## Color Stability of Ten Resin-Based Restorative Materials

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#### ABSTRACT

Despite significant developments to improve the optical properties of composites, color stability remains a challenge with changes still observed immediately after polymerization and after some time of storage.

*Objective:* This study aimed to evaluate the color change of ten commercially available resin composite systems immediately after polymerization, at 24 hours, and at 1 month of water storage.

*Materials and Methods:* Five discs of two thicknesses (1 mm/3 mm) in two shades (A3/Bleach) were made from ten commercially available brands of resin composite. Color measurements were recorded with a colorimeter and expressed in terms of the CIE  $L^*a^*b^*$  scale. Color change was calculated between baseline and immediate polymerization ( $\Delta E^*_1$ ), between immediate polymerization and 24 hours ( $\Delta E^*_2$ ), and between 24 hours and 1 month ( $\Delta E^*_3$ ). Values  $\Delta E^* \ge 3.3$  were considered clinically unacceptable as color shifts over this threshold value may be noticeable. The results were analyzed using a three-way analysis of variance (ANOVA) and Student–Newman–Keuls's tests. A significance level of 0.05 was used for all tests.

**Results:** Color change was evidenced for all brands, shades, and thickness. The greatest color change was observed immediately after initial polymerization with  $\Delta E^*$  values ranging from 2.4 to 12.0. Color change after 24 hours and 1 month were significantly less than those observed after polymerization. Immediately after polymerization, only Tetric EvoCeram (Ivoclar-Vivadent, Amherst, NY, USA) showed changes below 3.3. At 24 hours, 20 out of the 40 groups including all materials in both shades and thicknesses showed changes above 3.3. At 1 month, color changes for all brands remained under 3.3.

**Conclusions:** Overall, clinically relevant color changes ( $\Delta E^* \ge 3.3$ ) took place immediately after polymerization of current light-activated composites. These changes were predominantly attributed to shifts in the  $L^*$  and  $b^*$  parameters towards the dark and blue region, respectively. Color shifts continued at 24 hours with changes in the  $L^*$  and  $b^*$  parameters towards the light and yellow region, respectively. After I month, only minor changes under the 3.3 threshold took place.

#### **CLINICAL SIGNIFICANCE**

Ensuring an accurate color match of resin composite restorations to the surrounding tooth structure is a critical aspect of any esthetic restorative procedure. A better understanding of the changes taking place during the different stages of maturation and storage of composites may be of significant benefit to clinicians as this may help minimize shade mismatch issues.

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#### INTRODUCTION

Achieving an optimal color match of resin composite restorations represents a challenge especially in highly poly-chromatic anterior teeth where even small shade discrepancies represent an issue for the patient with high esthetic demands. Visual comparisons using resin colored porcelain or acrylic shade guides are subjective and do not always yield an accurate color match.<sup>1,2</sup> In addition, polymerization-dependent color changes are known to take place and must be taken into consideration during shade selection. Fabrication of a resin color mock-up has been advocated as the standard recommended procedure for shade selection for greater accuracy of color match.<sup>3</sup> The composite is applied to the tooth in the desired thickness and polymerized for 10 seconds after which the shade is selected. Provided that a good initial color match has been obtained, additional changes may occur further compromising the final overall esthetics of the restoration.

The CIE  $L^*a^*b^*$  scale, developed by the International Commission on Illumination (Commission Internationale d'Eclairage), is commonly used to describe color characteristics of an object based on three parameters: lightness–darkness  $(L^*)$ , red–green ( $a^*$ ), yellow-blue ( $b^*$ ).  $L^*$  is achromatic with 0 = black and 100 = white, whereas  $a^*$  and  $b^*$  represent the chromatic coordinates with +a = red, -a = green,+b = yellow, and -b = blue. Similar to an object that can be described by the three dimensions of physical form, length, width, and depth, color can be described in terms of its three coordinates  $L^*$ ,  $a^*$ , and  $b^*$ . Color change is described quantitatively in Delta E ( $\Delta E^*$ ) units, which combines changes in each of the individual parameters  $L^*$ ,  $a^*$ , and  $b^*$  into a single value. This single number represents the "distance" between two colors and it includes changes in each of the individual parameters  $L^*$ ,  $a^*$ , and  $b^*$ . Since the ability to perceive color differences varies from individual to individual, the smallest color difference that the human eye can detect has been the subject of debate. Different  $\Delta E^*$ values have been proposed to determine a "clinically acceptable" color change value, or in other words, a change that may either be unnoticeable or noticeable

but very subtle. Different studies have reported unacceptable color change values at  $\Delta E^* \ge 2,^4$  $\Delta E^* \ge 3.3,^{5-8}$  and  $\Delta E^* \ge 3.7,^{9-11}$  with most available studies applying a value  $\Delta E^* \ge 3.3$  as the threshold for clinical acceptability of color change.

Research efforts have been directed towards modifying the composite's chemical composition in a way that the least amount of color change is experienced. However, despite significant developments to improve the optical properties of composites, color stability remains a challenge with changes observed immediately after polymerization<sup>12,13</sup> and after some time of storage.<sup>14</sup> Discolorations are known to arise from intrinsic or extrinsic factors. Filler and matrix composition,<sup>15–17</sup> photoinitiator,<sup>18,19</sup> wavelength,<sup>12</sup> degree of conversion,<sup>19</sup> shade,<sup>20,21</sup> and water sorption<sup>22</sup> relate to the composition and properties inherent of the material itself. An increased filler content has been reported to yield enhanced color stability.<sup>15,21</sup> Similarly, a higher resin volume fraction has been reported to yield greater discoloration.<sup>5,23</sup> The nature of the matrix is also known to affect the color stability of composites with more hydrophilic monomers resulting in greater water absorption and therefore greater color change,<sup>22</sup> and more hydrophobic monomers resulting in less water sorption and enhanced color stability.<sup>5,22,24</sup> In addition to these intrinsic factors, exposure to different staining agents and accumulation of their products on the surface of teeth has also been shown to cause discolorations over time.24,25

The effect of different extrinsic and intrinsic factors on the color stability of composites requires further study. A better understanding of the changes taking place during polymerization and storage may help minimize shade mismatch issues through a more appropriate management of the color shifts expected to occur. A number of studies have reported on the color stability of resin composite materials after polymerization and storage with changes taking place either to the dark<sup>3,15,26</sup> or light region,<sup>3,27</sup> as well as in the chromatic coordinates of color.<sup>12,15,26,28</sup> Most of these studies describe only a few commercially available products, a single shade or thickness of composite. The authors are not aware of any large comparative studies evaluating

the behavior of newer formulations of nanohybrid and microhybrid composites relative to variables such as thickness, shade, and time of storage. The objective of this study was to evaluate the color change of ten commercially available resin composite systems, Ceram X (Dentsply-Caulk, Milford, DE, USA), Filtek Supreme (3M-ESPE, St. Paul, MN, USA), Four Seasons and Tetric EvoCeram (Ivoclar-Vivadent, Amherst, NY, USA), Point 4 and Premise (Kerr, Orange, CA, USA), Venus (Heraeus-Kulzer, Hanau, Germany), Gradia Direct (GC America, Alsip, IL, USA), Vit-l-Escence (Ultradent, South Jordan, UT, USA), and Artiste (Pentron, Wallingford, CT, USA), immediately after polymerization, at 24 hours, and after 1 month of storage (100% humidity at 37°C). Furthermore, the study aimed to evaluate the effect of shade (A3/Bleach) and thickness (1 mm/3 mm) on the color change.

### MATERIALS AND METHODS

Table 1 summarizes the ten commercially available light-activated resin composites tested in this study. Two thicknesses (1 mm/3 mm) and two shades (A3/Bleach) were tested. Enamel shades were used unless the particular composite system did not discriminate between enamel and dentin shades, as it was the case for Point 4, Venus, and Gradia Direct, for which the only A3 and Bleach shades available were selected instead. Five specimens per study group yielded a total of 220 specimens.

#### Specimen Preparation

The composite was inserted into a white polytetrafluoroethylene (PTFE) mold of 10 mm in diameter and a thickness of either 1 or 3 mm. The composite was condensed against a microscope glass slab with care to avoid internal void formation. A second glass slab was stabilized in contact with the uncured composite and pressed to the thickness of the mold. Glass slabs were used to provide flat specimens of uniform surface that would be less likely to introduce variations in the color measurements.<sup>15</sup>

#### Color Measurements

Color measurements were recorded before polymerization, immediately after polymerization, at 24 hours, and after 1 month of storage (100% humidity at 37°C) with a colorimeter (Minolta Chroma Meter model CR-321/Minolta Corp., Ramsey, NJ, USA). Calibration of the device was performed against a white calibration tile provided by the manufacturer. A measuring area of 3 mm in diameter with 45° circumferential illumination and 0° viewing angle were used. Therefore, the source of light shined on the sample at an angle of 45° and the detector received the reflected light at an angle of 0°. The colorimeter device records the color of a specimen placed against a black background and exposed to a standard light source (D65 or regular daylight). Color values were expressed according to the CIE  $L^*a^*b^*$  scale color coordinates: lightness-darkness ( $L^*$ ), red-green ( $a^*$ ), yellow-blue  $(b^{*}).$ 

The specimens were placed individually in contact with the optical geometry of the colorimeter. Baseline color measurements of the unpolymerized composite were recorded through the glass microscope slab. Although the effect of the glass cover on the color measurements was determined to be negligible in previous studies,<sup>15</sup> subsequent color measurements were recorded through the glass slab to eliminate the potential influence of specular and diffuse reflectance from the glass. Because the diameter of the colorimeter optical geometry was smaller than the diameter of the specimens, 3 mm and 10 mm, respectively, measurements were recorded in three overlapping areas and averaged to determine a single baseline color value. LED light curing unit (Bluephase16i/Ivoclar-Vivadent, Amherst, NY, USA) with a customized light curing tip ( $\geq 10$  mm in diameter) was used to polymerize the specimens for 20 seconds. An irradiance of 1,600 mW/cm<sup>2</sup> was ensured at all times by periodically monitoring the light curing output with a radiometer (Demetron/Kerr, Orange, CA, USA). Only 3 mm specimens were polymerized from both sides. Immediately after polymerization, a second set of color measurements was recorded following the same procedures as described for baseline color measurements. Individual specimens were stored in

Product	Manufacturer	Туре	Shade	Matrix	Particle	Filler	Filler content			
			A3/Bleach		sıze (µm)	type	% <b>W</b> t	% Vol		
Ceram X Duo	Dentsply-Caulk (Milford, DE, USA)	Nanohybrid	Duo E2/Duo DB	Methacrylate modified polysiloxane/dimethacrylate resin	nodified 0.02–0.03 Barium-aluminum-borosilicate /dimethacrylate glass; methacrylate functionalised silicon dioxide nanofiller		76	57		
Filtek Supreme Plus	3M-ESPE (St. Paul, MN, USA)	Nanohybrid	A3 E/White Enamel	Bis-EMA/UDMA/Bis-GMA/ TEGDMA	0.02	SiO <sub>2</sub> Nanosilica filler, ZrO <sub>2</sub> //SiO <sub>2</sub> nanoclusters	78.5	59.5		
Four Seasons	lvoclar-Vivadent (Amherst, NY, USA)	Hybrid	Standard A3/Bleach XL	Bis-GMA/UDMA/TEGDMA	0.6	Barium glass, ytterbium trifluoride, Ba-Al fluorsilicate glass, dispersed silicon dioxide, spheroid mixed oxide	76	56.5		
Tetric EvoCeram	lvoclar-Vivadent (Amherst, NY, USA)	Nanohybrid	Standard A3/Bleach XL	Dimethacrylates	0.55	Barium glass, ytterbium trifluoride, mixed oxide, prepolymers	75.5	54		
Point 4	Kerr (Orange, CA, USA)	Microhybrid	A3 Base shade/XLI	Bis-GMA/UDMA/TEGDMA	0.4	Barium aluminoborosilicate glass, fumed silicon dioxide filler particles	77	59		
Premise	Kerr (Orange, CA, USA)	Nanohybrid	A3 Body shade/XLI	Bis-EMA/UDMA/TEGDMA	0.02–0.4	Prepolymerized filler, barium glass, silica nanoparticles	84	69		
Venus	Heraeus-Kulzer (Hanau, Germany)	Hybrid	Standard A3/Super Bleach SB1	Bis-GMA/TEGDMA	0.7	Barium aluminum boron fluoride silica glass, dispersed silicon dioxide	78	61		
Gradia Direct	GC America (Alsip, IL, USA)	Hybrid	Standard A3/Bleach white	UDMA co-monomer matrix	0.85 Silica, prepolymerized fillers, fluoro- alumino-silicate glass		77	65		
Vit-I-Escence	Ultradent (South Jordan, UT, USA)	Hybrid	A3 Enamel/ Opaque white	Bis-GMA	0.7	0.7 Barium alumina silicate		58		
Artiste™	Pentron (Wallingford, CT, USA)	Nanohybrid	A Enamel/Bleach Enamel	PCBisGMA/BisGMA/UDMA/ HDDMA	0.02–0.7	Barium boro-silicate glass, nanoparticulated silica, zirconium silicate	75	66		
Bis-EMA=ethoxylated bisphenol A dimethacrylate; Bis-GMA=bisphenol A glycidyl dimethacrylate; TEGDMA=triethylene glycol dimethacrylate; UDMA=urethane dimethacrylate.										

hermetically sealed plastic bags. The individual bags were stored for 24 hours in a dry oven at 37°C after which a third set of color measurements was recorded. Specimens were placed in hermetically sealed plastic bags containing distilled water and the bags were stored for 1 month in a dry oven at 37°C after which the last set of color measurements was recorded.

#### Color Change

Overall color change ( $\Delta E^*$ ) was calculated using the following equation:  $\Delta E^* = ([\Delta L^*]^2 + [\Delta a^*]^2 + [\Delta b^*]^2)^{1/2}$ . Mean  $\Delta E^*$  values for the four study groups were calculated between baseline and immediate polymerization ( $\Delta E^*_1$ ), between immediate

polymerization and 24 hours ( $\Delta E_2^*$ ), and between 24 hours and 1 month ( $\Delta E_3^*$ ). Total net color change result after 1 month relative to baseline was also calculated. Values  $\Delta E^* \ge 3.3$  were considered clinically unacceptable based on available studies.<sup>5–8</sup> Further analysis of the changes taking place on each of the individual color parameters ( $\Delta L^*$ ,  $\Delta a^*$ ,  $\Delta b^*$ ) was completed only for those groups showing values  $\Delta E^* \ge 3.3$ .

#### Statistical Analyses

A three-way analysis of variance (ANOVA) was used to evaluate whether significant differences in color change existed among study groups at each of the testing times. If differences were found, Student–Newman–Keuls's tests were used to identify the differences. A significance level of 0.05 was used for all tests.

#### RESULTS

Overall, when all materials in both shades were analyzed as a group, 3 mm specimens showed greater color change than 1 mm specimens at baseline (p < 0.001) and

#### **TABLE 2.** Three-way analysis of variance results for $\Delta E^*$

24 hours (p < 0.001). No differences were observed between the two thicknesses at 1 month (p = 0.384). Similarly, when all materials in both thicknesses were analyzed as a group, bleach shades showed significantly greater color change than A3 shades (p < 0.001) at 24 hours and 1 month. Immediately after polymerization, A3 shades showed significantly greater color change than bleach shades (p < 0.001).

# $\Delta E^*$ Immediately after Polymerization and at 24 Hours

Three-way ANOVA results are summarized in Table 2. Immediately after polymerization and at 24 hours, ANOVA results revealed statistically significant effects of the brand, thickness, shade, and all their interactions on color change (p < 0.001). Mean color change values are shown in Figure 1. Immediately after polymerization, all materials in both shades and thicknesses produced  $\Delta E^* \ge 3.3$  except for Tetric EvoCeram in bleach shade. At 24 hours, 16 out of the 20 groups of 1 mm thickness showed  $\Delta E^* \ge 3.3$ , whereas only four out of the 20 groups of 3 mm thickness showed  $\Delta E^* \ge 3.3$ .

Source of	DF		Immedia	ate ( $\Delta E_1$ )		24 hours (ΔE <sub>2</sub> )			l month (ΔE <sub>3</sub> )				
variation		SS	MS	F	Þ	SS	MS	F	þ	SS	MS	F	Þ
Thickness	I	6.188	6.188	24.142	<0.001	219.430	219.430	1357.355	<0.001	0.215	0.215	0.763	0.384
Shade	I	13.447	13.447	52.462	<0.001	123.701	123.701	765.189	<0.001	3.430	3.430	12.202	<0.001
Brand	9	429.300	47.700	186.092	<0.001	283.470	31.497	194.833	<0.001	31.348	3.483	12.392	<0.001
Thickness × shade	I	4.917	4.917	19.184	<0.001	24.760	24.760	153.159	<0.001	1.665	1.665	5.925	0.016
Thickness × brand	9	.893	12.433	48.503	<0.001	103.935	11.548	71.436	<0.001	2.395	0.266	0.947	0.486
Shade × brand	9	228.578	25.398	99.083	<0.001	325.514	36.168	223.730	<0.001	17.652	1.961	6.978	<0.001
Thickness × shade × brand	9	42.809	4.757	18.557	<0.001	81.362	9.040	55.921	<0.001	4.377	0.486	1.730	0.086
Residual	160	41.012	0.256			25.866	0.162			44.972	0.281		
Total	199	878.144	4.413			1188.038	5.970			106.054	0.533		
DF=degrees of freedom; SS=sum of squares; MS=mean squares.													





#### $\Delta E^*$ at 1 Month

At 1 month, statistically significant effect of the brand and shade was found (p < 0.001) but no effect of the thickness was detected in this case (p = 0.384). The interactions between thickness and shade (p = 0.016) and between shade and brand (p < 0.001) were also found to be statistically significant. Conversely, the interaction between thickness and brand (p = 0.486) and the triple interaction thickness × shade × brand (p = 0.086) did not show statistical significance. Mean color change values are shown in Figure 1. All groups showed  $\Delta E^*$  values below 3.3 after 1 month. Therefore, no further analysis of the contribution of each of the individual parameters  $L^*$ ,  $a^*$ , and  $b^*$  was conducted for these groups.

# $\Delta L^*,\,\Delta a^*,\,\Delta b^*$ Immediately after Polymerization and at 24 Hours

Immediately after polymerization, ANOVA results revealed statistically significant effects of the brand, thickness, shade, and all their interactions on parameters  $\Delta L^*$ ,  $\Delta a^*$ , and  $\Delta b^*$  (p < 0.05). Similar results were found at 24 hours, except for the interaction between thickness and shade for  $\Delta a^*$ , which became non-statistically significant (p = 0.200). Figures 2 and 3 summarize the mean change on  $L^*$ ,  $a^*$ , and  $b^*$  parameters for the four study groups immediately after polymerization and at 24 hours.

#### $\Delta L^*$ , $\Delta a^*$ , $\Delta b^*$ at 1 Month

After 1 month, the interactions thickness × shade (p = 0.410), thickness × brand (p = 0.425) and the triple interaction thickness × shade × brand (p = 0.912) did not have a statistically significant effect on  $\Delta L^*$ . No statistically significant effect of the thickness was detected on either  $\Delta a^*$  or  $\Delta b^*$  with p = 0.111 and p = 0.464, respectively. The interaction thickness × shade was also shown to be non-statistically significant for both  $\Delta a^*$  and  $\Delta b^*$  with p = 0.475 and p = 0.512, respectively. Statistically significant effect of the brand, thickness, shade, and the interaction shade × brand was shown for  $\Delta L^*$  (p < 0.05). The brand, shade, and the

interactions thickness × brand, shade × brand, and the triple interaction thickness × shade × brand were also shown to have a statistically significant effect on  $\Delta a^*$  and  $\Delta b^*$  (p < 0.05).

#### DISCUSSION

This study evaluated the color change of ten commercially available brands of composite immediately after polymerization, at 24 hours, and after 1 month of water storage. In addition to evaluating the color stability behavior of a number of composites during the different stages of maturation and storage, this study also intended to try and isolate the variables that may be responsible for the observed color changes at each of the different stages such as for example post-polymerization reaction and water storage. With this purpose, the color change analysis was broken down in three segments, changes observed immediately after polymerization, at 24 hours, and after 1 month of water storage.

Under the tested conditions, color changes at 24 hours could be attributed to the composite post-polymerization reaction and changes after 1 month could be attributed to the resin water sorption. The total net color change result after 1 month relative to baseline was also calculated in order to determine if changes taking place at any given stage were neutralized by changes taking place at a later stage. In most cases, the total net color change result after 1 month relative to baseline was clinically unacceptable. This was true for 12 out of the 20 1 mm specimens and for 18 out of the 20 3 mm samples. Most of the clinically relevant color changes occurred after initial polymerization and at 24 hours. Color changes after 1 month relative to 24 hours were negligible and remained under the threshold of 3.3 for all materials indicating that they played only a minor role in the total net color change result.

The inclusion of ten brands of composite, two shades, two thicknesses, and two testing times was intended to provide an overall assessment of the different materials'





behavior when applied and tested according to the different study variables. The tested composites included a number of the most representative hybrid, microhybrid, and nanohybrid materials, at least in the United States market, all of which can be used for anterior and posterior applications. Shades A3 and bleach were included as two end values in a range of commonly used shades. Although most color studies normally test specimens in 2 mm thickness,<sup>3,28,29</sup> 1 mm and 3 mm thicknesses were investigated in order to observe the behavior of the different materials in two extreme situations, but yet within a clinically acceptable range. Based on most available studies, values  $\Delta E^* \ge 3.3$ were considered clinically unacceptable.<sup>5,6,8</sup> For changes above the 3.3 level, further analysis of  $\Delta L^*$ ,  $\Delta a^*$ ,  $\Delta b^*$ was conducted to determine the contribution of each of the individual parameters to the overall color change as well as direction of the observed color shifts.

As shown in Table 2, the ANOVA results demonstrated that the color change was dependent on the specific brand, shade, and thickness of composite as well as on the interactions among these variables. When all materials in both shades were analyzed as a group, 3 mm samples showed greater color change than 1 mm samples. This was true immediately after polymerization and at 24 hours. No differences between the two thicknesses were observed at 1 month. Our results differ from those by Arikawa who found greater color change with thinner samples.<sup>30</sup> Although the translucency parameter was not measured as a part of this study, the greater color change of 3 mm samples when all materials in both shades were analyzed as a group may be the result of the greater effect of the black background in 1 mm samples. Perhaps the apparent greater translucency of 1 mm samples made true color shifts less evident as compared to the 3 mm samples. Translucency has been shown to increase exponentially as the thickness of composite decreases.<sup>31</sup> However, the similar trends observed for the two thicknesses tested seem to indicate that the opacity of all tested materials might have been enough to avoid this effect. Similarly, when all materials in both thicknesses were analyzed as a group, bleach shades showed significantly greater color change than A3 shades. This was only true at 24 hours and 1 month.

These results are in agreement with those by Hirobumi who found that lighter shades of composite were likely to exhibit higher color degradation after 24 hours of water storage.32 However, immediately after polymerization, A3 shades showed significantly greater color change than bleach shades. These results are in agreement with those obtained by Shin who demonstrated that the use of alternative photoinitiators present in bleach shades resulted in an increased color stability of composites.<sup>19</sup> Light-activated composites commonly use camphorquinone (CQ)/amine as their photoinitiator system. Although present in very small amounts, CQ can significantly affect the material's color.<sup>12,19</sup> Alternative white and colorless photoinitiators, such as lucirin TPO and phenylpropanedione are used in lighter shades, either alone or in combination with CQ, to avoid the photo-yellowing effect of the composite.<sup>19</sup> The use of photoinitiators that provide greater color stability during the polymerization process in bleach shades could explain the less color shift observed for bleach shades as compared with A3 shades after initial polymerization for most of the materials tested. As shown in Figure 1, a somewhat consistent behavior through the different stages of maturation and storage was observed when the different materials were tested in A3 shade. Except for Point 4 and Vit-l-Escence, which remained under 3.3 at 24 hours, all 1 mm samples showed changes above 3.3 after polymerization and at 24 hours, while all 3 mm samples showed changes above 3.3 after polymerization and below 3.3 at 24-hours. On the contrary, no consistent behavioral pattern across the tested materials was evident when they were tested in bleach shades with changes observed above and below 3.3 initially and at 24 hours.

Figure 1 summarizes the overall color change ( $\Delta E^*$ ) immediately after polymerization, at 24 hours, and after 1 month of water storage as well as the total net color change result between baseline and 1 month. In general, color change decreased with time, and changes at 24 hours and 1 month were less than those observed immediately after polymerization. Immediately after polymerization, all materials in both shades and thicknesses produced changes beyond the clinically acceptable threshold of 3.3 except for Tetric EvoCeram in bleach shade, which remained below 3.3. Interestingly, Tetric in bleach shade displayed the greatest color change of all brands at 24 hours. Color changes after polymerization have been attributed to the shifts in the optical properties of the resin known to take place during the cross-linking of the monomers into polymeric chains.<sup>16</sup> The degree of polymerization is in turn dependent on factors such as the radiant intensity of the polymerization unit, polymerization time, shade and translucency/opacity of a composite, as well as the absorption, scattering characteristics<sup>33,34</sup> and thickness of the resin.<sup>30</sup> To avoid under-polymerization and its potential effect on the color stability, 3 mm samples were polymerized from both sides. Although conversion ratios were not calculated as a part of this study, it is possible that the degree of polymerization could have been maximized for the 3 mm samples with the 40 second exposure to light. Conversely, the 20 second exposure time of the 1 mm samples might have resulted in less initial polymerization and therefore less associated changes in the optical properties and color after initial polymerization.

#### Color Change at 24 Hours

At 24 hours, less color changes were observed when the materials were applied in thicker increments regardless of the shade. When 3 mm specimens were tested in shade A3, all samples showed color change at 24 hours lower than that observed immediately after polymerization, and below the  $\Delta E^*$  threshold of 3.3. The same behavior was observed for six out of the ten brands of the 3 mm samples in bleach shade. Conversely, 1 mm samples in both shades showed color change at 24 hours above the critical  $\Delta E^*$  of 3.3 and in some cases even larger than the change observed immediately after polymerization. Under the testing conditions used in this study, the color change observed at 24 hours could be attributed to the post-irradiation polymerization reaction that composites are known to undergo following light activation and that lasts for up to 24 hours.<sup>35</sup> This dark cure has been shown to be quite extensive with as much as 19–26% of the final conversion occurring during this time period.<sup>36</sup> It is possible that the greater color change of the 1 mm samples at 24 hours could have been the product of a

reduced degree of polymerization following initial light irradiation resulting in a greater amount of unreacted monomer left to react over a 24-hour period and consequently undergoing greater color change. This was particularly true for Tetric EvoCeram in bleach shade, which showed changes below 3.3 on initial polymerization, but at 24 hours, it displayed the greatest color change of all brands when tested in both thicknesses. It is possible that the degree of polymerization on initial light irradiation of Tetric EvoCeram in bleach shade might have been insufficient to generate a clinically evident color change. Hence, an increased post-irradiation polymerization reaction, with the associated greater change in the optical properties of the material may have been responsible for the large color change observed at 24 hours. Coincident with the results from our study, the color stability of Tetric EvoCeram has been previously questioned in the literature with shifts mainly taking place to the dark and yellow region.37

The amount of TEGDMA present in the resin matrix has also been reported to affect the extent of post-irradiation polymerization.<sup>38</sup> As TEGDMA increases, the amount of post-irradiation polymerization decreases because TEGDMA generates higher initial conversion.<sup>38</sup> As shown in Figure 1, of the five materials reported by their manufacturer to contain TEGDMA, only three, Four Seasons, Point 4, and Filtek, demonstrated a color change at 24 hours of less magnitude than that observed after initial polymerization for all combinations of shades and thicknesses. Tetric and Premise in bleach shade showed greater color change at 24 hours than after initial polymerization.

Clinically, provided that a resin color mock up is fabricated for shade selection, color mismatch of a restoration could be avoided or minimized by identifying a shade that provides a better match to the tooth structure once it is polymerized. In this regard, a composite material with "clinically unacceptable" color change immediately after polymerization may represent much less of a concern than a material that does not change on initial polymerization but instead shows a rather large color change after 24 hours of placement. The concern may even be greater if the direction and magnitude of these changes cannot be either understood or anticipated. For example, despite the large change shown immediately after polymerization for Point 4, only minor changes took place at 24 hours indicating that after initial mock-up and shade selection, the color of the restoration remains relatively stable. The opposite was true for Tetric EvoCeram in bleach shade, which showed only minor changes following initial polymerization, but at 24 hours showed the largest color change of all brands with the shift primarily attributed to an increase on the  $L^*$  parameter indicating that the material became lighter.

#### Color Change at I Month

After 1 month of water storage, all groups produced color change values under the threshold of 3.3 and no significant differences among the brands were detected. Under the testing conditions used in this study, the observed color changes could be attributed to the water sorption over a 1 month storage period. However, all changes remained clinically acceptable indicating that water itself may not play a major role in the color change of composites unless it acts as a carrier for staining agents such as coffee, tea, wine, and others, in which case it may promote their absorption with the consequent discoloration over time. A number of studies have reported on the effect of different staining agents in the color stability of composites.<sup>24,25</sup> The present study was intended to evaluate color changes associated with factors inherent to the material composition, post-irradiation polymerization reaction, chemical breakdown of unreacted components and water sorption rather than the result of extrinsic factors on the composites. A controlled environment such as distilled water was used as storage media for this purpose.

#### Change in L\*, a\*, and b\* Parameters

The change in the individual color parameters,  $\Delta L^*$ ,  $\Delta a^*$ , and  $\Delta b^*$ , for all materials, shades, and thicknesses immediately after polymerization and at 24 hours are represented in Figures 2 and 3. Analysis of the individual parameters  $\Delta L^*$ ,  $\Delta a^*$ , and  $\Delta b^*$  at 1 month

was not performed since these values remained under 3.3 and thus did not contribute greatly to the total net color change result. In both cases, after polymerization and at 24 hours, the parameters  $L^*$  and  $b^*$  were responsible for most of the observed changes. Under the tested conditions, only minor changes were observed in the  $a^*$  coordinate; therefore, no further analysis of this parameter was deemed necessary. This is coincident with other studies who found that the  $a^*$ parameter contributed the least to the overall color change.<sup>13,39</sup> As shown in Figure 2, immediately after polymerization, both  $L^*$  and  $b^*$  parameters shifted to the negative values indicating that the samples became darker and less yellow (more blue), respectively. The same behavior was observed across groups for all brands except for Ceram X and Vit-l-Escence in 3 mm bleach shade whose  $L^*$  parameter changed towards the positive values indicating that the samples became lighter. Coincident with our findings, most studies have reported a decrease in the  $L^{*3,15,26}$  and  $b^{*12,15,26,28}$ coordinates after polymerization irrespectiveof the brand and shade. It is common for materials containing camphorquinone as photoinitiator to become less yellow on irradiation with light as the photoinitiator is consumed.<sup>12</sup> Both A3 and bleach shades showed similar degree of change to the negative values for the  $b^*$  parameter (less yellow/more blue).

At 24 hours, further changes in the  $L^*$  and  $b^*$ parameters were observed with most of the shifts taking place in the opposite direction from the changes observed after polymerization (Figure 4). The positive values for the  $L^*$  and  $b^*$  parameters indicated a shift back to the lighter and yellower side, respectively, which was enough to neutralize the changes observed immediately after polymerization for most materials. This was especially true for 1 mm samples in shade A3. Gradia Direct and Filtek provide good examples of materials whose color change after initial polymerization and 24 hours were comparable (Figure 1) but in the opposite direction (Figures 2 and 3). Despite the clinically unacceptable changes observed after polymerization and at 24 hours, these neutralized each other yielding a negligible total net color change result. Later changes at 1 month were only minor and



**FIGURE 4.** Resin sample showing color shift at 24 hours to the light and yellow region. A, Sample after initial polymerization. B, Sample at 24 hours.

remained under 3.3. Of the four materials with a total net color change above 3.3 in that same group of 1 mm samples in shade A3, Four Seasons, Point 4, and Ceram X were derived predominantly from changes following initial polymerization, which were not neutralized by shifts at 24 hours. Although Artiste showed the same degree of color change immediately after polymerization and 24 hours, the extent of these changes for each of the individual  $L^*$  and  $b^*$  parameters was not equivalent yielding a net result after 24 hours, which was not able to neutralize the changes observed after initial polymerization. This, in addition to the minor changes observed after 1 month, resulted in an overall color change net result above 3.3.

As shown in Figure 1, changes immediately after polymerization for A3 samples in 3 mm thickness were above the 3.3 level. A detailed analysis of the  $L^*$ ,  $a^*$ , and  $b^*$  parameters for the changes occurred immediately after polymerization revealed that most of these changes were toward the dark and blue region as represented in Figure 2. Although the changes at 24 hours took place in the opposite direction (Figure 3), they remained under the 3.3 threshold level as shown in Figure 1, and therefore were not sufficient to counteract the changes occurred following initial polymerization. Similar to all other groups, the color change at 1 month was insignificant indicating that the net total color change result for eight out of the ten materials, when comparing baseline to 1 month, was predominantly the result of changes taking place initially after polymerization.

Bleach shades showed a less consistent behavior than A3 shades. As shown in Figure 2, the color changes following initial polymerization were primarily attributed to changes in the  $b^*$  parameter toward the blue (less yellow) region and to a lesser extent to changes in the *L*<sup>\*</sup> parameter towards the dark region except for Four Seasons and Filtek, whose changes were primarily attributed to shifts in the  $L^*$  parameter. The opposite was true at 24 hours as represented in Figure 3. Changes in the  $L^*$  parameter to the lighter side accounted for most of the observed changes, and shifts in the  $b^*$ coordinate to the yellow side provided less contribution to the overall color change. Twenty-four-hour shifts in the  $L^*$  parameter toward the lighter side were greater than the initial shifts to the darker side indicating that they not only counteracted but also exceeded the changes observed initially with the resultant net result of a brighter shade. The described behavior was especially evident in 1 mm samples and may be considered particularly advantageous for bleach shades since a final net result of brighter teeth may be highly desirable. Nevertheless, the prior statement should be interpreted cautiously as the described shifts to the lighter side may, more often than not, be derived from changes occurred

at 24 hours, which as explained previously, is less than ideal.

Further research is needed in order to gain a better understanding of the different stages in the process of maturation and storage where most of these changes are likely to take place. Only when a reproducible behavioral pattern can be established for each of these materials, clinicians will be able to make more accurate predictions regarding the direction and magnitude of the changes that are expected to occur, and therefore, they will be able to master color matching situations through a better understanding of the material of their choice. Ceram X and Vit-l-Escence showed changes in the  $L^*$  parameter that were opposite from those observed for all other materials when tested in bleach shade. Immediately after polymerization, the  $L^*$ parameter changed to the positive values indicating that the materials became lighter, and at 24 hours, the materials showed less important changes back to the darker side. However, as shown in Figure 1, changes at 24 hours and 1 month were minor for both materials indicating that the large total net color change result at 1 month relative to baseline was primarily attributed to shifts towards the lighter and bluer (less yellow) side that took place initially after polymerization.

### CONCLUSIONS

Within the limitations of this in vitro study, the following can be concluded:

- A large color shift occurs immediately after polymerization of current resin composite materials. The extent of these changes is dependant on the brand, thickness, and shade of composite.
- 2 To minimize shade mismatch issues, fabrication of a resin mock-up is still the standard recommended procedure for shade selection.
- 3 Overall, composites became darker and less yellow immediately after polymerization. At 24 hours, color changes were still observed with a shift back to the lighter and yellower side. For some materials, changes observed at 24 hours were sufficient to neutralize the changes occurred after initial

polymerization. After 1 month, only negligible color shifts took place, and thus did not contribute considerably to the total net color change result.

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