount of information is available concerning the effect of filler properties on the color of composite resins. Therefore, it is important to understand how different filler properties affect the optical properties and color of composite resins with regard to effective color match to the teeth.

The purpose of this investigation, therefore, was to examine the light transmittance characteristics, including light diffusion characteristics, of experimental composite resins containing different filler types with different particle sizes and shapes and filler contents. We also investigated a possible relationship between the filler properties and color of composite resins.

MATERIALS AND METHODS

Experimental composite resins and specimen preparation

A blended mixture of 30 mol% Bis-GMA (bisphenol A bis 2-hydroxy propyl methacrylate, Polysciences Inc., Warrington, PA, USA) and 70 mol% TEGDMA (triethyleneglycol dimethacrylate, Wako Pure Chemical Industries Ltd., Osaka, Japan), both individually available as commercial products, was used

Table 1 Fillers used in this study.

Filler	Shape	Average particle size (μm)	Specific density (g/cm^3)	Specific surface area (m^2/g)
VX-X	Irregular	1.9	2.72	10.24
VX-SR	Irregular	3.6	2.69	7.06
EMB-10	Spherical	5.4	2.60	0.73
EMB-20	Spherical	11.1	2.61	0.48

Table 2 Materials used.

Code	Filler	Mixing proportion		Amount of silane (g) against filler (100 g)
		Base monomer (vol%)	Filler (vol%)	
$M_{80}F_{VX\text{-}20}$	VX-X	80	20	3.25
$M_{70}F_{VX\text{-}30}$		70	30	
$M_{60}F_{VX\text{-}40}$		60	40	
$M_{80}F_{VR\text{-}20}$	VX-SR	80	20	2.25
$M_{70}F_{VR\text{-}30}$		70	30	
$M_{60}F_{VR\text{-}40}$		60	40	
$M_{80}F_{E1\text{-}20}$	EMB-10	80	20	0.24
$M_{70}F_{E1\text{-}30}$		70	30	
$M_{60}F_{E1\text{-}40}$		60	40	
$M_{80}F_{E2\text{-}20}$	EMB-20	80	20	0.16
$M_{70}F_{E2\text{-}30}$		70	30	
$M_{60}F_{E2\text{-}40}$		60	40	
$M_{100}F_0$	—	100	0	—

as a base monomer[14]. In addition, a photoinitiator (camphorquinone, Wako Pure Chemical Industries Ltd.) and a co-initiator (N,N-dimethylaminoethyl methacrylate, Tokyo Chemical Industry Co. Ltd., Tokyo, Japan) equivalent to 1.0 wt% of the base monomer were added.

Four different filler types used in this study are listed together with their filler properties in Table 1. Two of the fillers (VX-X and VX-SR, Tatsumori, Tokyo, Japan) consisted of irregular-shaped quartz glass particles, while the other two fillers (EMB-10 and EMB-20, Toshiba-Ballotini Co. Ltd., Tokyo, Japan) consisted of spherical-shaped borosilicate glass particles. The refractive indexes of the fillers were 1.46 and 1.48 with reference wavelength held at 589 nm of wavelength. Surfaces of the fillers were treated with γ-MPTS (γ-methacryloxy propyl-trimethoxy silane, Shin-Etsu Chemical Co. Ltd., Tokyo, Japan), where the corresponding amount of γ-MPTS for each filler[14] are shown in Table 2. Both filler and γ-MPTS corresponding to the filler were then added to a 95% ethanol aqueous solution, and mixed using a magnetic stirrer and an ultrasonic washing machine. After mixing was completed, ethanol was evaporated from the slurry and the filler dried at 130℃.

For each of the four fillers, specimens with three different filler volume contents of 20, 30, and 40 vol% (Table 2) were prepared by adding the calculated quantity of filler to the base monomer and then blending sufficiently by hand. The reason for a maximum filler content of 40 vol% was that no more than 40 vol% could be mixed with the base monomer before wettability; otherwise, translucency problems would occur. Test specimens were prepared by inserting resin paste into a Teflon cylindrical mold (12ϕ×1.0 mm) open at both ends and placed on a glass slide. The resin paste was slightly overfilled and covered by a thin glass plate (0.15 mm thick), and a glass slide was placed over the thin glass plate. Finger pressure was exerted on this plate to extrude excess resin. The top glass plate was then removed, and a light source unit with a tungsten-halogen lamp (Luxor model 4000, 200 mW/cm^2, ICI PLC, Macclesfield, UK) irradiated through the thin glass plate for 180 seconds. Once the curing process was completed, specimens were removed from the mold and carefully checked to be free of scratches, impurities, porosities, and other defects that could cause irregular optical behavior.

Filler size, specific density, and specific surface area

Filler properties—namely, filler particle size, specific density, and specific surface area—of the fillers were measured. Average filler particle size was determined with a centrifugal particle size analyzer (SA-CP2, Shimadzu Corp., Kyoto, Japan). Specific density was determined with an air comparison pyknometer (Model 930, Beckman-Toshiba Ltd., Tokyo, Japan). Specific surface area was also determined with a surface area analyzer (Model 4200, Nikkiso Co. Ltd., Tokyo, Japan) using a mixed gas of nitrogen and helium in a proportion of 3:7. Filler particles were kept in a vacuum oven for 40 minutes at 150°C and then stored in a desiccator before the tests. For each of the four fillers, three measurements were conducted and used to determine the respective mean values.

Light transmittance characteristics

Light transmittance and its wavelength distribution were measured with a spectral transmittance meter with an integrating sphere (TM-1, Topcon Co., Tokyo, Japan)[15]. Measurements were recorded at 5-nm intervals from 380 nm to 700 nm of wavelength, which included most wavelengths detected by the human eye. Ratio of intensity of the incident light and the transmitted light passing through the specimen was calculated as the light transmittance (%) of the specimen at each wavelength. To evaluate the effect of base resin on light transmittance spectra, base resin without filler was also measured.

Light diffusion characteristics

Angular intensity distribution of transmitted light was measured using a gonio-photometer (GP-1C, Optec Co., Tokyo, Japan)[7]. Light diffusion characteristics are thought to mirror any optical scattering which occurs within a material[16]. Intensity of the collimated incident light and diffused light in the material were measured using an optical power meter (TQ82017, Advantest Co., Tokyo, Japan) rotated around the fixed specimen at different angles to the specimen's surface. Light transmittance (%) was recorded at 5° intervals from a diffusion angle of -70° to +70°.

Color

Color values of the specimen on the CIE L*a*b* color system were measured using a spectral transmittance meter with the standard illuminant D_{65}. The L* value represents lightness, and a* and b* are chromatic coordinates of the red-green and yellow-blue axes respectively[17]. In the apparatus, the spectrum of light transmitted through the specimen was categorized into the spectral colors. From the color values, the color difference ΔE^* for the 30 and 40 vol% filler content specimens was compared with the minimum 20 vol% specimen. It was calculated using the following equation:

$$\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$$

where ΔL^*, Δa^*, and Δb^* are the changes in L*, a*, and b* between each filler content and 20 vol%.

Measured properties among the materials were statistically compared using Student's t-test at a significance level of $p=0.05$. The influence of filler properties on the differences in color values was analyzed by three-way analysis of variance (ANOVA). Correlation between color differences and filler shape was analyzed using linear regression analysis.

RESULTS

Filler size, specific density, and specific surface area

The results are shown in Table 1. Irregular-shaped fillers had markedly larger values of specific surface area than the two spherical-shaped fillers.

Light transmittance characteristics

Light transmittance wavelength distribution spectra of the materials containing four filler types with filler content of 40 vol% in the 380-700 nm wavelength region are shown in Fig. 1. There were significant differences in spectral distribution among the materials. All materials showed a distinct absorption peak in the wavelength range of 440 to 550 nm. Base resin without filler transmitted more than 90% of incident light, except at the absorption peak wavelength. Figure 2 shows the values of overall light transmittance for all materials. Overall light transmittance was calculated as the average of overall values measured from 380 to 700 nm. For all materials, the overall light transmittance decreased with increased filler content.

Light diffusion characteristics

Angular intensity distributions of the transmitted light of materials containing four filler types with filler content of 40 vol% as a function of diffusion angle are shown in Fig. 3. In this figure, relative transmittance was the ratio of light transmittance value at each diffusion angle to the value at 0°. A material with broader distribution suggests that more light is scattered within the material than that with sharper distribution. VX-X and -SR materials showed sharper distribution than the other two fill-

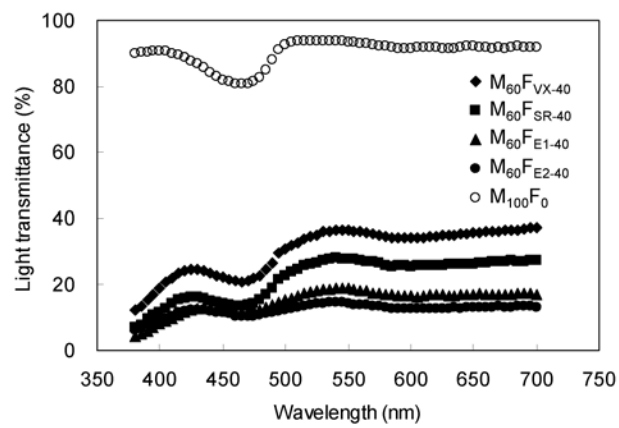

Fig. 1 Spectral distributions of transmitted light with wavelength from 380 nm to 700 nm of the materials without and with four types of filler in filler content of 40 vol%.

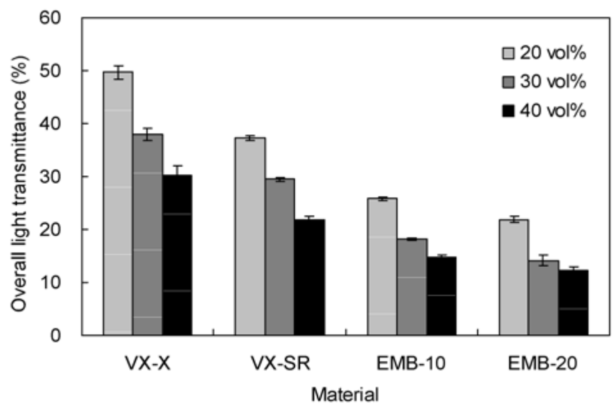

Fig. 2 Values of overall light transmittance from 380 to 700 nm for materials containing four types of filler in filler contents of 20, 30, and 40 vol%.

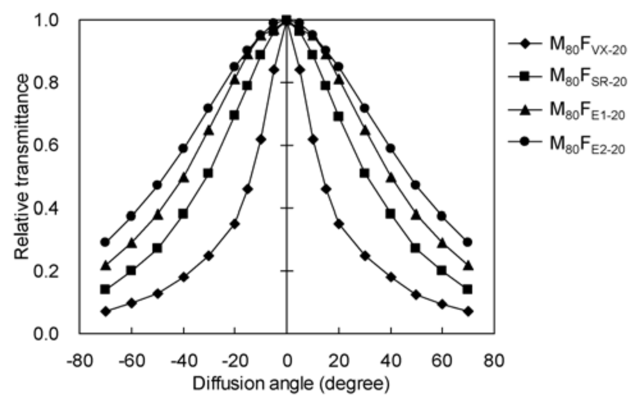

Fig. 3 Angular intensity distributions of transmitted light as a function of diffusion angle of materials containing four types of filler in filler content of 20 vol%.

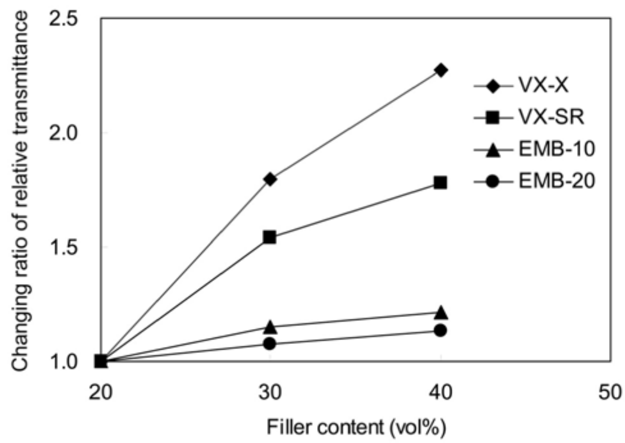

Fig. 4 Variations of changing ratio of overall relative transmittance for materials containing four types of filler in filler contents of 30 and 40 vol% compared with those in 20 vol%.

ers. To consider the effect of filler content on angular distribution of diffuse light, the changing ratios of overall relative transmittance—which were the ratios of the values at filler contents of 30 and 40 vol% to the values at 20 vol%—were calculated. Overall relative transmittance was calculated as the average of overall values from -70° to +70°. The results are shown in Fig. 4. For all filler types, the scattered light increased with increased filler content. Values of changing ratio of overall relative transmittance at 40 vol% filler content ranged from 1.14 for EMB-20 to 2.27 for VX-X. VX-X and -SR materials showed higher changing ratios with increasing filler content, whereas for EMB-10 and-20, the values were relatively small.

Color

Changes in color values of Δa^* and Δb^*—which reflect the chromatic components of color difference—for all materials with 30 vol% and 40 vol% of filler content compared with those of 20 vol% are shown in Table 3. ANOVA showed that filler particle size (a^*: F=233.0; b^*: F=125.6), shape (a^*: F=247.5; b^*: F=212.5), and filler content (a^*: F=38.3; b^*: F=23.2) statistically influenced the color values (p=0.0001). Values of color difference ΔE^* calculated are also shown in Fig. 5. VX-X and -SR materials with irregular shape showed significantly higher values of color difference than those with spherical shape.

DISCUSSION

The optical properties of a composite resin consisting of two different transparent base monomers and small filler particles is characterized by the differences in optical properties between the resin matrix and filler particles. In particular, the difference in refractive index may be one critical factor that determines the optical properties of the composite

Table 3 Values of Δa* and Δb* for all materials of 30 and 40 vol% filler contents *versus* those of 20 vol% filler content. All values are given as mean±SD.

Filler	Filler content (vol%)	Δa*	Δb*
VX-X	30	0.2±0.25	5.1±0.53
VX-X	40	-2.2±0.38	12.0±1.04
VX-SR	30	-1.9±0.21	4.5±0.60
VX-SR	40	-2.2±0.46	8.6±0.52
EMB-10	30	0.9±0.16	-2.4±0.14
EMB-10	40	4.9±0.02	-12.2±0.42
EMB-20	30	1.0±0.17	-1.7±0.35
EMB-20	40	4.0±0.34	-10.3±0.17

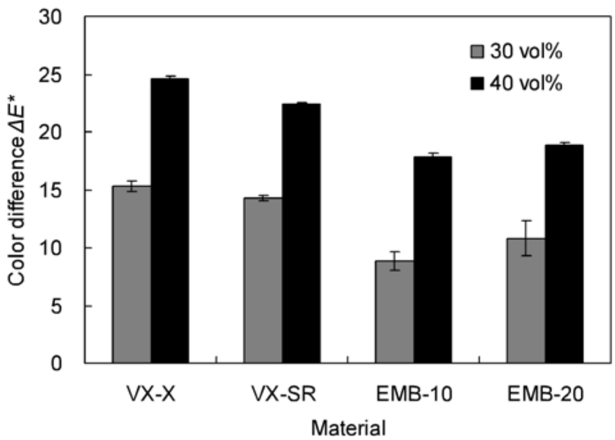

Fig. 5 Values of color difference ΔE* of materials containing four types of filler in filler contents of 30 and 40 vol% compared with those in 20 vol%.

resin[18,19]. Although the refractive indexes of fillers used in this study were not the same, the differences in their refractive indexes were considerably small. Therefore, it seemed that differences in optical properties and color appearance among materials were expected to be dominated by filler properties such as particle size, shape, and filler content[9].

In the light transmittance spectra, there were significant differences in wavelength distribution characteristics among the materials (Fig. 1). The average light transmittance of the base resin without filler was more than 90% from 380 nm to 700 nm, whereas for materials containing 40 vol% filler light transmittance ranged between 12.5 and 30.2%. This indicated that incorporation of small filler particles caused significant reduction in light transmitted through the composite material. As for the cause, it could be due to diffuse light transmission by multiple scattering of light within the material as shown by the angular distribution of diffuse light for each material (Fig. 3).

Moreover, for all materials, there was a trend for light transmittance to decrease as the wavelength decreased, especially at short-wavelength range below 520 nm. Light scattering increased for incident light with shorter wavelengths, whereas light with longer wavelengths transmitted more easily through the material[3,20]. This wavelength dependence of light scattering was especially acute for cases where the size of the scattering particles was near the wavelength of incident light[21]. The materials containing smaller and irregular-shaped fillers, VX-X and -SR, showed stronger wavelength dependence of light transmittance as compared with those containing larger and spherical-shaped fillers. This result suggested that differences in the size and shape of fillers incorporated in the materials might lead to significant differences in the distribution spectrum of transmitted light among the materials.

For all materials, a broad and wide absorption peak was also observed in the wavelength range between 430 nm and 550 nm with a peak at around 470 nm. Absorption peak was also observed for the base resin without filler. Other studies reported that the camphorquinone photoinitiator has an absorption peak at about 468 nm, and no other absorption bands were present in either the resin or filler particles within the spectrum of interest[9,22]. Therefore, the specific absorption peak observed for all materials was caused by the camphorquinone photoinitiator, and not attributable to filler properties.

The VX-X material with the smallest filler particle size showed the highest values of overall light transmittance for all filler contents, whereas those with the larger-sized fillers showed lower light transmittance for all filler contents (Fig. 2). This result agreed with another study[9] which investigated the light attenuation characteristics of composite

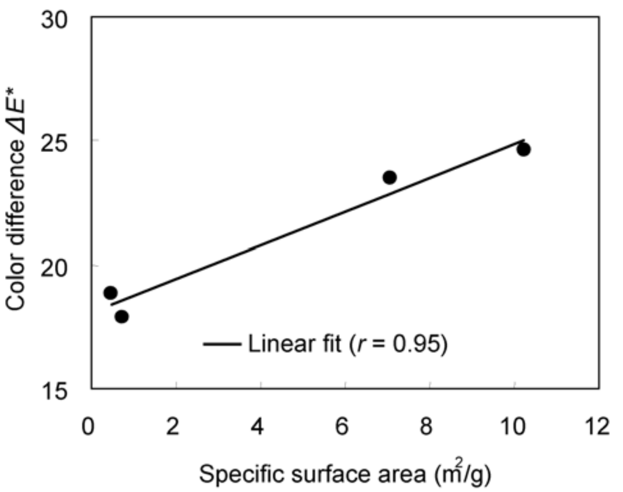

Fig. 6 Relationship between specific surface area of four filler types and color difference of materials in filler content of 40 vol% compared with those in 20 vol%.

resins with different filler particle sizes, which were almost the same as or smaller than the fillers used in this study. On the other hand, materials with smaller filler particle size showed sharper angular distribution of diffuse light (Fig. 3), indicating that less light was scattered within the material. As light scattering is expected to increase with increasing filler particle diameter[9,23], the larger scattering caused by larger fillers thus resulted in higher transmittance loss in comparison with materials containing smaller filler particles.

At this juncture, it has been established that filler content and filler particle size significantly affected the optical properties, and hence the color, of composite resins. In addition, it was found in this study that the shape of filler particles also significantly influenced the color rendition. Between materials with irregular- and spherical-shaped fillers, there were significant differences in a* and b* values, which describe the chromatic components of color difference (Table 3). With irregular-shaped fillers of VX-X and -SR, the a* value decreased and b* value increased with increasing filler content. With spherical-shaped fillers of EMB-10 and -20, the a* value increased and b* value decreased. These results suggested that the color of materials containing irregular-shaped fillers shifted more to green and yellow on the red-green and yellow-blue axes, while those containing spherical-shaped fillers shifted more to red and blue respectively.

The difference in color-changing behavior may be predominantly due to the difference in wavelength distribution characteristics of the light transmittance of the materials (Fig. 1). A lower light transmittance at shorter wavelengths and a higher transmittance at longer wavelengths for the materials containing irregular-shaped fillers resulted in shifting the color to more green and yellow, while a comparatively flat distribution of transmittance for the given wavelength range caused an opposite color shift in materials containing spherical-shaped fillers. These results thus indicated that differences in light transmittance characteristics caused by filler properties played a pivotal role in the color of composite resins.

Similarly, the color differences obtained in this study showed the same trend as that of the chromatic coordinates, with a significant difference in color difference ΔE^* between the different filler shapes (Fig. 5). Figure 6 shows the relationship between the color difference for materials with four different filler types in 40 vol% filler content compared with those of 20 vol% filler content, *versus* the respective specific surface area. The two measured properties were well correlated. A linear regression analysis gave a correlation coefficient of 0.95, which was significant ($p<0.05$). In general, the specific surface area of filler is highly related to the shape of filler particles, *i.e.*, filler with irregular shape and rougher surface has larger surface area than that with spherical shape and smoother surface. In this study, it was found that the surface area of filler particles also affected the light scattering behavior of the material. The VX-X and -SR materials with larger surface area showed significantly larger changes in light scattering behavior with increasing filler content than the other two fillers (Fig. 4). It should be noted that difference in light scattering behavior caused by filler shape would result in different color renditions among the materials[21].

The color of a composite material is a complex combination of optical behaviors within the material. To date, neither theoretical nor experimental treatment on this subject is known[24], which means that more investigation concerning the accurate mechanism of color changes caused by filler properties is much needed in future studies. Within the limitations of the present study, it was found that filler shape strongly affected the color of composite resins, and other filler properties such as filler particle size and filler content exerted significant influences too. On this note, filler particle size and filler content must be harnessed and leveraged to achieve optimum mechanical and physical properties. Finally, in the context of aesthetics properties, it certainly seems beneficial and expedient to improve the color appearance of composite resins by focusing on filler shape.

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