Color Changes of Dental Resin Composites before and after Polymerization and Storage in Water

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ABSTRACT

The aims of this study were to: (1) evaluate the A2 shades of various types and brands of resin composites to determine if any color differences occurred before and after polymerization and after 1 month of storage in water and (2) examine the correlation among the color changes and changes in Commission internationale de l'éclairage L*, a*, and b* values after polymerization and after 1 month of storage in water. One submicron-hybrid (Spectrum TPH3, DENTSPLY DeTrey, Milford, DE, USA), one nano-filled (Filtek Supreme XT, 3 M ESPE, St. Paul, MN, USA), three micro-hybrid (Filtek Z250, 3 M ESPE; Esthet X, DENTSPLY DeTrey; and Gradia Direct, GC, Tokyo, Japan), and five nano-hybrid (Ceram X, DENTSPLY DeTrey; Clearfil Majesty Esthetics, Kuraray, Osaka, Japan; Premise, Kerr Corporation, Orange, CA, USA; Tetric Evo Ceram, Ivoclar Vivadent, Schaan, Liechtenstein and Tetric N Ceram, Ivoclar Vivadent) light-curing resin composites were tested. The specimens (N = 10 for each composite) were prepared as discs, 12 mm in diameter and 2 mm in thickness, using round molds. The measurements were performed "before polymerization," "after polymerization," and "after I month of storage in water'' using a contact type dental spectrophotometer (Vita Easyshade, Vident, Brea, CA, USA). The range of ΔE^* values after polymerization (ΔE^*) and storage in water (ΔE^* 2) were 4.59 to 14.13 and 1.26 to 6.29, respectively. Nested analysis of variance and post hoc tests revealed that the type of resin composites significantly affected Δa^* , Δa^{*2} , Δb^{*1} , Δb^{*2} , ΔE^{*2} -values, whereas the brand of resin composites affected the changes in all color parameters (p < 0.05). In spite of many improvements in chemical compositions and fillers of the contemporary composites, color changes after polymerization were perceptible in all resin composites. However, color changes after storage in water were in the acceptable ranges for all resin composites except Clearfil Majesty Esthetic and Gradia Direct.

CLINICAL SIGNIFICANCE

In spite of many improvements in chemical compositions and fillers of the contemporary resin composites, the color changes after polymerization were still perceptible in all the resin composites tested in this study, regardless of their types and brands. Such changes may cause esthetic problems clinically, thus should be taken into account when the shade selections are performed. Alternatively, a piece of unpolymerized resin material can be placed on, or adjacent to the tooth to be restored and polymerized to confirm the selected shade of the esthetic material.

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INTRODUCTION

Providing patients with a natural-looking restoration is an important goal for dental professionals. However, obtaining an accurate shade match for restorations can be a difficult and sometimes frustrating experience. The initial selection of a proper shade is carried out by an unpolymerized resin composite, thus, the color stability

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of the resin composite should be acceptable after polymerization as well. Ideally, the color of esthetic dental materials should not be affected by the polymerization mechanism or any aging procedures. However, significant color changes have been reported in several studies on the color changes of resin composites after polymerization and aging, and the magnitude of this change varied among the brands and composition of the resin composites.^{1–3}

The color change of resin composites after polymerization revealed characteristic chromatic shifts.³ Generally, resin composites become lighter or more translucent after irradiation with a light as the b^* value of light-curing resin composites shifted toward the blue region of the color space, which resulted in a perceived decrease in the yellow chroma. The changes for the lightness were greater than for the chromaticity. That being said, selecting an initial color shade more yellow or more chromatic than the desired final color was sometimes advised.^{3,4}

Resin composites have a tendency to discolor during long-term service in the oral cavity.⁵ The internal color change of resinous materials is one of the reasons for these discolorations.⁶ Some of the previous studies have evaluated the color changes of resin composites over specified periods of time. These studies have been performed by Katayama and colleagues⁷ in 37°C of water for 1 month, by Tamura and colleagues⁸ in 37°C of artificial saliva for 40 days, by Morikawa and colleagues⁹ in 60°C of water for 60 days, and by Peutzfeldt and Asmussen¹⁰ in 60°C of water for 1 month. Storing polymerized resin composites in water or artificial saliva was found to cause color changes that were rated barely perceptible to perceptible for the resin composites studied in these researches.

Although polymerization-dependent color changes of resin composites have been extensively reported, resin-based restorative materials have been continually improving so new contemporary restorative materials would be more color stable after polymerization and storage in water for a period of time. In addition, composites are formulated with a variety of chemical and structural variations thus there could be differences among different resin composite types and brands with the same shade designation regarding optical property changes after polymerization and aging.

In the light of all these findings, the aims of our study were to: (1) evaluate the A2 shades of various types and brands of resin composites for any color differences that may have occurred before and after polymerization, and after 1 month of storage in water, and (2) examine the correlation among the color changes and changes in Commission internationale de l'éclairage (CIE) L*, a*, and b* values after polymerization and after 1 month of storage in water. The null hypotheses tested were that: (1) the amount of the changes in color and color parameters before and after polymerization and after polymerization and after 1 month of storage in water were similar regardless of the type and brand of resin composites used, and (2) there is no correlation among the color changes and changes in CIE L^* , a^* , and b^* values after polymerization and after 1 month of storage in water.

MATERIALS AND METHODS

One submicron-hybrid, one nano-filled, three micro-hybrid, and five nano-hybrid contemporary light-curing resin composites of A2 shade were tested in this study (Table 1). The specimens were prepared with polytetrafluoroethylene disc molds (12 mm in diameter and 2 mm in thickness). Mylar strips and glasses were positioned under and over the molds and the resin specimens were light cured through. Ten specimens were prepared for each material. Color measurements for all samples were obtained "before polymerization," "after polymerization," and "after 1 month of storage in water" from the middle of each sample. The "before polymerization" measurements were taken in the mold and over the mylar strips attached.

Color was measured according to the CIELAB color scale relative to the standard illuminant D65 against a white background (CIE $L^* = 94.48$, $a^* = -0.16$,

Code	Brand name	Composite type	Compositions*	Batch # LOT	Manufacturer
TPH3	Spectrum TPH3	Submicron-hybrid	Matrix: Bis-GMA-adduct, Bis-EMA, TEGDMA, photo initiators, and stabilizers	0802002064	DENTSPLY DeTrey, Milford, DE, USA
			Filler: 57 vol% (77 wt%) Bariumaluminumborosilicate glass (mean particle size < 1 μ m), bariumfluoroaluminioborosilicate (mean particle size < 1 μ m), and highly dispersed silicon dioxide (particle size 10–20 nm)		
FZ2	Filtek Z250	Micro-hybrid	Matrix: Bis-GMA, Bis-EMA, UDMA, photo initiators, and stabilizers	6TH	3M ESPE, St. Paul, MN, USA
			Filler: 60 vol% (84.5 wt%) Zirconium/silica filler (0.01–3.5 $\mu\text{m})$		
CRX	Ceram X Duo	Nano-hybrid	Matrix: Methacrylate modified polysiloxane, dimethacrylate resin, fluorescence pigment, UV stabilizer, stabilizer, dl-Camphorquinone and ethyl–4 (dimethylamino) benzoate	0611001601	DENTSPLY DeTrey, Milford, DE, USA
			Filler: 57 vol% (76 wt%) Bariumaluminiumborosilicate glass (1–1.5 μm) and methacrylate functionalised silicon dioxide nano filler (10 nm)		
CME	Clearfil Majesty Esthetics	Nano-hybrid	Matrix: Bis-GMA, hydrophobicaromatic dimethacrylates, and hydrophobicaliphatic dimethacrylates, dl-Camphorquinone	00002B	Kuraray, Osaka, Japan
			Filler: 66 vol% (78 wt%) Silanated barium glass (average particle size 0.7 $\mu m)$ and pre-polymerized organic filler		
PRM	Premise	Nano-hybrid	Matrix: Bis-EMA, TEGDMA, initiators, and stabilizers	424864	Kerr Corporation, Orange, CA, USA
			Filler: 69 vol% (84 wt%) Prepolymerized filler (30 to 50 μm), barium glass (0.4 μm), and silica nanoparticles (0.02 μm)		
TEC	Tetric Evo Ceram	Nano-hybrid	Matrix: Dimethacrylates, additives, catalysts, stabilizers, and pigments	H29941	Ivoclar Vivadent, Schaan, Liechtenstein
			Filler: 53–55 vol% (75–76 wt%) Barium glass (1 μ m), ytterbium trifluoride, mixed oxide (<100 nm), and prepolymers (0.4–3 μ m)		
FSX	Filtek Supreme XT	Nano-filled	Matrix: Bis-GMA, TEGDMA, UDMA, Bis-EMA resins	7JP	3M ESPE, St. Paul, MN, USA
			Filler: 59.5 vol% (78.5 vt%) Nanoagglomerated/nonaggregated silica filler (20 nm) and aggregated zirconia/silica cluster filler with an average particle size of 0.6–1.4 µm with primary particle size of 5–20 nm		
ESX	Esthet X	Micro-hybrid	Matrix: Bis-GMA adduct, TEGDMA, Bis-EMA adduct, camphorquinone, photoinitiator, stabilizer, and pigments	0802154	DENTSPLY DeTrey, Milford, DE, USA
			Filler: 60 vol% (61 wt%) Barium aluminofluorosilicate glass (0.02–2.5 $\mu m)$ and silica dioxide (10–20 nm)		
TNC	Tetric N Ceram	Nano-hybrid	Matrix: phosponic acid acrylate, HEMA, Bis-GMA, UDMA, ethanol, catalyst, and stabilizers	K12971	Ivoclar Vivadent, Schaan, Liechtenstein
			Filler: 55–57 vol% (79 wt%) Barium glass, ytterbium trifluoride, mixed oxide, and copolymers (between 40–3000 nm)		
GRD	Gradia Direct	Micro-hybrid	Matrix: dimethacrylate co-monomers, UDMA, camphorquinone and amine catalysts, and pigments	0710022	GC, Tokyo, Japan
			Filler: 65 vol% (77 wt%) Fluoro alumino silicate glass (average 0.85 μm), prepolymerized filler, and silica (average 0.85 μm)		
Bis-EMA = ethoxylated bisphenol a dimethacrylate; Bis-GMA = bisphenol-glycidyl methacrylate; HEMA = 2-hydroxyethyl methacrylate; TEGDMA = triethyleneglycol dimethacrylate; UDMA = urethane dimethacrylate.					

*The data regarding the compositions of resin composites were obtained from the manufacturers of these composites.

and $b^* = -0.21$) using a contact type dental spectrophotometer (Vita Easyshade, Vident, CA, USA).

The polymerization tip of a conventional halogen polymerization lamp (Optilux 501, Kerr, Orange, CA, USA) was held against the top cover glass centered over the specimen. The specimens were light cured for 40 seconds with a minimum intensity of 450 mW/cm². The output of the curing light was checked with the radiometer incorporated in the device after 10 consecutive measurements. "After polymerization" measurements were taken following storage in distilled water at 37°C for 24 hours and after blot drying of the specimens.

For the evaluation of the color stability of the resin composites, all specimens were stored in distilled water at 37°C for 1 month. Since color formation is a process of oxidation,¹¹ a hole in the stopper of the water containers was prepared in order to provide free access of air to the water baths. Following storage in water, the specimens were washed in distilled water, blot dried, and "after 1 month of storage in water" measurements were undertaken again with a spectrophotometer (Vita Easyshade).

Color differences were calculated using the following formula, with the color coordinates over the white background; $\Delta E^*_{ab} = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$. ΔL^*1 , Δa^*1 , Δb^*1 , and ΔE^*1 indicate the differences between before and after polymerization values; ΔL^*2 , Δa^*2 , Δb^*2 , and ΔE^*2 indicate the differences between after polymerization and after storage in water values.

The changes in color and color parameters were evaluated in two ways: (1) according to the type of composites, and (2) according to the brand of composites (Table 2).

Statistical Analysis

The statistical analysis was processed with the SPSS 13.0 software system (SPSS Inc., Chicago, IL, USA). Nested ANOVA and post-hoc tests (Dunnett t3

TABLE 2. Study groups

Groups	Type of resin composites
I	Submicron-hybrid ($N = 10$)
2	Nano-filled ($N = 10$)
3	Micro-hybrid (N=30)
4	Nano-hybrid (N=50)
	Brand of resin composites
I	Spectrum TPH3 (N=10)
2	Filtek Z250 (N=10)
3	Ceram X Duo (N=10)
4	Clearfil Majesty Esthetics ($N = 10$)
5	Premise (N=10)
6	Tetric Evo Ceram (N=10)
7	Filtek Supreme XT ($N=10$)
8	Esthet \times (N = 10)
9	Tetric N Ceram (N=10)
10	Gradia Direct (N=10)

correction was used as the equality of variances were not assumed) were performed at $\alpha = 0.05$ to test the differences in the changes in color (ΔE) and color parameters (ΔL , Δa , Δb) between brands of composites and differences between types of composites. A multiple regression analysis was used in order to determine the correlation among the color changes and changes in CIE L^* , a^* , and b^* values. A regression analysis with forward data input was selected (p = 0.05). When the tolerance between independent variables was lower than 0.3, the variable with the lower β -value (standardized correlation of each variable) was excluded from the regression to eliminate the impact of interrelated independent variables.¹²

RESULTS

The mean changes in color and color parameters, and standard deviations by the type and brand of resin

composites are presented in Tables 3 and 4, respectively. Ranges of ΔL^*1 , Δa^*1 , Δb^*1 , and ΔE^*1 were 12.54, 3.67, 13.04, and 9.54, respectively, after polymerization. Ranges of ΔL^*2 , Δa^*2 , Δb^*2 , and ΔE^*2 were 5.56, 1.37, 3.45, and 5.03, respectively, after storage in water. Nested ANOVA and post hoc tests revealed that the type of composite significantly affected Δa^*1 , Δa^*2 , Δb^*1 , Δb^*2 , ΔE^*2 values, whereas the brand of composite affected the changes in all color parameters (p < 0.05) (Tables 3 and 4).

Groups Assigned According to the Type of Composites

The a^* values decreased for the submicron-hybrid composite after polymerization whereas the others became reddish after polymerization and storage in water. The b^* values decreased in the submicronhybrid, nano-filled, and nano-hybrid composites after polymerization, and submicron-hybrid and nano-filled composites after storage in water, whereas it increased in the micro-hybrid composites after polymerization

TABLE 3. Mean changes and (SD) in color and color parameters of groups assigned according to the type of resin composites after polymerization and after storage in water

	After polymerization				After storage in water			
	∆L*I	∆a*I	∆b*l	∆E*I	∆ L*2	∆ a*2	∆ b*2	∆E*2
Submicron-hybrid composite	5.71ª (2.69)	-0.13ª (0.03)	-2.6 ^{a.b} (1.07)	6.64ª (1.80)	0.53ª (0.11)	0.45ª (0.12)	-0.4 ^{a.b} (0.73)	1.26 ^a (0.78)
Nano-filled composite	5.98ª (2.33)	I.II [♭] (0.27)	-0.09ª (0.03)	6.24ª (2.12)	1.38ª (0.36)	1.02 ^{ab} (0.12)	-0.22ª (0.10)	1.78 ^{a.b} (0.27)
Micro-hybrid composite	6.62ª (4.15)	1.53 ^b (0.43)	2.92⁵ (3.11)	7.88ª (4.71)	1.75ª (1.49)	1.05 ^{ab} (0.4)	I.83° (I.07)	3.16 ^b (1.42)
Nano-hybrid composite	7ª (3.97)	1.02 ^b (1,18)	-1.08 ^{a,b} (1.83)	8.63ª (2.5)	1.85ª (2.17)	1.26 ^b (1.18)	0.64 ^b (0.72)	2.92 ^b (2.05)
Different letters in the same column indicate significant differences between the mean values of study groups.								

TABLE 4. Mean changes and	(SD) in color and color par	ameters of groups assigned	l according to the brand o	of resin composites
after polymerization and after	storage in water			

	After polymerization					After storage in water			
	∆ <i>L</i> *I	∆a*I	∆ b *I	∆E*I	∆L* 2	∆a*2	∆ b*2	∆ E*2	
TPH	5.71 ^{a.b} (2.69)	-0.13ª (0.03)	-2.6 ^{a.} (1.07)	6.64ª (1.80)	0.53 ^{ab} (0.11)	0.45ª (0.12)	-0.4ª (0.73)	1.26 ^a (0.78)	
FZ2	3.83ª (1.86)	1.35 ^{b.c} (0.58)	1.96ª (1.23)	4.91 ^b (1.04)	2.41ªb (1.85)	0.83 ^{a.b.c} (0.49)	1.74 ^{b.c} (0.69)	3.44 ^{b.c} (1.42)	
CRX	10.87 ^{c.d} (2.37)	-0.93 ^d (0.34)	0.24 ^b (0.12)	11.06° (2.39)	1.19 ^{ab} (0.68)	0.95 ^{b.c} (0.25)	-0.53 ^{a.d} (1.59)	2.19 ^d (0.79)	
CME	9.69 ^{c.e} (1.40)	1.53 ^{c.e} (0.41)	2.18ª (1.25)	10.1° (1.62)	5.93° (1.36)	1.02 ^{b.c} (0.19)	1.79 ^{b.c} (0.55)	6.29° (1.40)	
PRM	5.14 ^{a.b} (1.34)	-0.49ª (0.16)	-3.22° (0.66)	6.48ª (1.18)	0.37ª (0.15)	1.82 ^d (0.28)	$0.26^{a.d}$ (0.84)	2.22 ^d (1.97)	
TEC	8.74° (1.77)	2.74 ^f (0.33)	1.1 ^{a.b} (0.2)	9.3° (1.75)	1.26 ^{a.b} (0.51)	1.28 ^{b.c} (0.23)	1.3 ^d (0.55)	2.31 ^b (0.39)	
FSX	5.98 ^b (2.33)	1.11 ^{b.d} (0.27)	-0.09 ^b (0.03)	6.24 ^{a.b} (2.12)	1.38 ^b (0.36)	1.02 ^b (0.12)	-0.22ª (0.10)	1.78 ^d (0.27)	
ESX	11.95 ^f (1.83)	1.45 ^{b.c.e} (0.39)	7.36 ^d (1.16)	14.13 ^d (2.04)	0.39 ^{ab} (0.18)	I ^{b.c} (0.35)	$0.82^{a.b.d}$ (0.25)	1.94 ^d (0.79)	
TNC	0.59 ^g (0.35)	2.27 ^g (0.21)	-5.68° (0.65)	6.23ª (0.61)	0.49 ^{ab} (0.12)	I.22 ^b (0.25)	0.38 ^{a.d} (0.13)	1.6 ^{a.d} (0.45)	
GRD	4.07 ^{a.b} (1.21)	1.78° (0.28)	-0.57 ^b (0.28)	4.59 ^b (1.02)	2.44 ^b (1.09)	1.32° (0.18)	2.92° (0.73)	4.11° (1.09)	
Different letters in the same column indicate significant differences between the mean values of study groups									

and micro-hybrid and nano-hybrid composites after storage in water.

The submicron-hybrid composite revealed significantly lower mean $\Delta a1^*$ value compared with the other composites, whereas its mean $\Delta a2^*$ value only differed from that of nano-hybrid composites. Nano-filled composites revealed significantly lower mean $\Delta b1^*$ value than micro-hybrid composites. Mean $\Delta b2^*$ values of nano-filled, nano-hybrid, and micro-hybrid composites ranged, respectively (p < 0.05), and changes in $b2^*$ value of the submicron-hybrid composite were significantly lower compared with the micro-hybrid composite. Mean $\Delta E2^*$ value of the submicron-hybrid composite was significantly different compared with that of micro-hybrid and nano-hybrid composites (Table 3).

Groups Assigned According to the Brand of Composites

The changes in color and color parameters after polymerization and storage in water varied significantly among the brand of resin composites (Table 4). CIE L^* value increased after polymerization in all resin composites except for TNC. CIE a^* value increased after polymerization except for CRX, TPH, and PRM. CIE b^* value decreased after polymerization except for FZ2, CRX, CME, TEC, and ESX. After storage in water, CIE L^* and a^* values increased in all resin composites. CIE b^* value increased except for CRX, TPH, and FSX.

After polymerization, the highest mean ΔL^*1 , Δb^*1 , and ΔE^*1 values were observed in the ESX group, whereas the highest mean Δa^*1 -value was observed in the TEC. The lowest mean ΔL^*1 , Δa^*1 , Δb^*1 , and ΔE^*1 values were observed in the TNC, TPH, FSX, and GRD, respectively. After storage in water, the highest mean ΔL^*2 and ΔE^*2 -values were observed in CME, whereas the highest mean Δa^*2 and Δb^*2 -values were obtained from the GRD. The lowest mean Δa^*2 and ΔE^*2 -values were observed in the TPH, whereas the lowest mean ΔL^*2 and Δb^*2 -values were obtained from the GRD. The lowest mean Δa^*2 and ΔE^*2 -values mean ΔL^*2 and Δb^*2 -values were obtained from the GRD. The lowest mean Δa^*2 and Δb^*2 -values were obtained from the GRD. The lowest mean ΔL^*2 and Δb^*2 -values were obtained from the GRD. The lowest mean ΔL^*2 and Δb^*2 -values were obtained from the GRD. The lowest mean ΔL^*2 and Δb^*2 -values were obtained from the GRD. The lowest mean ΔL^*2 and Δb^*2 -values were obtained from the GRD. The results of multiple regression analysis are presented in Table 5. ΔL^*1 and Δb^*1 were found to be significant predictors of color change after

TABLE 5. The correlation among the color change and changes in color parameters

Color change	The significant predictors of color change in order		
ΔE^* after polymerization	ΔL^* l after polymerization (β = 0.857), Δb^* l after polymerization (β = 0.410)		
	r*=0.932		
ΔE*2 after storage in water	ΔL^{*2} after storage in water (β =0.798), Δa^{*2} after storage in water (β =0.321), and Δb^{*2} after storage in water (β =0.127)		
	r=0.881		
β =standardized correlation of each variable; r=the product moment correlation between the dependent variable and the predicted values as generated by the multiple regression equation. It can assume values between 0 and 1.			

polymerization, whereas ΔL^*2 , Δa^*2 , and Δb^*2 were found to be significant parameters of color change after storage in water (p < 0.05).

DISCUSSION

Small color differences and their limits in terms of perceptibility and acceptability are of greatest importance.^{13,14} In the literature, although the data on acceptability and perceptibility limits are controversial, generally a ΔE^* value of 1 was found to be undetectable to 50% of the observers.¹⁵ A ΔE^* value of 2 or less was found to be in the acceptable clinical limits¹⁶; the 50:50 ΔE^* replacement point of esthetic dental materials was 2.7,¹⁷ whereas a $\Delta E^* \ge 3.7$ was found to be a poor color match value.¹⁸

Groups Assigned According to the Type of Composites

In this study, all type of composites revealed ΔE^* values greater than 3.7 after polymerization, whereas none of them had ΔE^* values higher than this limit after storage in water. Changes in color and color parameters were significant among the type of composites. No significant differences were observed among ΔE^* values of different composite types after polymerization, where

submicron-hybrid composite tested had lower ΔE^* values compared with nano-hybrid and micro-hybrid composites after storage in water. There were limited investigations regarding the effect of type of the resin composites on the color change after polymerization, although various investigations on the color changes of new composites after polymerization have been performed.^{2,3,7,10,19} In those studies the comparisons were generally made according to brands, in spite of a few earlier ones.9,10,20 The authors of these earlier investigations reported that microfilled composites showed more color change than semi-hybrid and conventional resin composites.^{9,10} On the other hand, some researchers found that microfilled composites showed superior color stability than that of conventional resin composites.²⁰ In this study, submicron-hybrid and nano-filled composites generally revealed lower ΔL^* , Δa^* , Δb^* , and ΔE^* values than nano-hybrid and micro-hybrid resins, although the differences were not significant at all times. The reason for the better color stability of both composites may be attributed to their organic filler size. All of the organic fillers of both composites were submicron and nanometer sized. However, there were differences in the sample size of each group when they were assigned according to the type of composites. This limitation should be considered during the evaluation of the results.

Groups Assigned According to the Brand of Composites

In this study, the L^* values decreased after polymerization for all resin composites except for TNC. The a^* values decreased for the CRX, TPH, and PRM, whereas the FZ2, CME, TEC, FSX, ESX, TNC, and GRD became reddish. The b^* values after polymerization decreased in the TPH, PRM, FSX, TNC, and GRD, whereas it increased in the FZ2, CRX, CME, TEC, and ESX. Different results were obtained from the literature; some investigators reported a decrease in L^* and in b^* values,^{2,3,19,21} and an increase in a^* values,²¹ In contrast, others reported an increase in L^* values,²¹ a decrease in a^* values,²¹ and a combined decrease/increase in b^* values.² The different results regarding the changes in color parameters of different brands of resin composites may be caused by the variations in the chemical compositions of the resin composites tested, such as the activator, initiator, inhibitor, or pigments.

The color changes after polymerization varied among the resin composites tested (p < 0.05). GRD and FZ2 revealed relatively small color changes, whereas the CME, CRX, and ESX groups showed greater color changes. However, all resin composites had polymerization dependent differences in color according to the ΔE^* formula greater than 3.7. The ΔE^* values obtained after polymerization were recorded as 4.59 to 14.13 and were greater than the reported polymerization-dependent differences in color.^{1,2,19,21,22}

Eldiwany and colleagues¹ evaluated the color stability of five resin composites (Clearfil, TrueVitality, Charisma, Conquest C&B and Herculite XRV) after light-curing and reported color differences ranging from 3.1 to 7.4. Yap and colleagues² investigated the color parameters of five different shades of Z100 (a mini-filled resin composite), Fuji II LC (a resin-modified glass-ionomer cement), and Dyract (a polyacid-modified resin composite) after postcuring and reported ΔE^* intervals of 3.3 to 6.1. Johnston and Reisbick¹⁹ evaluated various types of resin composites (Prisma APH, Charisma, Durafill VS, Helio Progress, Herculite XRV, PertacHybrid, Tetric, TPH, Z100) and glass ionomer restorative materials (Fuji LC, VariGlass VLC, Vitremer) of various shades to determine if the color differences occurred during curing and after storage in water and reported a ΔE^* interval of approximately 2.2 to 8.2. Paravina and colleagues²¹ investigated the curingdependent changes in color and translucency parameter values of resin composite bleach shades of micro-hybrid (3-D Direct, Esthet X, TPH Spectrum, Charisma, Point 4, Filtek Z250, Tetric Ceram, Synergy, Renamel Universal, Renew) and microfill composites (Durafill VS, Filtek A110 and Renamel Microfill), and reported color differences ranging from 3.7 to 12.0. Lee and colleagues²² studied the color changes after curing, polishing, and thermocycling of a nano-filled resin composite (Filtek Supreme) and reported a ΔE^* value of 3.27 to 11.7 for different shades. Based on the above studies, it can be assumed that most of the resin composites tested in different investigations revealed

relatively large amounts of change in optical properties. For this reason, these changes should be taken into account during any initial clinical color match selection. To be more prudent, an unpolymerized resin material of the selected shade should be placed and cured on, or adjacent to the undried tooth to confirm the shades of the esthetic restoratives before undergoing any restorative procedures.

The color stability of esthetic restorative materials is determined not only by more macroscopic phenomena such as matrix and filler composition and filler content, but also by relatively minor pigment additions and potentially by all other chemical components of these materials, including the resin initiator and inhibitor, activator progress, type, and quality of inhibitor, oxidation of unreacted carbon-carbon double bonds, ultraviolet light illumination and filler coupling agent.^{9,23} Direct grafting of the resin to the filler, provides optimal coupling and the resin composite will undertake lower water sorption.²⁴ Previous investigations have revealed that the optically observed color changes of resin composites were proportional to the time elapsed.^{24–27} In the present study, changes in color after storage in water were small (less than 3.7) and would reveal a clinically acceptable color shift, except for the CME and GRD. However, the degree of color changes after storage in water varied among the resin composites tested (p < 0.05). TPH and TNC revealed relatively small changes, whereas CME showed a greater color change. Chemical differences among the resin components, such as purity of the oligomers and monomers, and concentration/type of the activators, initiators, inhibitors, the oxidation of the unreacted carbon-carbon double bonds and fillers might have affected the results of color stability of the resin composites tested in this study.

In the present study the color measurements were carried out after storage in water at 37°C for 1 month. In the oral cavity, the influence of heat caused by hot drinks and hot food may be more noteworthy. In addition, the resin surface roughness was changed by the mastication process and the discoloring factors and deposits can stay longer on rough resin surfaces. Moreover, it is possible that resin composite

discolorations in the oral cavity might be greater than the results obtained in this study.

The degree of change of which color parameter influenced the most color changes was determined using a regression analysis. The results after polymerization indicated that ΔL^*1 and Δb^*1 positively influenced the color change (although five composites shifted toward blue region), whereas changes in CIE L^* parameters had the greatest influence on the overall polymerization color change. In previous studies, color changes of resin composites were negatively influenced by the changes of CIE L^* , a^* , or $b^{*, 2,22,23,27}$ except the study of Kim and Lee in which CIE L*and C_{ab}* positively influenced the color change after polymerization.²⁸ In addition, the power of the influence of the changes in CIE L^* , a^* , and b^* values on the color changes varied among different investigations.^{2,22,23,27,28} The reason for the different results from the different studies may be attributed to the study design and the tested materials. Some of the studies compared the different shades of restorative materials, whereas only one shade was used in the present study.^{2,22,28} The others used the earlier versions of composite resins tested in this study.^{23,27}

Changes in L^*2 , a^*2 , and b^*2 positively influenced the color changes after storage in water. It is generally thought that the color change of the resin composites in the oral cavity is caused by the increase of yellow and especially red color factors.²⁹ Similar with this observation, most of the resin composites tested in the present study changed to yellowish colors (except TPH, CRX, and FSX groups), whereas all of the resin composites tested became reddish after storage in water.

CONCLUSION

In spite of many improvements in chemical compositions and fillers of the contemporary composites, color changes after polymerization were perceptible in all resin composites tested here. Color changes after storage in water were acceptable in all resin composites except for CME and GRD. The large variations in the color change values of different resin composites after polymerization indicated that: (1) such changes should be taken into account when selecting a specific shade, and (2) a piece of unpolymerized resin material should be placed on, or adjacent to, the undried tooth and polymerized to confirm the selected shade of the material before undergoing any restorative procedures. A long-term in vitro study is required for further investigations regarding the color changes after storage in water as the color changes are thought to continue even after 1 month of storage. The null hypotheses tested were all rejected except for the amount of changes in L^* value after polymerization and after storage in water and changes in color after polymerization for different types of composites and for the correlation among the color changes and changes in CIE a^* values after polymerization.

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