Effect of staining solutions on discoloration of resin nanocomposites

Jeong-Kil Park, DDS, PhD, Tae-Hyong Kim, DDS, MS, Ching-Chang Ko, DDS, PhD, Franklin García-Godoy, DDS, MS, Hyung-Il Kim, DDS, PhD & Yong Hoon Kwon, PhD

ABSTRACT: Purpose: To examine the effect of staining solutions on the discoloration of resin nanocomposites. Methods: Three resin nanocomposites (Ceram X, Grandio, and Filtek Z350) were light cured for 40 seconds at a light intensity of 1000 mW/cm^2. The color of the specimens was measured in %R (reflectance) mode before and after immersing the specimens in four different test solutions [distilled water (DW), coffee (CF), 50% ethanol (50ET) and brewed green tea (GT)] for 7 hours/day over a 3-week period. The color difference (ΔE*) was obtained based on the CIEL*a*b* color coordinate values. Results: The specimens immersed in DW, 50ET and GT showed a slight increase in L* value. However, the samples immersed in CF showed a decrease in the L* value and an increase in the b* value. CF induced a significant color change (ΔE*: 3.1~5.6) in most specimens but the other solutions induced only a slight color change. Overall, coffee caused unacceptable color changes to the resin nanocomposites. (*Am J Dent* 2010;23:39-42).

CLINICAL SIGNIFICANCE: Within the limits of this study, coffee can induce an unacceptable color change in resin nanocomposites if used regularly for a long time.

✉: Prof. Yong Hoon Kwon, Department of Dental Materials, School of Dentistry, Pusan National University, Yangsan 626-870, Korea. E-✉: y0k0916@pusan.ac.kr

Introduction

Recently, advances in nanotechnology have led to several dental resin nanocomposites becoming available in dental clinics. One of the achievements in nanotechnology is the development of nanofillers ranging in size from 0.1 to 100 nm. The inclusion of nanofillers in resin composites has many advantages, such as increased filler content through decreased empty space within the resin matrix, increased continuity between the host material (teeth) and the restorative materials, and increased material strength and durability.[1-3] Since the size of nanofillers is much smaller than the incident blue light emitted from the light-curing unit, nanofillers tend to scatter or absorb less visible incident light, which can increase the translucency and esthetics of the resin nanocomposites.[4-6]

Discoloration of restorative materials is an unwanted effect on esthetic dental resin composites, even though it is inevitable in the oral environment. Discoloration can be caused by intrinsic and (or) extrinsic factors. Incomplete polymerization of the resin composites leaves unreacted monomers, which can cause discoloration by aging and sub-sequent reactions with other substances. Other components, such as initiators, fillers and pigments, can affect the color stability.[7-12] Since polymerization is achieved by light or heat, discoloration can occur from exposure to these stimu-li.[13,14] Discoloration can also occur through water sorption and food intake. Since the foods consumed contain a variety of coloring agents, they can alter the color of the resin composites through absorption and/or adsorption of colorants during the long period of exposure.[7,15] The surface roughness of the restoratives can also affect discoloration because a roughened surface has different surface dimensions, and dif-ferent contact rates with coloring agents.[16,17]

In order to test the staining effect of solutions on resin composites, a variety of beverages, such as water, coffee, wine, tea and soft drinks have been used.[18-22] The specimens showed a range of discoloration depending on the resin composites themselves and test solutions.[8,23-25]

This study tested the effect of various staining solutions (distilled water, ethanol, coffee, and brewed green tea) on discoloration of resin nanocomposites.

Materials and Methods

Three different resin nanocomposites [Ceram X^a (CX), Grandio^b (GD), and Filtek Z350^c (Z3)] were chosen for the study, and their characteristics are listed in Table 1. A quartz-tungsten-halogen (QTH) lamp-based unit (Optilux 501^d) was used for light curing. To prepare the specimens, a metal ring mold (2 mm in height with an inner diameter of 8 mm) was filled with resin. The top and bottom surfaces were then covered with a thin glass slide to make the surfaces flat. The specimens were light cured for 40 seconds under 1000 mW/cm^2 light intensity. The light-cured specimen was then removed from the mold and aged for 24 hours in a dark chamber at 37°C for the next measurement. The following four different test solutions were prepared:

Distilled water (DW);

Coffee (CF) - 0.45 g of instant coffee (Taster's Choice^e) powder/50 cc distilled water, without sugar or cream;

Green tea (GT) - 0.45 g of green tea leaves (Sulloc Cha^f)/ 50 cc distilled water. The green tea leaves were steeped in 80°C water for 10 minutes, and the leaves were then removed from the water.); and

50% Alcohol (50ET) - distilled water 25 cc + ethanol (99.9%) 25 cc.

The color change during staining with different solutions was measured using a spectrophotometer (CM-3600^g). Speci-mens (n= 5 for each test solution) of three different shades (M1, M2, M5 for CX; A1, A3, B2 for GD and Z3) were chosen and prepared using the procedures described before. Calibration was carried out according to the procedures of the system between 360 and 740 nm. After calibration, the initial color of the light-cured specimen was measured by placing

Table 1. Characteristics of resins tested in this study.

	Composition	Filler type	Filler* vol%/wt%
CX	Methacrylate modified polysiloxane dimethacrylate resin	Ba-Al-borosilicate glass, methacrylate functionalized SiO_2 nanofiller	57/76
GD	Bis-GMA, TEGDMA, UDMA	Ba-Al-borosilicate Glass filler, SiO_2 nanofillers	71.4/87
Z3	Bis-GMA, UDMA, TEGDMA, Bis-EMA	Non-aggregated zirconia/silica	59.5/78.5

CX: Ceram X; GD: Grandio; Z3: Filtek Z350
*: According to the manufacturers.

the specimen at the center of the target mask under %R (reflectance) mode. This target mask has a hole in the center, 7 mm in size. This hole maintains the consistency of specimen placement during the measurements.

After the first color measurement, the specimens were immersed in 1.5 ml of the test solutions for 7 hours and distilled water for 17 hours per day over a 3-week period. The test solutions were stored in a 37°C chamber and renewed daily. While changing the test solutions, the specimens were cleaned with running water without brushing. After 3 weeks, the specimens were removed from the test solutions and rinsed with running water. The remaining water was removed with tissue paper. The second measurement of the %R was performed immediately under the same conditions. Based on the measured %R data, the color values were evaluated according to the CIEL*a*b* color coordinate system using the internal software of the measurement system. The color difference, ΔE*, was obtained using the following equation:

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

where ΔL*, Δa*, and Δb* represent changes in L*, a*, and b*, respectively. Here, L* represents the degree of gray corresponding to a lightness, a* is the red (for + a* value) - green (for - a* value) axis, and b* is the blue (for - b* value) - yellow (for + b* value) axis.

The result of the color change was analyzed by a two-way ANOVA for the shade and test solution. A multiple-comparison was then performed using a Tukey's test. A P value < 0.05 was considered significant.

Results

After immersing the specimens for 3 weeks, those in DW, 50ET, and GT became slightly brighter (whiter). Depending on the product and shade, the L* value increased by 0~2.1. However, in CF, the L* value of the specimens decreased slightly, approximately 0.3~1.3. Among the test solutions, only CF made the specimens more yellow. The b* value increased 1.7~5.5 after immersion in CF depending on the product and shade. Among the specimens, GD showed the greatest change in b* (Table 2).

According to the two-way ANOVA, the shade and test solution in CX and GD were significant (P≤ 0.001). On the other hand, in Z3, only the test solution showed statistical significance (P< 0.001). After 3 weeks, the color change in

CX, GD, and Z3 ranged from 1.6~4.0, 0.8~5.6, and 2.0~3.4, respectively, depending on the test solution and shade (Table 3). In CX, only CF induced significant color changes. In GD, CF induced marked (appreciable) color changes (ΔE*: 3.1~5.6). The value was the highest among the test conditions examined. In the other solutions, a slight (ΔE*: 0.8) or perceivable (noticeable) (ΔE*: 1.7~2.7) color change was observed. In addition, in Z3, CF induced the greatest color change (ΔE*: 2.5~3.4) among the test solutions. In the same test solution, Z3 showed a similar color change regardless of the differences in shade.

Discussion

This study examined the influence of the type of test solution and shade of the product on the color of the resin nanocomposites. In CX and GD, the color change of the original CIEL*a*b* color values was affected significantly (P≤ 0.001) by the shade and type of test solution. On the other hand, Z3 was significantly affected only by the type of test solutions. In general, the color stability of a resin composite is affected by a variety of factors, such as resin matrix, initiator concentration, oxidation of unreacted monomers, filler loading and pigments.[7-13] According to Table 1, each resin product had different filler content. GD and CX showed the highest and lowest filler content, respectively. However, there was no correlation between the resulting color change and filler loading. No consistent tendency was observed in this study.

The color change in the resin nanocomposites may be related to the absorption and/or adsorption of the colorant contained in the test solution. The test solution containing CF produced the greatest color change in this study. Coffee has a strong staining effect on resin composites and natural tooth structures.[7,18] The brown coloring agent may be compatible with the polymer network. Therefore, the facilitated adsorption and penetration of the coloring agent produced the greatest color change in the specimens immersed in the coffee-containing solution. The specimens immersed in CF showed the largest change in b* value among the test solutions examined. A decrease in the L* value should be related to the adsorption and/or absorption of brown colorant. Tea has a naturally high tannin content, which is responsible for the significant color change in the resin composites after immersion in tannin-containing solutions.[19,20] However, according to the report, tea extracts do not contain tannin.[21] Brewed green tea has a thin green color. In most cases in the present study, the green colorant had no greening effect on the specimens regardless of product or shade. The original a* value changed more toward the direction of red (+a) than green (-a). The original L* value increased in all products and shades after immersion. This trend suggests that either the test solution does not contain tannin or the concentration of green colorant must be too low to be effective. The process is probably related not to the adsorption or absorption of green colorant, but to the dissolution of pigment(s) in the specimen. The same trend was observed in specimens immersed in transparent DW and 50ET solutions. An increase in L* value may be achieved by dilution of the original pigment within the specimen through absorption of the test solution. Alcohol was reported to roughen and degrade the surface resulting in increased staining.[10,22] How-

Table 2. CIEL*a*b* color coordinate values before and after immersion in test solutions for 3 weeks.

		Code	M1 L*	M1 a*	M1 b*	M5 L*	M5 a*	M5 b*	M2 L*	M2 a*	M2 b*
CX	A	0	58.1±0.1	-1.5±0.1	6.9±0.2	51.9±0.1	1.3±0.0	12.6±0.2	54.2±0.3	0.4±0.0	10.6±0.1
		3	59.6±0.2	-0.5±0.1	6.3±0.3	51.9±0.2	2.0±0.1	11.2±0.1	55.5±0.4	1.2±0.0	9.6±0.2
	B	0	58.2±0.5	-1.5±0.1	6.8±0.2	51.8±0.4	1.2±0.1	12.4±0.2	53.7±0.5	0.5±0.0	10.5±0.3
		3	57.5±0.4	-0.3±0.2	10.4±0.2	50.5±0.6	2.8±0.1	14.1±0.5	53.4±0.3	1.8±0.0	12.8±0.4
	C	0	57.8±0.1	-1.4±0.0	6.8±0.3	52.1±0.5	1.1±0.0	12.0±0.3	54.3±0.5	0.3±0.0	10.2±0.4
		3	59.6±0.3	-0.2±0.0	6.8±0.2	52.7±0.5	1.6±0.0	11.1±0.2	56.0±0.7	1.3±0.0	9.8±0.2
	D	0	57.9±0.4	-1.4±0.0	6.4±0.4	52.0±0.3	1.2±0.0	12.3±0.1	53.6±0.5	0.5±0.0	10.4±0.1
		3	59.1±0.4	-0.6±0.0	7.3±0.2	53.0±0.1	2.1±0.2	11.9±0.3	55.6±0.5	1.3±0.1	10.2±0.3
			A1			A3			B2		
GD	A	0	57.0±0.3	-2.3±0.0	0.7±0.1	52.9±0.7	-0.8±0.1	5.6±0.3	56.9±0.5	-2.8±0.1	3.2±0.2
		3	58.4±0.3	-1.7±0.6	1.6±0.3	53.4±0.5	-0.6±0.0	5.9±0.2	57.9±0.5	-2.2±0.0	5.1±0.2
	B	0	57.2±0.4	-2.2±0.0	0.7±0.2	52.0±0.2	-0.9±0.1	5.3±0.1	56.8±0.1	-2.8±0.1	3.3±0.2
		3	55.9±0.2	-1.6±0.0	5.8±0.5	51.5±0.3	-0.3±0.1	8.2±0.2	56.1±0.3	-1.9±0.2	8.8±0.7
	C	0	57.2±0.1	-2.2±0.1	0.3±0.1	52.6±0.2	-1.0±0.1	4.8±0.3	57.0±0.2	-2.7±0.0	3.5±0.2
		3	57.9±0.2	-1.8±0.1	2.1±0.2	53.5±0.3	-0.3±0.0	6.9±0.3	57.8±0.3	-2.4±0.1	5.4±0.3
	D	0	57.1±0.5	-2.2±0.1	0.6±0.2	52.4±0.4	-1.1±0.1	4.6±0.2	56.9±0.2	-2.7±0.0	3.6±0.3
		3	57.9±0.3	-2.5±0.2	3.1±0.5	53.2±0.5	-0.9±0.1	6.6±0.4	58.3±0.6	-2.7±0.1	5.7±0.2
Z3	A	0	59.8±0.2	-1.9±0.1	9.1±0.1	57.2±0.2	0.8±0.0	14.3±0.2	57.0±0.1	-0.8±0.0	11.8±0.3
		3	61.9±0.2	-0.7±0.1	8.3±0.2	59.2±0.3	2.1±0.0	14.6±0.2	59.0±0.3	0.3±0.0	12.7±0.2
	B	0	59.8±0.1	-1.9±0.1	9.1±0.2	57.0±0.4	0.9±0.1	14.4±0.3	56.9±0.4	-0.7±0.1	12.0±0.1
		3	59.0±0.5	-0.2±0.1	11.6±0.7	56.7±0.6	2.5±0.4	16.1±0.5	56.4±0.5	1.0±0.2	14.7±0.5
	C	0	59.7±0.4	-1.7±0.0	8.4±0.2	57.9±0.4	0.9±0.0	13.9±0.2	57.1±0.2	-0.7±0.0	12.1±0.1
		3	60.9±0.5	-0.6±0.1	8.6±0.0	59.1±0.6	2.2±0.1	14.9±0.2	58.6±0.2	0.4±0.0	13.0±0.1
	D	0	60.1±0.5	-1.8±0.0	8.7±0.3	57.6±0.4	0.8±0.1	14.1±0.4	57.0±0.3	-0.7±0.0	12.0±0.3
		3	61.6±0.2	-1.0±0.0	8.9±0.4	59.1±0.4	1.8±0.1	15.0±0.5	58.6±0.3	0.0±0.0	13.1±0.2

Code A: DW; B: CF; C: 50ET; D: GT; 0: Control (before immersion); 3: immersion for 3 weeks.

Table 3. Color differences in products for different shades and test solutions.

		DW[1]	CF[2]	50ET[1]	GT[1]	P-values
			Test solutions			
CX	M1[A]	1.9 ± 0.1	4.0 ± 0.3	2.1 ± 0.3	1.8 ± 0.3	α < 0.001
	M5[B]	1.6 ± 0.1	2.7 ± 0.4	1.2 ± 0.2	1.6 ± 0.3	β < 0.001
	M2[A]	1.9 ± 0.3	2.7 ± 0.5	2.1 ± 0.5	2.2 ± 0.7	
		DW[1]	CF[2]	50ET[3]	GT[3]	P-values
GD	A1[A]	1.7 ± 0.5	5.3 ± 0.5	2.0 ± 0.4	2.7 ± 0.5	α < 0.001
	A3[B]	0.8 ± 0.3	3.1 ± 0.2	2.5 ± 0.3	2.2 ± 0.2	β < 0.001
	B2[A]	2.2 ± 0.2	5.6 ± 0.7	2.1 ± 0.6	2.6 ± 0.4	
		DW[1]	CF[2]	50ET[3]	GT[13]	P-values
Z3	A1[A]	2.6 ± 0.2	3.4 ± 0.8	1.7 ± 0.1	1.9 ± 0.4	α = 0.37
	A3[A]	2.3 ± 0.1	2.5 ± 1.0	2.0 ± 0.3	2.1 ± 0.3	β < 0.001
	B2[A]	2.5 ± 0.3	3.3 ± 0.5	2.0 ± 0.3	2.2 ± 0.1	

* Statistically significant difference on shade is shown by superscript letters A, B, on test solution by superscript numbers.[1,2,3] Same letters or numbers are not significantly different (P< 0.05).
* On P-values, the letters α and β denote shade and test solution, respectively.

ever, in this study, no increased staining was observed. Different experimental conditions may be one of the reasons. Many studies have examined staining through a dynamic roughening process. However, the present study does not involve a dynamic roughening process. Specimen and solution were maintained in the container statically.

The test solutions in this study induced a different degree of color change in the specimens. DW, 50ET and GT induced a similar color change (in DW 0.8~2.6, in 50ET 1.2~2.5, and in GT 1.6~2.7 depending on product and shade). This was an acceptable degree range. On the other hand, CF induced a significantly different color change (ΔE^*: 2.5~5.6 depending on

product and shade). In most products, the degree range was unacceptable.

In conclusion, within the limits of this study, only CF induced an unacceptable (marked or appreciable) degree of color change. In contrast to the other test solutions, the L* value decreased and the b* value changed significantly after immersion in CF for 3 weeks due to the brown coloring agent. Regardless of the test solutions, the filler loading and copolymer of the monomers had no significant influence on the color change.

Dr. Park is Assistant Professor, Mr. T-H. Kim is a graduate student, Department of Conservative Dentistry, Dr. H-I. Kim is Professor, Dr. Kwon is Associate Professor, Department of Dental Materials, School of Dentistry and Medical Research Institute, Pusan National University, Yangsan, Korea. Dr. Ko is Associate Professor, Department of Orthodontics, School of Dentistry, University of North Carolina, Chapel Hill, North Carolina, USA. Dr. García-Godoy is Professor and Senior Executive Dean for Research, College of Dentistry, University of Tennessee, Memphis, Tennessee, and Senior Clinical Investigator, The Forsyth Institute, Boston, Massachusetts, USA.

References

1. Freitas RA Jr. Nanodentistry. *J Am Dent Assoc* 2000;131:1559-1565.
2. Davis N. A nanotechnology composite. *Compend Contin Educ Dent* 2003;24:662-670.

3. Baseren M. Surface roughness of nanofill and nanohybrid composite resin and ormocer-based tooth-colored restorative materials after several finishing and polishing procedures. *J Biomater Appl* 2004;19:121-134.

4. Kawaguchi M, Fukushima T, Miyazaki K. The relationship between cure depth and transmission coefficient of visible-light-activated resin composites. *J Dent Res* 1994;73:516-521.

5. Ure D, Harris J. Nanotechnology in dentistry: Reduction to practice. *Dent Update* 2003;30:10-15.

6. Kim JJ, Moon HJ, Lim BS, Lee YK, Rhee SH, Yang HC. The effect of nanofiller on the opacity of experimental composites. *J Biomed Mater Res Part B: Appl Biomater* 2007;80B:332-338.

7. Um CM, Ruyter IE. Staining of resin-based veneering materials with coffee and tea. *Quintessence Int* 1991;22:377-386.

8. Abu-Bakr N, Han L, Okamoto A, Iwaku M. Color stability of compomer after immersion in various media. *J Esthet Dent* 2000;12:258-263.

9. Eldiwany M, Friedl KH, Powers JM. Color stability of light-cured and post-cured composites. *Am J Dent* 1995;8:179-181.

10. Reis AF, Giannini M, Lovadino JR, Ambrosano GM. Effects of various finishing systems on the surface roughness and staining susceptibility of packable composite resins. *Dent Mater* 2003;19:12-18.

11. Eliades T, Gioka C, Heim M, Eliades G, Makou M. Color stability of orthodontic adhesive resins. *Angle Orthod* 2004;74:391-393.

12. Bagheri R, Burrow MF, Tyas M. Influence of food-simulating solutions and surface finish on susceptibility to staining of aesthetic restorative materials. *J Dent* 2005;33:389-398.

13. Janda R, Roulet JF, Kaminsky M, Steffin G, Latta M. Color stability of resin matrix restorative materials as a function of the method of light activation. *Eur J Oral Sci* 2004;112:280-285.

14. Ferracane JL, Moser JB, Greener EH. Ultraviolet light-induced yellowing of dental restorative resins. *J Prosthet Dent* 1985;54:483-487.

15. Scotti R, Mascellani SC, Formiti F. The *in vitro* color stability of acrylic resin for provisional restorations. *Int J Prosthodont* 1997;10:164-168.

16. Sarac D, Sarac YS, Kulunk S, Ural C, Kulunk T. The effect of polishing techniques on the surface roughness and color change of composite resins. *J Prosthet Dent* 2006;96:33-40.

17. Guler AU, Kurt S, Kulunk T. Effects of various finishing procedures on the staining of provisional restorative materials. *J Prosthet Dent* 2005;93:453-458.

18. Chan KC, Hormati AA, Kerber PE. Staining calcified dental tissues with food. *J Prosthet Dent* 1981;46:175-178.

19. Ertas E, Guler AU, Yucel AC, Koprulu H, Guler E. Color stability of resin composites after immersion in different drinks. *Dent Mater J* 2006;25:371-376.

20. Omata Y, Uno S, Nakaoki Y, Tanaka T, Sano H, Yoshida S, Sidhu SK. Staining of hybrid composites with coffee, oolong tea, or red wine. *Dent Mater J* 2006;25:125-131.

21. Wheeler SR. Tea and tannins. *Science* 1979;204:6-8.

22. Sarrett DC, Coletti DP, Peluso AR. The effects of alcoholic beverages on composite wear. *Dent Mater* 2000;16:62-67.

23. Iazetti G, Burgess JO, Gardiner D, Ripps A. Color stability of fluoride-containing restorative materials. *Oper Dent* 2000;25:520-525.

24. Gurdal P, Akdeniz BG, Sen BH. The effect of mouth rinses on micro hardness and color stability of esthetic restorative materials. *J Oral Rehabil* 2002;29:895-901.

25. Stober T. Gilde H, Lenz P. Color stability of highly filled composite resin materials for facings. *Dent Mater* 2001;17:87-94.