

Influence of color coordinates on the color changes after polymerization and thermocycling of dental resin composites

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Color changes after polymerization and aging of dental resin composites are perceptible. If these color changes can be predicted by the shade of material before polymerization or clinical use, color matching of composites with teeth would be improved. The objectives were to measure the correlations among the color coordinates ($CIE L^*$, a^* and b^*) of dental resin composites and the changes in color (ΔE_{ab}^*) and color parameters (ΔL^* , ΔC_{ab}^* and ΔH_{ab}^*) after polymerization (PO) and thermocycling (TC), and to determine whether the range of color coordinates influenced these correlations. Color of two resin composites (26 shades) was measured before and after PO, and after TC with a reflection spectrophotometer. Regression analyses were performed among the color coordinates and the changes in color and color parameters after PO and TC. After PO, ΔE_{ab}^* was correlated with $CIE L^*$ value measured after PO, and $CIE a^*$ and b^* values measured before PO ($p < 0.01$). After TC, ΔE_{ab}^* was correlated with $CIE b^*$, a^* , and L^* measured after PO ($p < 0.01$). Correlations among the color coordinates and the changes in color parameters after PO or TC varied, but were generally significant ($p < 0.01$). Multiple regression analyses showed that ΔE_{ab}^* values after PO and TC were mainly influenced by $CIE L^*$ after PO (multiple $r = 0.87$ after PO and 0.58 after TC). Correlations were also influenced by the range of color coordinates. Within the limit of this study, shade of resin composites significantly influenced the changes in color and color coordinates after PO and TC. Light shades (high $CIE L^*$ shades measured after polymerization) showed small color changes after polymerization and thermocycling. © 2005 Springer Science + Business Media, Inc.

1. Introduction

The relatively large color changes of dental resin composites after polymerization and aging indicate that these changes must be taken into account during initial clinical shade match at placement of the unpolymerized composite [1]. Light-cured resin composites produce a perceptible color change as a result of the polymerization for most shades of materials, and generally showed a characteristic chromatic shift toward the blue region of color space [2]. Changes in color coordinates after polymerization were shade dependent, and changes in $CIE L^*$ values had the greatest influence on the overall color changes [3]. Polymerization color changes of composites for bleach shades varied by the composites, and microhybrid composites became less saturated and darker after polymerization [4]. After polymerization, $CIE L^*$ decreased in hybrid resin composites and changes in $CIE a^*$ were small [5].

After aging, resin composites became to have lower luminous reflectance and excitation purity and higher dominant wavelength [6], and became darker and more chromatic [7]. Because the direction and magnitude of the color changes differed for the thermally and pho-

tochemically induced color changes, it seemed that different mechanisms were involved [8]. Regardless of the aging conditions, color shifts, chroma changes, and hue differences were observed in resin composites [9, 10]. After aging, composites for bleach shades became darker and more saturated [11].

Color changes after polymerization and aging of resin composites are visually significant in many cases. Two clinical methods were recommended to solve the problems of color changes after polymerization. Selection of a more yellow or more chromatic shade than the restored tooth was recommended as the first method [2]. Clinical practice of polymerizing some material on or adjacent to the undried tooth to confirm shade of material before restorative procedures was the second method [3]. Changes in color and optical properties after polymerization and aging of resin composites for bleach shades were compared with those of conventional shades [12].

Currently, correlations among color coordinates of resin composites and the changes in color and color parameters are not completely understood, although the directions of changes in color parameters have been

confirmed in previous studies [2, 4, 9]. The working hypothesis of the present study was that there were significant correlations among the color coordinates of resin composites and the changes in color and color parameters after polymerization and aging. The objectives of this study were to measure the correlations among the color coordinates of resin composites and the changes in color and color parameters after polymerization and thermocycling, and to determine the change of these correlations by the range of color coordinates.

2. Material and methods

Two brands of light-cured dental resin composites were studied. Synergy (Coltene/Whaledent, Mawah, NJ, USA) was composed of resin matrix containing BisGMA, BisEMA and TEGDMA and 59 vol% strontium-barium-borosilicate filler particles of 0.04–2.5 μm (average; 0.6 μm). The shades investigated were N, O, P, Transparent, A1, A2, A3, A3.5, A4 and C2. Vitalescence (Ultradent Products, South Jordan, UT, USA) was composed of BisGMA-based resin matrix and 58 vol% of micro-hybrid particles of average 0.7 μm . The shades investigated were Pearl Frost, Pearl Snow, Trans Gray, Trans Ice, Trans Mist, Trans Yellow, A1, A2, A3, A3.5, B1, B2, B3, C1, C2, and C3.

Composites were packed into a polytetrafluoroethylene mold (8 mm in diameter and 2 mm in thickness). Five specimens were made for each shade. Color measurement before polymerization was made with the cover glasses attached on the specimen. Each specimen was then light cured for 40 s using a light-curing unit (Spectrum 800, Dentsply/Caulk, Milford, DE, USA) with an intensity setting of 400 mW/cm², and the output of the light was checked with a radiometer (Demetron, Danbury, MA, USA). Color of polymerized specimen was measured after removing both cover glasses. Color was measured according to CIELAB color scale relative to the CIE standard illuminant D65 over a white background ($CIE L^* = 96.68$, $a^* = -0.18$, and $b^* = -0.22$) on a reflection spectrophotometer (CM-3500d, Minolta, Osaka, Japan) with specular component excluded (SCE) geometry. $CIE L^*$ means lightness, $CIE a^*$ means red-green parameter, and $CIE b^*$ means yellow-blue parameter. Illuminating and viewing configuration was CIE diffuse/10° geometry. The influence of the cover glasses on the color measurement was determined to be negligible in the previous studies [13, 14]. Polymerized specimens were thermocycled between 5 and 55°C distilled water for 3,000 cycles with a dwell time of 15 s, and color was measured again. CIELAB color difference was calculated as $\Delta E_{ab}^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$. Chroma was calculated as $C_{ab}^* = [(\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$. Changes in lightness or chroma were calculated as ΔL^* (or ΔC_{ab}^*) = $CIE L^*$ (or C_{ab}^*) after polymerization/thermocycling— $CIE L^*$ (or C_{ab}^*) before polymerization/thermocycling. Changes in hue (ΔH_{ab}^*) was calculated as $\Delta H_{ab}^* = [(\Delta E_{ab}^*)^2 - (\Delta L^*)^2 - (\Delta C_{ab}^*)^2]^{1/2}$ [15].

Regression analysis was used to determine the correlations between ΔE_{ab}^* after polymerization and each

color coordinate measured before and after polymerization, between ΔE_{ab}^* after thermocycling and each color coordinate measured after polymerization, and between the changes in color parameters after polymerization or thermocycling and the color coordinates ($\alpha = 0.01$). Multiple regression analysis was used to determine the significantly influencing variables (predictors) on the color changes after polymerization or thermocycling by a forward regression analysis ($\alpha = 0.01$). The influence of the range of the most influencing predictor on the multiple regressions was also calculated. To eliminate the impact of interrelated independent variables, the variable which showed the lower standardized partial correlation coefficient (β) was not included in the regression equation when the tolerance between two influencing variables was lower than 0.30 [16].

3. Results

Ranges of color coordinates and changes in color and color parameters after polymerization and thermocycling are presented in Table I. After polymerization, mean values for the changes in color parameters were -5.3 in lightness, -2.9 in chroma, and 3.1 in hue. After thermocycling, those were 4.5 , -0.2 , and 1.0 , respectively.

Correlations between color coordinates and color changes (ΔE_{ab}^*) are presented in Table II and Figs 1–2. ΔE_{ab}^* values after polymerization (PO) and thermocycling (TC) were significantly correlated with several color coordinates. In Fig. 1, correlation between ΔE_{ab}^* after PO and $CIE L^*$ after PO [which showed the highest absolute correlation coefficient (r) with ΔE_{ab}^*] is presented. In Fig. 2, correlation between ΔE_{ab}^* after TC and $CIE b^*$ after PO (highest r value) is presented. In these graphs, second order regression equations are presented.

TABLE I Ranges and average values of color coordinates of specimens and changes in color and color parameters after polymerization and thermocycling

Parameter	Condition	Range	Average
$CIE L^{*a}$	Before PO ^b	41.4–66.7	53.0 (7.1) ^c
	After PO	34.3–60.7	47.7 (6.8)
$CIE a^*$	Before PO	–6.0–0.0	–2.3 (1.2)
	After PO	–7.1–2.8	–2.4 (1.2)
$CIE b^*$	Before PO	0.7–12.4	6.9 (3.2)
	After PO	–8.6–9.1	2.4 (3.6)
C_{ab}^*	Before PO	3.3–12.4	7.7 (2.5)
	After PO	1.1–9.3	4.8 (1.7)
ΔE_{ab}^{*d}	After PO	4.2–14.7	8.4 (2.1)
	After TC	1.2–9.8	4.8 (1.7)
ΔL^*	After PO	–14.0–4.0	–5.3 (4.2)
	After TC	–0.7–9.4	4.5 (1.8)
ΔC_{ab}^*	After PO	–7.8–5.9	–2.9 (2.7)
	After TC	–4.2–3.0	–0.2 (1.2)
ΔH_{ab}^*	After PO	0.0–9.8	3.1 (1.9)
	After TC	0.0–6.5	1.0 (0.9)

^aColor coordinates of specimens.

^bPO means polymerization and TC means thermocycling.

^cStandard deviations are in parentheses.

^d ΔE_{ab}^* means color changes, and ΔL^* , ΔC_{ab}^* , and ΔH_{ab}^* mean changes in lightness, chroma and hue (value after PO or TC—value before PO or TC).

TABLE II Correlations between the color coordinates and the color changes

Dependent variable	Independent variable	Correlation coefficient (r) ^a
ΔE_{ab}^* after PO ^b	<i>CIE L*</i> before PO	NS ^c
	<i>CIE L*</i> after PO	-0.51
	<i>CIE a*</i> before PO	-0.35
	<i>CIE a*</i> after PO	NS
	<i>CIE b*</i> before PO	-0.32
	<i>CIE b*</i> after PO	NS
	C_{ab}^* before PO	-0.30
	C_{ab}^* after PO	0.22
ΔE_{ab}^* after TC	<i>CIE L*</i> after PO	-0.22
	<i>CIE a*</i> after PO	-0.31
	<i>CIE b*</i> after PO	-0.32
	C_{ab}^* after PO	0.16

^aAll the correlation coefficients were significant at the significance level of 0.01.

^bPO means polymerization and TC means thermocycling.

^cNS means no significant correlation.

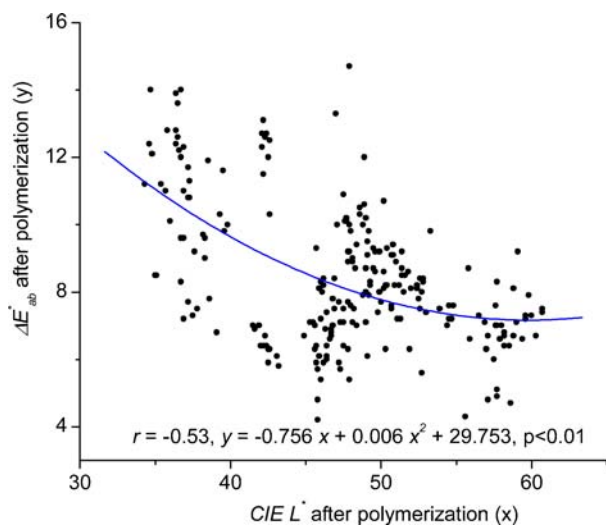


Figure 1 Correlation between *CIE L** and color change after polymerization.

Correlations between each color coordinate and changes in each color parameter are listed in Table III. Changes in lightness (ΔL^*) after PO were correlated with *CIE a** and *L** values measured before PO. Change in chroma (ΔC_{ab}^*) after PO was highly correlated with chroma measured before and after PO. Change in hue (ΔH_{ab}^*) after PO was correlated with many color coordinates. ΔL^* after TC showed negative correlation with *CIE b** after PO, and ΔC_{ab}^* after TC showed positive correlation with *CIE a** after PO.

Influencing variables (predictors) on the color changes by multiple regressions are listed in Table IV. The range of multiple correlation coefficients among the predictors and the changes in color/color parameters was 0.67 to 0.99 after PO, and was 0.30 to 0.58 after TC.

Because the most highly influencing variable on color change after PO was *CIE L** value measured after PO and that after TC was *CIE b** after PO, full ranges of two predictors were divided into three equal parts to observe the difference in the correlations by

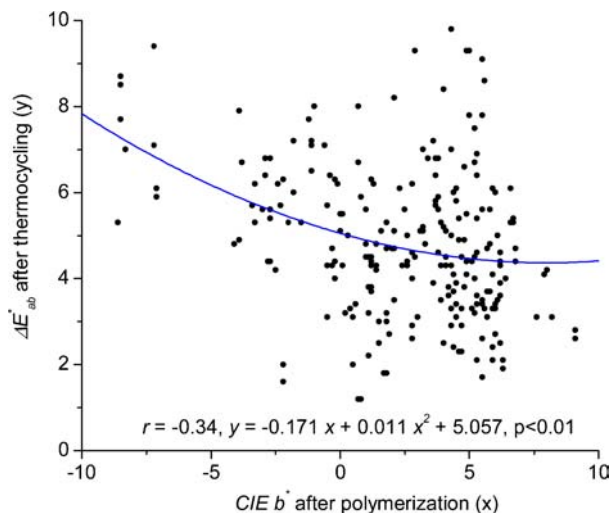


Figure 2 Correlation between *CIE b** and color change after thermocycling.

the range of predictor (Table V). Predictors by multiple regressions varied by the range of predictors and were different from those based on the all data. However, the multiple correlation coefficients were similar to those based on all the data (0.87 vs. 0.81–0.86 after PO; 0.58 vs. 0.54–0.59).

4. Discussion

The working hypothesis of the present study was accepted because there were significant correlations among the color coordinates of resin composites and the changes in color/color parameters after polymerization and thermocycling. Predictability for the color change after polymerization was as high as 0.87, and that for the color change after thermocycling was 0.58 based on the multiple regression analyses (Tables IV). It was supposed to be possible to develop a predictive model for the color changes after polymerization or aging from the color coordinates of materials because a predictive model has already been developed for the efficacy of tooth bleaching depending on the color coordinates of tooth [17]. However, simple correlations coefficients between color coordinates and color changes (Table II) and between color coordinate and changes in color parameters were not high except some cases in the present study. This implies that changes in color and color parameters are influenced simultaneously by three color coordinates, not by a single coordinate.

Changes in color parameters varied after polymerization and thermocycling. In both conditions, changes in lightness were the highest (Table I), which were in agreement with the previous studies [3, 5, 18]. This result might reflect the fact that the magnitude of the distribution range of *CIE L** values of the specimens was higher than those of *CIE a** and *b** (26.5 vs. 10.0 in *CIE a** and 17.8 in *CIE b** after PO condition). The difference of two composites was not considered in this study because two composites showed similar range of changes in color and color parameters. Mean ΔE_{ab}^* was 7.4 (± 1.4) in Synergy and 9.0 (± 2.2) in

TABLE III Correlations between the color coordinates and the changes in color parameters

Independent variable	ΔL^* after PO ^a	ΔC_{ab}^* after PO	ΔH_{ab}^* after PO	ΔL^* after TC	ΔC_{ab}^* after TC	ΔH_{ab}^* after TC
<i>CIE L*</i> before PO	-0.37 ^b	NS	NS ^c	NA ^d	NA	NA
<i>CIE a*</i> before PO	0.43	-0.69	-0.31	NA	NA	NA
<i>CIE b*</i> before PO	0.18	-0.83	-0.51	NA	NA	NA
<i>C_{ab}^*</i> before PO	0.17	-0.78	-0.49	NA	NA	NA
<i>CIE L*</i> after PO	0.24	NS	0.22	-0.21	-0.38	NS
<i>CIE a*</i> after PO	NS	-0.37	-0.59	-0.31	0.45	-0.24
<i>CIE b*</i> after PO	-0.31	-0.58	-0.77	-0.33	0.29	NS
<i>C_{ab}^*</i> after PO	-0.19	0.41	NS	NS	-0.29	NS

^aPO means polymerization and TC means thermocycling.

^bAll the correlation coefficients (*r*) were significant at the significance level of 0.01.

^cNS means no significant correlation.

^dNA means not applicable.

TABLE IV Predictors on the changes in color and color parameters from multiple regressions

	Change	<i>r</i> ^a	Predictor (β , <i>t</i> -value) ^b
PO ^c	ΔE_{ab}^*	0.87	<i>CIE L*</i> AP ^d (-1.57, -26.32); <i>CIE L*</i> BP (1.23, 20.88) <i>CIE b*</i> AP(-0.37, -10.81)
	ΔL^*	0.67	<i>CIE a*</i> BP (0.64, 12.07); <i>CIE b*</i> AP(-0.55, -10.35)
	ΔC_{ab}^*	0.99	<i>CIE b*</i> BP(-0.92, -117.44); <i>C_{ab}^*</i> AP (0.56, 71.55)
	ΔH_{ab}^*	0.83	<i>CIE b*</i> AP(-0.78, -12.57); <i>CIE L*</i> BP(-0.23, -5.05) <i>C_{ab}^*</i> AP (0.14, 3.50); <i>CIE b*</i> BP (0.12, 2.16)
TC	ΔE_{ab}^*	0.58	<i>CIE L*</i> AP(-0.47, -7.50); <i>CIE a*</i> AP(-0.40, -5.44) <i>CIE b*</i> AP(-0.23, -3.52); <i>C_{ab}^*</i> AP (0.17, 2.88)
	ΔL^*	0.55	<i>CIE L*</i> AP(-0.46, -7.27); <i>CIE a*</i> AP(-0.45, -6.32) <i>CIE b*</i> AP(-0.18, -2.91);
	ΔC_{ab}^*	0.56	<i>C_{ab}^*</i> AP(-0.28, -4.79); <i>CIE L*</i> AP(-0.24, -3.77) <i>CIE b*</i> AP (0.22, 3.32); <i>CIE a*</i> AP (0.18, 2.35);
	ΔH_{ab}^*	0.30	<i>CIE a*</i> AP(-0.34, -4.66); <i>CIE L*</i> AP(-0.20, -2.73)

^aMultiple correlation coefficient (*p* < 0.01). For PO analysis, color coordinates measured before and after PO were applied. For TC analysis, color coordinates measured after PO were used.

^bIn the order from the most influencing determined by the absolute *t*-value. Predictor (β = standardized partial correlation coefficient, *t*-value) (*p* < 0.01).

^cPO means polymerization and TC means thermocycling.

^dBP means 'before polymerization', and AP means 'after polymerization'.

TABLE V Influence of the range of the most highly influencing predictors on the correlations

	Predictor	Range	<i>r</i> ^a	Predictor (β , <i>t</i> -value) ^b
ΔE_{ab}^* after PO ^c	<i>CIE L*</i> AP ^d	34.3-43.0	0.82	<i>C_{ab}^*</i> BP (-0.75, -9.88) <i>CIE L*</i> AP (-0.25, -3.32)
		43.1-51.8	0.81	<i>CIE a*</i> AP (-0.47, -6.43) <i>CIE L*</i> BP (0.52, 6.77) <i>CIE b*</i> AP (-0.23, -2.94)
		51.9-60.7	0.86	<i>CIE L*</i> AP (-2.36, -12.53) <i>CIE L*</i> BP (2.22, 11.81)
ΔE_{ab}^* after TC	<i>CIE b*</i> AP	-8.6--2.8	0.54	<i>C_{ab}^*</i> AP (0.54, 2.73)
		-2.7-3.1	0.54	<i>CIE a*</i> AP (-0.66, -6.10) <i>CIE L*</i> AP (-0.29, -2.65)
		3.2-9.1	0.59	<i>CIE L*</i> AP (-0.62, -7.82) <i>CIE a*</i> AP (-0.31, -3.90)

^aMultiple correlation coefficient (*p* < 0.01). For PO analysis, color coordinates measured before and after PO were applied. For TC analysis, color coordinates measured after PO were used.

^bIn the order from the most influencing determined by the absolute *t*-value. Predictor (β = standardized partial correlation coefficient, *t*-value) (*p* < 0.01).

^cPO means polymerization and TC means thermocycling.

^dBP means 'before polymerization', and AP means 'after polymerization'.

Vitaescence after polymerization, which was not significantly different (*p* > 0.05). And mean ΔE_{ab}^* was 5.0 (± 1.7) in Synergy and 4.7 (± 1.8) in Vitaescence after thermocycling, which was also not significantly different (*p* > 0.05).

Concerning the color changes after polymerization, shades with high *CIE a** and *b** values before PO and high *CIE L** value after PO showed small color change (Table II). This means that more chromatic shades before PO and lighter shade after PO resulted in small

color changes. Concerning the color changes after TC, shades with high *CIE L**, *a**, and *b** values resulted in small color changes. To calculate a more highly correlated equation, second order polynomial regression analyses were performed (Figs 1–2). However, *r* value did not increase significantly compared to linear regression (–0.51 vs. –0.53 in Fig. 1, and –0.32 vs. –0.34 in Fig. 2). Therefore, first order linear regression analyses were used in this study.

Multiple regression results for the influencing variables on changes in color/color parameters were different from those based on simple regression between pairs of variables (Table IV). However, it was clear that color coordinates of resin composite influenced the color changes after PO significantly. Specifically, changes in chroma after PO was highly correlated with *CIE b** before PO and chroma value after PO, and multiple *r* value was as high as 0.99. After TC, *r* values were lower than those after PO, which implies that color changes after TC may be also influenced by other factors. Color coordinates after TC was not used in the analyses because those values could be changed by aging conditions. However, color coordinates measured before and after PO are fixed property, specific to each shade of composites. If other optical properties such as the optical constants and translucency parameter were included in the regression analysis, the predictability might be improved.

It was reported that color change after PO was negatively correlated with *CIE b** and *a** value measured after PO ($\beta = -0.73$ and -0.64 , respectively). And after aging, color change was also negatively correlated with *CIE b** ($\beta = -0.51$) and *CIE a** ($\beta = -0.50$) [12]. In the previous study, since the most shades investigated were bleach shades (eight bleach shades out of total 12 shades), the simple correlation results were different from the present study. This means that color changes after polymerization or aging are influenced by the color coordinates of resin composites. Therefore, the range of color coordinates was grouped and analyzed again (Table V). When the *CIE L** value after PO was in the range of 43.1–51.8, the results were similar to those of the previous study [12]. Color changes after polymerization for the bleach shades composites were from 3.7 to 12.0 ΔE_{ab}^* units [4], which was in the similar range of the present study (4.2–14.7). However, the influencing color coordinates varied by the range of *CIE L** value (Table V). This implies that the influencing variables vary by the range of color coordinates, although the amounts of color change were similar. The color and translucency of dental resin composites are determined not only by more macroscopic phenomena such as matrix and filler composition and filler content, but also by relatively minor pigments additions and potentially by initiation component and filler coupling agent [1]. Further study including all of these variables is indicated.

Concerning the influence of the range of *CIE L** values on the color change after PO, *r* values were similar to those based on all the data (Tables IV and V; 0.87 vs. 0.81–0.86). However, when *CIE L** values were lower than 51.8, different color coordinates such as C_{ab}^* be-

fore PO or *CIE a** after PO showed the highest influence. In these conditions, *CIE L** values influenced the color change anyway. Therefore, correlations between color coordinates and color changes after PO were influenced by the range of lightness. Concerning the ΔE_{ab}^* after TC, the orders of influencing variables were changed compared to those based in the all data.

As to the color change after aging or staining, low water sorption, high filler-resin ratio, reduced particle size, and optimal filler-matrix coupling system were related to improved color stability of resin composites [19]. The difference in the color stability of resin composite after exposure to water may be due to the different nature of the matrix and could also be correlated with filler particles dimensions [20]. In the present study, the correlations coefficients between color coordinates and color changes after TC were lower than those after PO. Therefore, it seemed that other factors influenced color changes after aging. Those may be the absorption of water during thermocycling and changes in refractive index of resin matrix and filler. Resistance of modern composites to discolorations still depends on their structure and manipulation [19].

Color coordinates of materials influenced color changes after polymerization and thermocycling. Multiple correlation coefficient between color coordinates and changes in color was 0.87 after polymerization and 0.58 after thermocycling. Correlations were changed when the ranges of the most influencing variables were divided into groups; however, the correlation coefficients were within the similar range (0.81–0.86 after PO and 0.54–0.59 after TC). Within the limit of this study, color changes after polymerization and thermocycling were significantly influenced by the shade of resin composites. After polymerization and aging, light composites would show smaller color changes than dark composites.

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