

Original

The influence of polymerization conditions on color stability of three indirect composite materials

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Abstract: The aim of the present study was to evaluate color change and color stability against aqueous media of three indirect composite materials (Epicord, Estenia, and Twiny) that were polymerized with different systems. Disk-shaped specimens were prepared with their proprietary polymerization systems or with a metal halide light polymerization unit (Twinkle X). The specimens were then immersed in water or tea. Change in color from baseline (24 h) to 4 weeks was determined with a chromameter (ShadeEye NCC) using a white background. The L^* , a^* , and b^* values for both states were determined, and ΔE^*_{ab} values were calculated. The Estenia specimens immersed in tea had significantly lower ΔL^* (-7.0 to -5.6) and significantly higher ΔE^*_{ab} values (6.5 to 8.6) than did the Epicord and Twiny specimens, under all polymerization conditions. The results indicate that after tea immersion the Estenia material was less stable against color change than were the other two materials. The Twinkle X metal halide unit was suitable for polymerization of the three composite materials, using an exposure period of 60 s or longer. (J Oral Sci 55, 51-55, 2013)

Keywords: color; composite; heat; light; polymerization.

Introduction

Use of tooth-colored composite materials for restorations and partial denture veneers has increased substantially, probably due to improvements in the properties of the materials. However, several concerns have been reported. One such concern is color stability. A number of studies have reported changes in the color of composites (1-10). In addition, the polymeric matrix in composite materials is stained by beverages, food, and drugs (11-15).

A high-intensity light-polymerization apparatus has been developed (16), and use of this apparatus in combination with metal halide light sources was found to considerably improve the mechanical properties (17) and wear resistance (18) of composite materials. Although the stability of restorative materials against color change has been extensively studied, there is limited information on the color stability of indirect composite materials polymerized with different devices (19). The aim of the current project was to evaluate the influence of different varying polymerization conditions on the color stability of three indirect composite materials.

Materials and Methods

Indirect composite materials and polymerization apparatus

Three indirect composite materials designed for restoration and partial denture veneers were assessed (Table 1). Epicord (Kuraray Medical Inc., Tokyo, Japan) is composed of urethane tetramethacrylate (UTMA), triethyleneglycol dimethacrylate (TEGDMA), and 76.0 wt% filler. Estenia C&B (Kuraray Medical Inc.) is composed

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Table 1 Indirect composite materials assessed

Trade name	Manufacturer	Lot number	Composition (wt%)
Epicord E2	Kuraray Medical Inc., Tokyo, Japan	0065CA 0066AA	UTMA, TEGDMA, <i>dl</i> -camphorquinone, pigment, 76.0% filler (prepolymerized organic filler, silanated glass filler, silanated colloidal silica), others
Estenia C&B E2	Kuraray Medical Inc.	0037BA 0037BB	UTMA, methacrylate, <i>dl</i> -camphorquinone, pigment, 87.9% filler (surface-treated alumina microfiller, silanated glass ceramic filler), others
Twiny E2	Yamamoto Precious Metal Co., Ltd., Osaka, Japan	1031128	UTMA, TEGDMA, 79.0% filler, pigment, others

UTMA, urethane tetramethacrylate; TEGDMA, triethyleneglycol dimethacrylate

Table 2 Polymerization apparatus employed

Trade name	Manufacturer	Source	Polymerization condition
α -Light II	J. Morita Corp., Suita, Japan	Halogen lamp: 360 W \times 1, 400-600 nm*; Fluorescent tubes: 27 W \times 2, 400-550 nm*	180 s
KL-310	J. Morita Corp.	Heat oven	110°C, 15 min
Twinkle X	Toho Dental Products, Saitama, Japan	Metal halide lamps, 150 W \times 2, 250-600 nm**	30 s, 60 s, and 90 s

*Reference 18; **Reference 16.

of UTMA, other methacrylate, and 87.9 wt% filler. Twiny (Yamamoto Precious Metal Co., Ltd., Osaka, Japan) is composed of UTMA, TEGDMA, and 79.0 wt% filler.

Three polymerization devices were used (Table 2). The α -Light II (J. Morita Corp., Suita, Japan) unit is equipped with a halogen lamp and two fluorescent tubes (18). The KL-310 (J. Morita Corp.) apparatus is a box-type heat oven. The Twinkle X (Toho Dental Products, Saitama, Japan) unit is equipped with two high-intensity discharge metal halide lamps (16).

Specimen preparation

The composite materials were filled into steel molds (15 mm in diameter by 2 mm in height) placed on a glass plate (thickness, 1.3 mm; Micro Slide Glass, Matsunami Glass Ind., Ltd., Osaka, Japan). The composite material was filled into the mold, and the surface was covered with the 1.3-mm glass plate. The specimen was then polymerized with its proprietary system or with the Twinkle X unit for 30-90 s (Table 2). The proprietary system for the Epicord material was light exposed for 180 s with the α -Light II unit, and the polymerization system for the Estenia C&B and Twiny materials was light exposed with the α -Light II unit for 180 s, followed by heating with the KL-310 oven for 15 min at 110°C. After polymerization, both surfaces of each specimen were ground with silicon-carbide (SiC) abrasive papers (#800-#2,000, WetorDry Tri-M-ite, 3M Corp., St. Paul, MN, USA) under running water. The ground specimens were then polished with felt (TexMet 1500, Buehler Ltd., Lake Bluff, IL, USA) and diamond slurry (3, 1, and 0.25 μ m, MetaDi, Buehler Ltd.). The specimen thickness was adjusted to 1.0 mm using a digital caliper (Mitutoyo Corp., Kawasaki, Japan). The prepared specimens were

then placed in an incubator at 37°C for 24 h in a dry state.

Color determination and calculation of color difference

After 24-h storage, specimens were immersed in purified water or tea for 4 weeks (28 days). The tea solution was prepared by placing three tea bags (15 g; Day & Day Teabags, Mitsui Norin Co., Ltd., Tokyo, Japan) in 1.0 L of boiling water and brewing for 5 min (19). The color values (L^* , a^* , and b^*) of specimens were determined with a chromameter (ShadeEye NCC, Shofu Inc., Kyoto, Japan) using a white background (Konica Minolta Holdings Inc., Osaka, Japan). Mean color values of the white background were $L^* = 97.3$, $a^* = 0.1$, and $b^* = -1.3$.

The color of the dry-state specimens was measured 24 h after preparation, without immersion in solution. Immersed specimens (4 weeks) were rinsed with water and dried with absorbent paper. Color was measured according to the Commission Internationale de l'Eclairage (CIE) 1976 $L^*a^*b^*$ color space system relative to the standard illuminant D_{65} and observer functions (2° visual field). The system was calibrated at the start of each measurement with a white calibration tile provided by the manufacturer. The aperture of the chromameter was placed on the specimen surface. Color measurements were performed six times around the center of each specimen, and mean values were calculated. Nine specimens were tested for each material and condition. The color difference between pre- and post-immersion values (ΔE^*_{ab}) was determined for each specimen. The following equation was used to calculate ΔE^*_{ab} values:

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

Table 3 ΔL^* values for white background (4 weeks)

Polymerization mode	Water				Tea			
	Estenia	Epicord	Twiny	K-W test	Estenia	Epicord	Twiny	K-W test
Proprietary system	-0.8 (0.4)*	ab -0.3 (1.2)**	a 0 (0.3)**	ab Significant	-6.3 (0.4)*	ab -3.0 (1.1)**	a -2.0 (0.5)**	a Significant
Twinkle X 30 s	-1.2 (0.4)*,**	a -1.6 (0.7)*	b -0.8 (0.4)**	a Significant	-7.0 (0.4)*	a -2.7 (0.5)**	a -4.8 (1.5)**	b Significant
Twinkle X 60 s	-0.6 (0.6)*	ab -0.8 (0.8)*	ab 0.1 (0.8)*	b Not significant	-6.0 (0.9)*	ab -2.7 (1.7)**	a -3.0 (0.9)**	ab Significant
Twinkle X 90 s	-0.5 (0.3)*	b -0.6 (1.0)*	ab 0.2 (0.5)**	b Significant	-5.6 (0.7)*	b -2.8 (0.8)**	a -2.9 (0.6)**	ab Significant

$n = 9$. Means (SD). Identical letters indicate absence of a significant difference among polymerization modes (Dunn's test, $P > 0.05$).

K-W test, Kruskal-Wallis test. K-W test and Dunn's test, difference between Estenia, Epicord, and Twiny (significant; $P < 0.05$). Identical symbols indicate absence of a significant difference.

Table 4 Δa^* values for white background (4 weeks)

Polymerization mode	Water				Tea			
	Estenia	Epicord	Twiny	K-W test	Estenia	Epicord	Twiny	K-W test
Proprietary system	-0.1 (0.3)*	a 0 (0.2)*	a 0 (0.1)*	a Not significant	1.6 (0.2)*	a 1.2 (0.2)*,**	a 0.6 (0.2)**	a Significant
Twinkle X 30 s	0.7 (0.1)*	bc -0.5 (0.3)**	b 0.5 (0.1)***	bc Significant	2.3 (0.2)*	b 1.0 (0.3)**	ac 1.2 (0.4)**	b Significant
Twinkle X 60 s	0.6 (0.1)*	ac -0.1 (0.2)**	ac 0.2 (0.1)**	ac Significant	2.0 (0.2)*	bc 0.5 (0.5)**	bc 0.9 (0.2)**	ab Significant
Twinkle X 90 s	0.7 (0.1)*	bc -0.4 (0.2)**	bc 0.2 (0.1)***	a Significant	1.9 (0.2)*	ac 0.4 (0.3)**	b 0.7 (0.3)**	a Significant

$n = 9$. Means (SD). Identical letters indicate absence of a significant difference among polymerization modes (Dunn's test, $P > 0.05$).

K-W test, Kruskal-Wallis test. K-W test and Dunn's test, difference between Estenia, Epicord, and Twiny (significant; $P < 0.05$). Identical symbols indicate absence of a significant difference.

Table 5 Δb^* values for white background (4 weeks)

Polymerization mode	Water				Tea			
	Estenia	Epicord	Twiny	K-W test	Estenia	Epicord	Twiny	K-W test
Proprietary system	-1.0 (0.2)*	a 1.1 (0.3)**	ab -1.1 (0.2)*	a Significant	3.8 (0.4)*	ab 1.2 (0.3)**	a -0.6 (0.2)***	a Significant
Twinkle X 30 s	-0.3 (0.2)*	b -1.4 (0.1)**	b -2.5 (0.1)***	b Significant	4.4 (0.4)*	a 1.3 (0.2)**	a 1.9 (2.0)**	b Significant
Twinkle X 60 s	-0.5 (0.2)*	b 1.4 (0.3)**	ac -1.2 (0.2)***	a Significant	3.1 (0.4)*	b 1.7 (0.4)**	ac -0.7 (0.6)**	a Significant
Twinkle X 90 s	-1.0 (0.1)*	a 2.2 (0.3)**	c -0.7 (0.3)*	a Significant	2.7 (0.4)*	bc 2.8 (0.2)*	bc 0.2 (0.7)**	ab Significant

$n = 9$. Means (SD). Identical letters indicate absence of a significant difference among polymerization modes (Dunn's test, $P > 0.05$).

K-W test, Kruskal-Wallis test. K-W test and Dunn's test, difference between Estenia, Epicord, and Twiny (significant; $P < 0.05$). Identical symbols indicate absence of a significant difference.

Table 6 ΔE^*_{ab} values for white background (4 weeks)

Polymerization mode	Water				Tea			
	Estenia	Epicord	Twiny	K-W test	Estenia	Epicord	Twiny	K-W test
Proprietary system	1.4 (0.4)*	ab 1.5 (0.7)*	a 1.2 (0.3)*	a Not significant	7.6 (0.5)*	ab 3.5 (0.8)**	a 2.2 (0.5)**	a Significant
Twinkle X 30 s	1.5 (0.3)*	a 2.3 (0.5)**	ab 2.7 (0.2)**	bc Significant	8.6 (0.4)*	a 3.2 (0.5)**	a 5.6 (1.6)**	b Significant
Twinkle X 60 s	1.1 (0.3)*	b 1.8 (0.5)**	ab 1.4 (0.3)***	ac Significant	7.1 (0.7)*	b 3.4 (1.4)**	a 3.3 (0.9)**	ab Significant
Twinkle X 90 s	1.4 (0.1)*	ab 2.5 (0.4)**	b 0.9 (0.3)*	a Significant	6.5 (0.7)*	b 4.0 (0.5)**	a 3.1 (0.6)**	ab Significant

$n = 9$. Means (SD). Identical letters indicate absence of a significant difference among polymerization modes (Dunn's test, $P > 0.05$).

K-W test, Kruskal-Wallis test. K-W test and Dunn's test, difference between Estenia, Epicord, and Twiny (significant; $P < 0.05$). Identical symbols indicate absence of a significant difference.

Statistical analysis

The ΔL^* , Δa^* , Δb^* , and ΔE^*_{ab} values were analyzed by the Kolmogorov-Smirnov test for normality of distribution (SPSS 15.0, SPSS Inc., Chicago, IL, USA). When the results of the Kolmogorov-Smirnov test did not show normality of distribution, the Kruskal-Wallis test (SPSS 15.0) and Dunn's comparison test (GraphPad Prism 5, GraphPad Software Inc., La Jolla, CA, USA) were performed. The value for statistical significance was set at $\alpha = 0.05$.

Results

The Kolmogorov-Smirnov test showed that some categories in several groups did not have normal distributions. The results were therefore analyzed with the Kruskal-Wallis test and Dunn's comparison test. The mean (SD) of ΔL^* , Δa^* , Δb^* , and ΔE^*_{ab} are shown in Tables 3 through 6.

Table 3 summarizes mean ΔL^* values. The ΔL^* of specimens immersed in water ranged from -1.2 to -0.5 for Estenia, -1.6 to -0.3 for Epicord, and -0.8 to 0.2 for Twiny. For specimens immersed in tea, ΔL^* ranged from

-7.0 to -5.6 for Estenia, -3.0 to -2.7 for Epricord, and -4.8 to -2.0 for Twiny. The Estenia specimens immersed in tea had a significantly lower ΔL^* than the Epricord and Twiny specimens under all polymerization conditions.

Table 4 shows Δa^* values. The Δa^* of specimens immersed in water ranged from -0.1 to 0.7 for Estenia, -0.5 to 0 for Epricord, and 0 to 0.5 for Twiny. For specimens immersed in tea, Δa^* ranged from 1.6 to 2.3 for Estenia, 0.4 to 1.2 for Epricord, and 0.6 to 1.2 for Twiny.

Mean Δb^* values are shown in Table 5. The Δb^* of specimens immersed in water ranged from -1.0 to -0.3 for Estenia, -1.4 to 2.2 for Epricord, and -2.5 to -0.7 for Twiny. For specimens immersed in tea, Δb^* ranged from 2.7 to 4.4 for Estenia, 1.2 to 2.8 for Epricord, and -0.7 to 1.9 for Twiny. The Δb^* of specimens polymerized with proprietary systems and immersed in tea was ranked in ascending order as Twiny (-0.6), Epricord (1.2), and Estenia (3.8), and the trend was statistically significant.

Mean ΔE^*_{ab} values are summarized in Table 6. The ΔE^*_{ab} of specimens immersed in water ranged from 1.1 to 1.5 for Estenia, 1.5 to 2.5 for Epricord, and 0.9 to 2.7 for Twiny. For specimens immersed in tea, ΔE^*_{ab} ranged from 6.5 to 8.6 for Estenia, 3.2 to 4.0 for Epricord, and 2.2 to 5.6 for Twiny. Estenia specimens immersed in tea had significantly higher ΔE^*_{ab} value than the Epricord and Twiny specimens for all polymerization conditions.

Discussion

This study evaluated color change after immersion in water or tea in three indirect composite materials polymerized with different systems. A number of storage media have been proposed, including coffee (10-13,20,21), cola (11,13), juice (13,15), red wine (11-13,20,21), soy sauce (15), and tea (11,12,20,21). This study employed tea as a coloring solution because it is relatively homogeneous as a coloring solution and because the color of tea is stable during storage.

L^* represents the brightness of materials. Table 3 shows that change in L^* did not significantly differ after immersion in water among the three materials. However, L^* was considerably lower after immersion in tea, especially for the Estenia material. The authors consider that the reduction in L^* after tea immersion was due to the coloring ingredients in tea. The results are in agreement with those of other studies that used tea as a storage medium (11,12,19). Also, the greater reduction in L^* for the Estenia material may be due to penetration of coloring ingredients into the interface between the matrix polymer and inorganic filler or into microvoids originally present in the Estenia material. The results shown in Table 1 show that the polymerization performance of the proprietary

systems and the Twinkle X unit did not particularly differ and that light exposure of longer than 60 s was acceptable for all materials treated with the Twinkle X unit.

The Δa^* value of the three composite materials was not greatly affected by water storage and increased after storage in tea. In the CIE 1976 $L^*a^*b^*$ color space system, an increase in a^* represents increased red, and a decrease in a^* represents increased green. The results of the present study show that redness increased in all materials, especially for Estenia, after tea immersion, regardless of polymerization system. Similar results were seen in the b^* values of the specimens, probably because yellowish color change was accelerated and bluish color change was suppressed by the original color of the tea solution.

According to the abovementioned equation, ΔE^*_{ab} represents total color change in specimens. The results for ΔE^*_{ab} (Table 6) indicate that light exposure for 30 s with the Twinkle X unit was insufficient for polymerization of Twiny. Also, Estenia was less stable against color change than were the other two materials after tea immersion, regardless of polymerization conditions. The results are likely due to the somewhat rougher surface characteristics of Estenia, which is a highly loaded composite material (18).

Dental technicians desire composite materials that can be simply and quickly polymerized. The Twinkle X unit, with a light exposure period greater than 60 s, had better polymerization performance than the proprietary systems of the three investigated materials. Application of high-intensity polymerization apparatus is recommended to improve stability against color change of indirect composite materials.

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References

1. Mutlu-Sagesen L, Ergün G, Ozkan Y, Semiz M (2005) Color stability of a dental composite after immersion in various media. *Dent Mater J* 24, 382-390.
2. Arikawa H, Kanie T, Fujii K, Takahashi H, Ban S (2007) Effect of filler properties in composite resins on light transmittance characteristics and color. *Dent Mater J* 26, 38-44.
3. Fontes ST, Fernández MR, de Moura CM, Meireles SS (2009) Color stability of a nanofill composite: effect of different

- immersion media. *J Appl Oral Sci* 17, 388-391.
4. Cruvinel DR, Garcia Lda F, Consani S, de Carvalho Panzeri Pires-de-Souza F (2010) Composites associated with pulp-protection material: color-stability analysis after accelerated artificial aging. *Eur J Dent* 4, 6-11.
 5. Mundim FM, Garcia Lda F, Pires-de-Souza Fde C (2010) Effect of staining solutions and repolishing on color stability of direct composites. *J Appl Oral Sci* 18, 249-254.
 6. Papadopoulos T, Sarafianou A, Hatzikyriakos A (2010) Colour stability of veneering composites after accelerated aging. *Eur J Dent* 4, 137-142.
 7. Lee YK, Yu B, Lim HN, Lim JI (2011) Difference in the color stability of direct and indirect resin composites. *J Appl Oral Sci* 19, 154-160.
 8. Domingos PA, Garcia PP, de Oliveira AL, Palma-Dibb RG (2011) Composite resin color stability: influence of light sources and immersion media. *J Appl Oral Sci* 19, 204-211.
 9. Güler AU, Duran I, Yücel AÇ, Ozkan P (2011) Effects of air-polishing powders on color stability of composite resins. *J Appl Oral Sci* 19, 505-510.
 10. Schmitt VL, Puppini-Rontani RM, Naufel FS, Nahsan FP, Alexandre Coelho Sinhorette M, Baseggio W (2011) Effect of the polishing procedures on color stability and surface roughness of composite resins. *ISRN Dent* 2011:617672.
 11. Ertaş E, Güler AU, Yücel AC, Köprülü H, Güler E (2006) Color stability of resin composites after immersion in different drinks. *Dent Mater J* 25, 371-376.
 12. Omata Y, Uno S, Nakaoki Y, Tanaka T, Sano H, Yoshida S et al. (2006) Staining of hybrid composites with coffee, oolong tea, or red wine. *Dent Mater J* 25, 125-131.
 13. Topcu FT, Sahinkesen G, Yamanel K, Erdemir U, Oktay EA, Ersahan S (2009) Influence of different drinks on the colour stability of dental resin composites. *Eur J Dent* 3, 50-56.
 14. Galvão AP, Jacques LB, Dantas L, Mathias P, Mallmann A (2010) Effect of lipstick on composite resin color at different application times. *J Appl Oral Sci* 18, 566-571.
 15. Soares-Geraldo D, Scaramucci T, Steagall-Jr W, Braga SR, Sobral MA (2011) Interaction between staining and degradation of a composite resin in contact with colored foods. *Braz Oral Res* 25, 369-375.
 16. Matsumura H, Tanoue N, Atsuta M, Kitazawa S (1997) A metal halide light source for laboratory curing of prosthetic composite materials. *J Dent Res* 76, 688-693.
 17. Satsukawa H, Koizumi H, Tanoue N, Nemoto M, Ogino T, Matsumura H (2005) Properties of an indirect composite material polymerized with two different laboratory polymerizing systems. *Dent Mater J* 24, 377-381.
 18. Hirata M, Koizumi H, Tanoue N, Ogino T, Murakami M, Matsumura H (2011) Influence of laboratory light sources on the wear characteristics of indirect composites. *Dent Mater J* 30, 127-135.
 19. Nakazawa M (2009) Color stability of indirect composite materials polymerized with different polymerization systems. *J Oral Sci* 51, 267-273.
 20. Stawarczyk B, Egli R, Roos M, Ozcan M, Hämmerle CH (2011) The impact of in vitro aging on the mechanical and optical properties of indirect veneering composite resins. *J Prosthet Dent* 106, 386-398.
 21. Gawriolek M, Sikorska E, Ferreira LF, Costa AI, Khmelinskii I, Krawczyk A et al. (2012) Color and luminescence stability of selected dental materials in vitro. *J Prosthodont* 21, 112-122.