분광광도계를 이용한 커피와 인공타액에서의 수복용 복합레진의 변색에 대한 연구

서울대학교 치과대학 치과보존학교실

엄승희 · 조효선 · 이정식

Abstract

A SPECTROPHOTOMETRIC STUDY ON DISCOLORATION OF RESTORATIVE COMPOSITE RESINS IN COFFEE AND ARTIFICIAL SALIVA

Seung-Hee Eom, D. D. S., Hyo-Sun Cho, D.D.S.,M.S.D.,Ph.D., Chung-Sik Lee, D. D. S., M. S. D., Ph.D.,

Department of Conservative Dentistry, College of Dentistry, Seoul National Universtiy

The purpose of this study was to investigate by spectrophotometry the changes in color parameters of composite resins after exposure to distilled water, filtered coffee, and artificial saliva. Five kinds of fine particle composite resin in experiment 1, and six kinds of composite resin in experiment 2 were used. In experiment 1, each group of composite resin was stored in distilled water at 4℃, 50℃, and filtered coffee solution at 4℃, 50℃. And then each specimen was measured by spectrophotometer. Measurements were repeated in 1 hour, 2 hours, 4 hours, 8 hours, 16 hours, 24 hours, and 48 hours. In experiment 2, all specimens of each brand were stored in distilled water at 37℃, artificial saliva not contained mucin at 37℃, and artificial saliva contained mucin at 37℃.

All specimens of each brand were measured by spectrophotormeter in 1 week, 2 weeks, 4 weeks, 8 weeks, and up to 12 weeks.

The results were as follows :

1. In cold coffee, five of fine particle composite resins showed ΔE^*_{ab} values less than 2. However, in hot coffee ΔE^*_{ab} values of Amelogen Universal and Prisma TPH were higher than those of Conquest Crystal, Ælitefil and Z100.
2. Z100 and Ælitefil had better cleansibility of extrinsic coffee staining than Amelogen Universal, Conquest Crystal and Prisma TPH.

* 본 연구는 95년도 서울대학교 병원 지정진료 연구비 지원에 의해 이루어 졌음.

3. In distilled water and artificial saliva not contained mucin, five of fine particle composite resin showed ΔE^*_{ab} values less than 2 up to 12 weeks. However, Silux Plus showed higher than ΔE^*_{ab} values of fine particle composite resin.
4. As an immersion solution for discoloration experiment, artificial saliva not contained mucin showed similar appearance as distilled water. However, artificial saliva contained mucin had different appearance from the others.
5. In artificial saliva contained mucin, Ælitefil, Prisma TPH and Z100 showed ΔE^*_{ab} values less than 1 up to 12 weeks. However, Silux Plus, Amelogen Universal and Conquest Crystal showed ΔE^*_{ab} values more than 1 up to 12 weeks. But, Conquest Crystal showed different characteristics of chromacity difference value(less chromatic) and lightness difference value(lighter) from Silux Plus and Amelogen Universal.

Key words : Spectrophotometer, ΔE^*_{ab}, Chromacity, Lightness

Introduction

The potential for composite resins to discolor in oral enviornment has been well recognized.[1,2] A number of factors contribute to the color change of composite resins in clinical situation. Chemical instability of the resin matrix may lead to endogeneous color change. Also it may arise from surface staining, changes in the opacity of the material due to the alteration of the interface between the resin and filler which affects the light scattering of particle, and the discoloration of resin matrix. Moreover, the ability to stain may be potentiated by surface conditions of the composite, such as the roughness or the surface chemistry. The another factor to be considered is the ability to be cleansed.

Some of these factors have been studied by a number of workers to compare color differences for a number of resins and different techniques.[3,4,5] Asmussen demonstrated that the color change produced in 15 proprietary composite resins by storing for 1 month in water at 60℃ is well correlated with the color change obtained after storing for 12 months in water at 37℃.[3] The effects of thickness, background color, specular reflection, and surface roughness on the color of five commercial restorative resins were studied by reflection spectrophotometry by Powers.[5] Because the human eye was not very reproducible, the quantification of color of composite resins has been studied using the reflection spectrophotometry and the Munsell color tabs. The efforts of many workers have lead to the application of good methods of color measurement in dentistry.[6,7,8,9] The spectrophotometric method of color measurement was applied to an investigation of the color stability of dimethacrylate based composite resins exposed to thermal and photochemical aging.[6] Brauer determined by visual inspection of the specimens by 5 people with normal color vision. Comparisons were made in bright diffuse daylight using a white background.[7] Powers studied the color stability of commercial resins under conditions of accelerated aging by reflection spectrophotometry and visually with Munsell color tabs.[8]

The purpose of this study was to investigate by reflectance spectrophotometry the changes in color parameters of composite resins after exposure to distilled water, filtered coffee, and artificial saliva. The effects of materials, the temperature of storage solution, and the component of saliva were studied.

Material and method

Experiment 1

Specimens preparation

For each material(Amelogen Universal, Conquest Crystal, Ælitefil, Prisma TPH, and Z 100), twelve specimens were prepared. Each material was placed inside a acryl mold onto an underlying layer of Mylar strip lying on a glass slab. The surfaces were covered with Mylar strip and glass slab, the two glass slabs were then compressed with a C-clamp. Each specimen was light polymerized with Curing Light XL 1000(3M Dental Products, USA : light bulb 600mW) positioned both below and above the specimen for 1 minutes. Each specimen was removed from the mold, placed in distilled water and stored in the dark. After a storage of 48 hours at room temperature, the specimens were then polished on both sides with Emery paper No. 400, 600, 800, 1000, and 1200 to a final thickness of approximately 1.5 ㎜ and a diameter of 18 ㎜. Emery paper is harder than any fillers in the resins used and so far is clinically accepted without negative results.[10] Those specimens were placed in a distilled water in the dark at room temperature for 72 hours. The specimens were not exposed to daylight or other light sources during storage. As the formation of color is a process of oxidation, free access of air to the water was secured.[4] The six composite resins used in this study are presented

Table 1. The six composite resins investigated

Material	Shade & batch No.	Manufacturer	*Filler, mean size & Vol%	*matrix phase
Amelogen Universal	A3 UP# 0933	Ultradent	Barium Glass, 0.7μm(0.1−1.8), 60 Vol%	BIS-GMA based
Conquest Crystal	A2 LOT# 3219302	Jeneric/Pentron	Glass, 0.6μm(narrow), 65 Vol%	PCDMA based Crystalline polymer
Ælitefil	A3 LOT# 069024	Bisco	Barium Glass, 0.7μm, 66 Vol%	BIS-GMA, UDMA based PAGDMA
Prisma TPH	A3.5 LOT# 021009	Caulk/Dentsply	Barium Glass, fumed silica 0.8μm(0.04−3), 57 Vol%	BIS-GMA, UDMA based TEGDMA
Z100	5904b A3	3M	Zirconia/silica(monomodal) 0.6μm(0.01−3.5), 66 Vol%	BIS-GMA based
Silux Plus	No.5702 U	3M	Silica, 0.04μm, 40Vol%	

UDMA(Urethane Dimethacrylate)

PCDMA(Polycarbonate Dimethacrylate)

PAGDMA(Polyalkyleneglycol Dimethacrylate)

TEGDMA(Triethylenegylcol Dimethacrylate)

* The type of filler, mean size, Vol% of filler, and component of matrix phase were based on Information manuals or Directions for use of each brands.

in Table 1.

Initial color measurement

After this period, initial spectrophotometric measurements were recorded on each specimen. The specimens were removed from the storage water and brushed with a soft tooth brush to remove any bateria or fungoid growth and were then wiped dry and the color measured.

The color characteristics of each sample were measured by the use of a spectrophotometer(Color Eye 3000, Mcbeth, USA). The spectrophotomer system was connected to a computer system(soft ware : Optiview Version 1.3.7.). The wavelength range was from 360nm to 740nm. Reflectance spectra were taken with a black background, i.e. 0% reflectance, and for calibration purposes a white standard was used. The tristimulus values(X, Y, Z) relative to the CIE color matching functions for CIE standard Illuminant D65(as a simulated natural daylight) was computed at 20nm intervals. The standard observer, spectrophotomer-computer system, obtained the color data for a field of vision of 10nm. The appearance of the samples were characterized by means of the white-black L*, redness-greenness a*, and yellowness-blueness b* uniform color space (CIELAB), which were calculated from the tristimulus values.

Immersion in test solutions

To prepare the coffee, 60 g of 'Rosebud Coffee' was put on the filter paper and 1ℓ of boiled distilled water was poured through the coffee. This solution was filtered again through two of the same filter papers and distilled water was added to make 1..

The four test solution used in this study are presented in Table 2.

Color measurement

After the 30 minutes' immersion period, the third group of specimens and the fourth group were removed from storage solution. All specimens were rinsed with running water for 5 minutes and then wiped dry.

Each specimen was measured by spectrophotometer as the same method as initial color measurement. After first measurement (before cleansing) all specimens were cleansed with soft tooth brush thirty times and then the surfaces were wiped dry with cotton prior to a second measurement (after cleansing). Measurements were repeated in the 1 hour, 2 hours, 4 hours, 8 hours, 16 hours, 24 hours and 48 hours. The first group and second group were measured in 48 hours.

Each sample was marked for identification with diamond point on its top surface and it was repeatably placed by a constant surface alignment to the slit. After one surface of each sample was measured three times, the other surface of the sample was measured three times and mean values were gained.

Experiment 2

For each material(Amelogen Universal, Conquest Crystal, Ælitefil, Prisma TPH, Z100,

Table 2. Test solution used

First grup	Distilled water at 4℃ maintained in a refrigerator
Second grup	Distilled water at 50℃ maintained in a water bath
Third grup	Filtered coffee at 4℃ maintained in a refrigerator
Fourth grup	Filtered coffee at 50℃ maintained in a water bath

Table 3. Test solution used

First grup	Distilled water at 37℃ in the dark
Second grup	Artificial saliva at 37℃ in the dark (without mucin)
Third grup	Artificial saliva at 37℃ in the dark (with mucin)

Table 4. Composition of synthetic artificial saliva

KCl	0.4 g/ℓ
NaCl	0.4 g/ℓ
$Mg_2P_2O_7$	0.0016 g/ℓ
Na_2HPO_4	0.6 g/ℓ
Na_2S	0.0016 g/ℓ
urea	1.0 g/ℓ
mucin	4.0 g/ℓ

and Silux Plus), nine specimens were prepared.

Specimen preparation and initial color measurement of experiement 2 were such as those of experiment 1.

Immersion in test solutions

The three test solution used in experiment 2 are presented in Table 3.

The composition of synthetic artificial saliva in this study are presented in Table 4.

Color measurement

All specimens of each brand were measured by spectrophotometer in 1 week, 2 weeks, 4 weeks, 8 weeks, and up to 12 weeks.

Color changes are determined for ΔL^*, which is the change in lightness or darkness, Δa^*, which is a change in hue along the red/green scale, and Δb^*, which is the change in hue along the yellow/blue scale. The total color differences are expressed by the formulation.

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

Perceived lightness differences, ΔL^* and a change in chroma, ΔC^*_{ab} were analyzed.

$$\Delta C^*_{ab} = \sqrt{(\Delta a^*)^2 + (\Delta b^*)^2}$$

Result

Experiment 1

Data obtained from the measurements are presented in Table 5 and Figure 1~5.

Figure 1 and 2 illustrate the effect of coffee at 50℃ on the test resins of each brand before cleansing and after cleansing. All resins showed an increase in ΔE^*_{ab} from 30 minutes to 48 hours. Before cleansing, until 24 hours Amelogen Universal showed the largest ΔE^*_{ab} and Prisma TPH showed relatively large ΔE^*_{ab}. At 24 hours Conquest Crystal and Z100 showed the least ΔE^*_{ab}. At 48 hours Conquest Crystal showed the least ΔE^*_{ab}. After cleansing, all of the resins showed a decrease in ΔE^*_{ab}. After up to 48 hours Amelogen Universal showed the most color change. At 24 hours Z100 showed the least color change($\Delta E^*_{ab}<3$). At 48 hours Conquest Crystal, Ælitefil, Z100 showed the small ΔE^*_{ab} values($\Delta E^*_{ab}<6$) in comparison to the values of Amelogen Universal and Prisma TPH.

Figure 3 and 4 illustrate the effect of coffee at 4℃ on the test resins of each brand before cleansing and after cleansing. All resins showed a very small increase in ΔE^*_{ab}. After cleansing ΔE^*_{ab} was the smaller than that before cleansing. However, before and after cleansing all specimes showed a very small ΔE^*_{ab}($\Delta E^*_{ab}<2$).

Table 5. Color difference of composite resins in coffee solutions.

Amelogen universal	0.5	0.5 b	1	1 b	2	2 b	4	4 b
cold coffee	1.087	1.922	1.220	1.061	1.817	1.749	1.203	1.126
hot coffee	1.262	1.016	2.735	2.226	3.604	3.683	4.495	3.846
Conquest Crystal	0.5	0.5 b	1	1 b	2	2 b	4	4 b
cold coffee	1.023	0.899	1.167	1.219	2.002	2.204	1.329	1.757
hot coffee	1.129	1.211	1.642	2.223	2.585	2.263	3.161	2.914
Ælitefil	0.5	0.5 b	1	1 b	2	2 b	4	4 b
cold coffee	1.139	1.025	1.467	1.065	1.237	1.608	1.285	1.360
hot coffee	1.138	1.25	2.658	1.975	2.787	3.702	3.256	3.742
Prisma TPH	0.5	0.5 b	1	1 b	2	2 b	4	4 b
cold coffee	1.018	0.68	0.793	0.756	0.682	0.782	0.787	0.711
hot coffee	0.89	0.692	2.354	2.292	3.132	3.085	3.816	3.690
Z100	0.5	0.5 b	1	1 b	2	2 b	4	4 b
cold coffee	0.991	1.211	1.791	1.398	1.675	0.795	2.125	1.385
hot coffee	0.659	1.075	2.500	2.054	2.875	2.603	3.621	3.262

Amelogen universal	8	8 b	16	16b	24	24	48	48 b
cold coffee	1.398	1.111	1.325	1.357	1.684	1.524	1.815	1.747
hot coffee	5.163	5.306	5.996	5.688	6.992	7.045	8.284	8.097
Conquest Crystal	8	8 b	16	16b	24	24	48	48 b
cold coffee	1.026	0.971	1.902	1.429	1.417	1.947	1.253	1.643
hot coffee	3.028	3.200	4.414	4.845	4.772	5.036	5.335	5.336
Ælitefil	8	8 b	16	16b	24	24	48	48 b
cold coffee	0.933	0.848	1.021	0.974	1.065	1.392	1.42	1.368
hot coffee	3.770	3.872	5123	4.319	4.854	6.058	8.041	5.545
Prisma TPH	8	8 b	16	16b	24	24	48	48 b
cold coffee	0.512	0.576	0.843	1.017	1.292	0.961	1.48	1.553
hot coffee	3.900	3.917	5.262	5.291	6.412	5.83	7.89	6.805
Z100	8	8 b	16	16b	24	24	48	48 b
cold coffee	1.007	0.943	1.226	1.181	1.29	1.224	1.565	1.596
hot coffee	3.969	3.990	4.073	5.287	4.472	2.43	7.564	5.348

Figure 5 illustrates the effect of Distilled water at 4℃ and 50℃ on the test resins of each brand at 48 hours. Although ΔE^*_{ab} values of control specimens did illustrate a change, none of the values showed more than 0.5.

After 2 hours' immersion, the specimens of Amelogen Universal in hot coffee displayed ΔE^*_{ab} values of more than 3.3. Finally after 48 hours' immersion ΔE^*_{ab} values of Amelogen Universal showed values over 8.0. The cleansing of specimens removes some of the surface staining. However, residual staining became cumulative. In cold coffee, after 48 hours, no ΔE^*_{ab} values of specimens were displayed more than 2.

The specimens of Conquest Crystal displa-

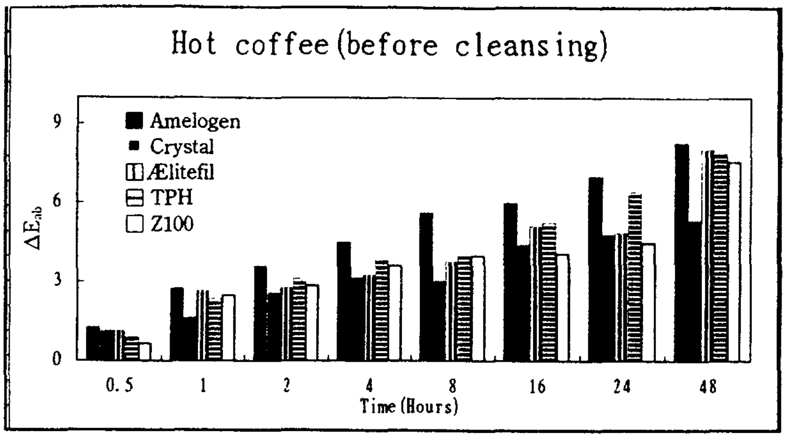

Fig. 1 ΔE^{*}_{ab} values of five composite resins in hot coffee (before cleansing).

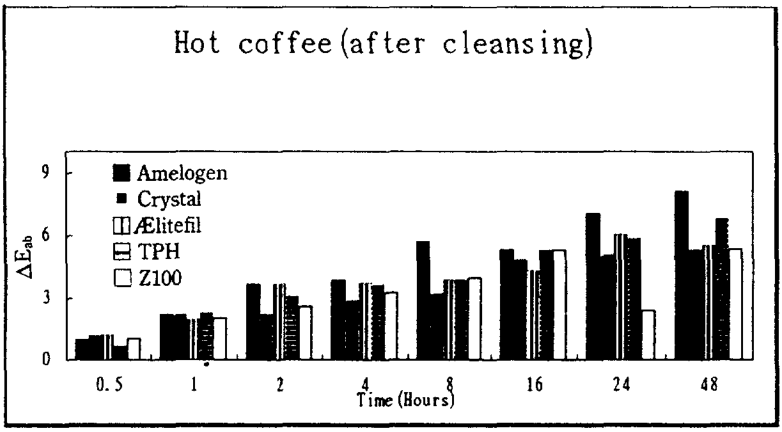

Fig. 2 ΔE^{*}_{ab} values of five composite resins in hot coffee (after cleansing).

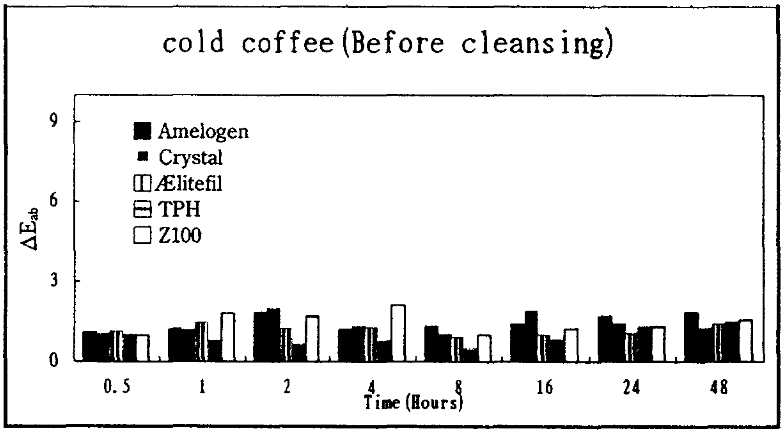

Fig. 3 ΔE^{*}_{ab} values of five composite resins in hot coffee (before cleansing).

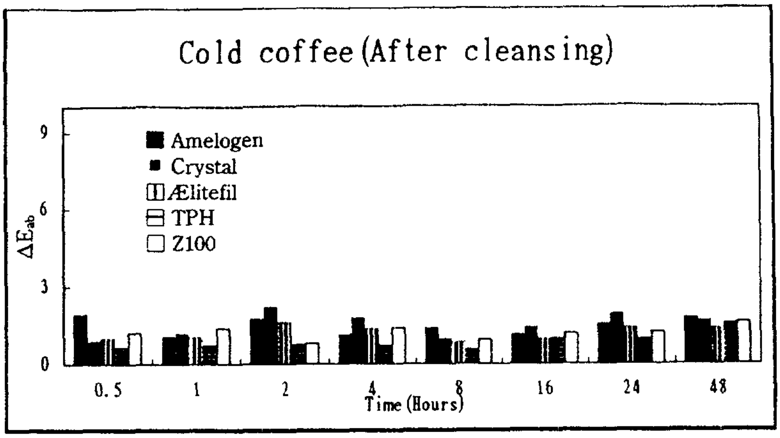

Fig. 4 ΔE^*_{ab} values of five composite resins in hot coffee (after cleansing).

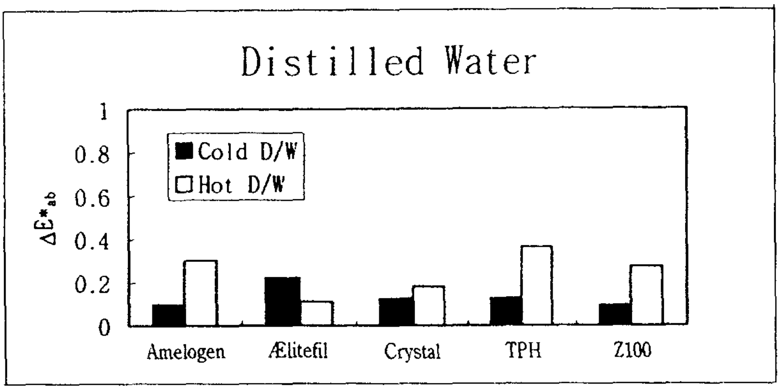

Fig. 5 Effect of distilled water at 48 hours.

yed ΔE^*_{ab} values of more than 3.3 in hot coffee after 4 hours. Finally after 48 hours' immersion ΔE^*_{ab} values showed approximately value of 5.3. The effect of cleansing on ΔE^*_{ab} values was minimal. In cold coffee, some of values were more than 2.

After 4 hours' immersion, the specimens of Ælitefil displayed ΔE^*_{ab} values of more than 3.3 in hot coffee. Finally after 48 hours' immersion ΔE^*_{ab} values showed approximately value of 8.0. However, the cleansing drop ΔE^*_{ab} values down to 5.545. In cold coffee, for 48 hours, ΔE^*_{ab} values of specimens displayed less than 2.

After 4 hours' immersion, the specimens of Prisma TPH displayed ΔE^*_{ab} values of more than 3.3 in hot coffee. Finally after 48 hours' immersion ΔE^*_{ab} values showed approximately value of 7.9. In cold coffee, for 48 hours, ΔE^*_{ab} values of specimens were displayed less than 2. For 24 hours, the color change of Z100 was the most stable. After 4 hours' immersion, the specimens displayed ΔE^*_{ab} values of more than 3.3 in hot coffee. Finally after 48 hours' immersion ΔE^*_{ab} values showed high value. However, the cleansing drop ΔE^*_{ab} values down to 5.348. In cold coffee, for 48 hours, ΔE^*_{ab} values of specimens were displayed less than 2.

Experiment 2

The total color difference, ΔE^*_{ab}, values for all of composite resins up to 12 weeks in distilled water at 37℃ and darkness is presented in figure 6. Corresponding values in artificial saliva not contained organic mucin and in artificial saliva contained mucin are presented in figure 9 and figure 12. From the results presented in figure 6 it is evident that the color stability of Silux Plus is significantly different from that of the other materials. ΔE^*_{ab} of high value in figure 6 reveals increasing discoloration of material. The discoloration of Silux Plus after 12 weeks is partially explained by increasing chromacity difference(more chromatic), ΔC^*_{ab}, values in figure 7 and negatively decreasing lightness difference(darker), ΔL^*, values, presented in figure 8. The chromacity difference is a measure of chroma of specimen after immersion in comparison with initial measure, and negative chromacity difference is a reflection of bleaching of specimens. Chromacity difference, ΔC^*_{ab}, values for Ælitefil and Prisma TPH were negative.

A chromacity difference, ΔC^*_{ab}, value for Z100 was approximately zero. A lightness difference, ΔL^*, value for only Prisma TPH was positive. It means that the specimens of Prisma TPH after immersion were lighter than before immersion.

From the results presented in figure 9 it is evident that the color stability of Silux Plus, Conquest Crystal, and Prisma TPH are different from that of the other materials. The discoloration of Silux Plus at 8 weeks is explained by increasing chromacity difference, ΔC^*_{ab}, values in figure 10. However, a high ΔE^*_{ab} value for Prisma TPH is explained by negatively decreasing chromacity difference(less

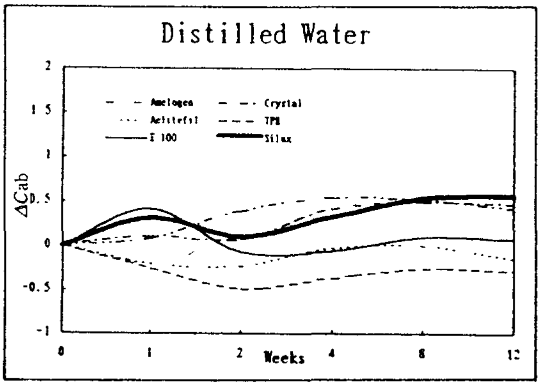

Fig. 7 ΔC^*_{ab} values of six composite resins in distilled water at 37℃.

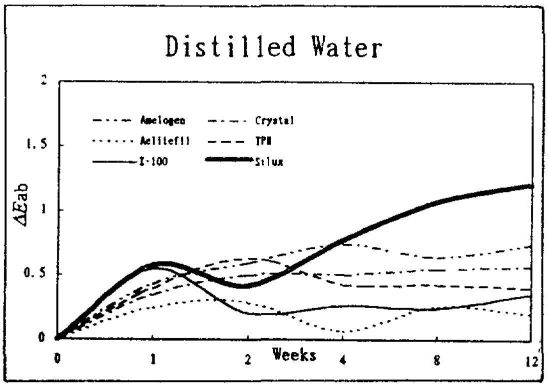

Fig. 6 ΔE^*_{ab} values of six composite resins in distilled water at 37℃.

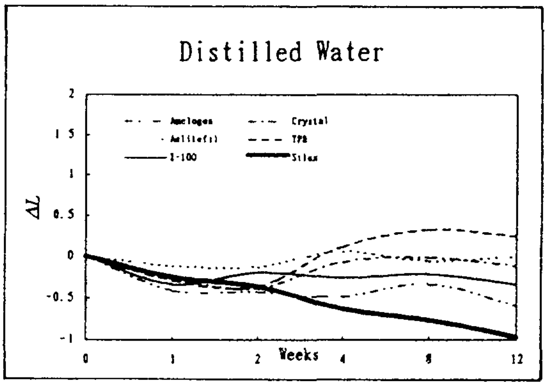

Fig. 8 ΔL^*_{ab} values of six composite resins in distilled water at 37℃.

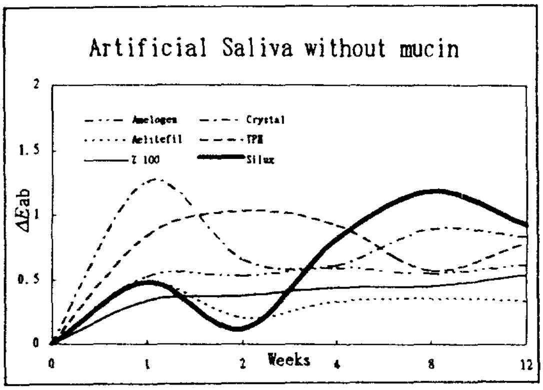

Fig. 9 ΔE^*_{ab} values of six composite resins in artificial saliva not contained mucin at 37℃.

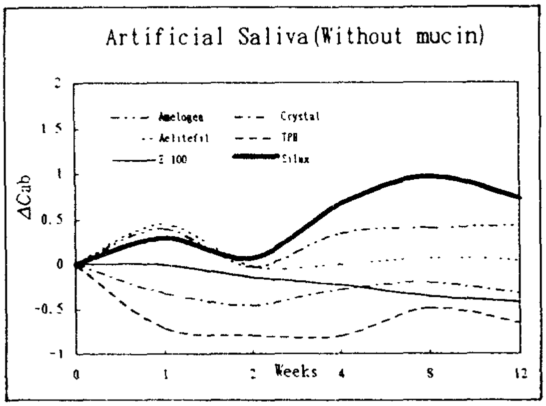

Fig.10 ΔC^*_{ab} values of six composite resins artificial saliva not contained mucin at 37℃.

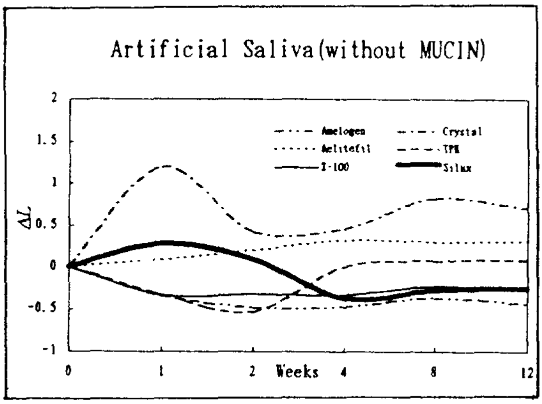

Fig.11 ΔL^*_{ab} values of six composite resins artificial saliva not contained mucin at 37℃.

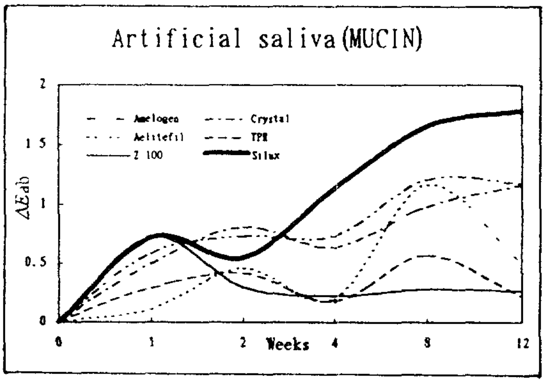

Fig.12 ΔE^*_{ab} values of six composite resins artificial saliva not contained mucin at 37℃.

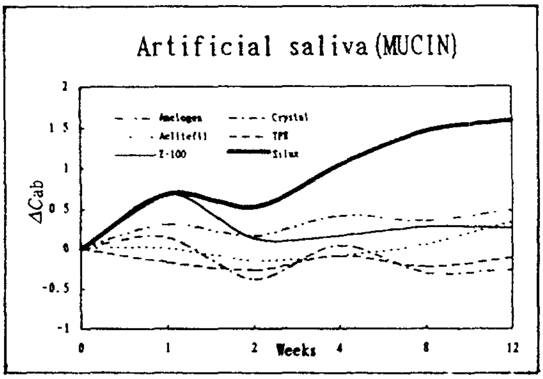

Fig.13 ΔC^*_{ab} values of six composite resins artificial saliva not contained mucin at 37℃.

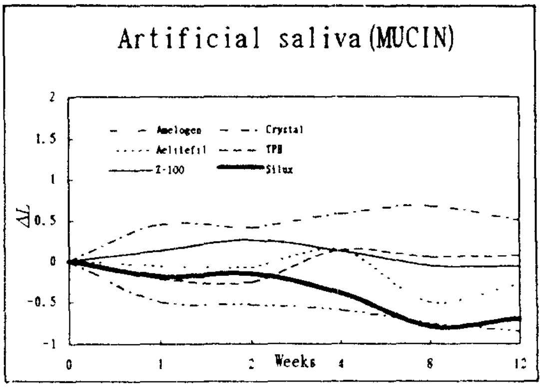

Fig.14 ΔL^*_{ab} values of six composite resins artificial saliva not contained mucin at 37℃.

chromatic), ΔC^*_{ab} values. Chromacity difference, ΔC^*_{ab}, values for Conquest Crystal, Prisma TPH, and Z100 were negative. A high ΔE^*_{ab} value for Conquest Crystal at 1 week is explained by negatively decreasing chromacity difference(less chromatic), ΔC^*_{ab}, values and increasing lightness difference, ΔL^*, values(lighter) presented in figure 11. Lightness difference, ΔL^*, values for Amelogen Universal and Z100 were negative.

From the results presented in figure 12 it is evident that the color stability of Silux Plus, Amelogen Universal, and Conquest Crystal are different from that of the other materials. The discoloration of Silux Plus and Amelogen Universal is explained by increasing chromacity difference, ΔC^*_{ab}, values in figure 13 and decreasing lightness difference, ΔL^* values (darker) presented in figure 14. However, a high ΔE^*_{ab} value for Conquest Crystal is explained by negatively decreasing chromacity difference, ΔC^*_{ab}, values and increasing lightness. Chromacity difference, ΔC^*_{ab}, values for Conquest Crystal and Prisma TPH were negative. A high ΔE^*_{ab} value for Z100 at 1 week is explained by increasing chromacity difference, ΔC^*_{ab}, value. However, from 2 weeks chromacity difference, ΔC^*_{ab}, value for Z100 decreased, and was stable up to 12 weeks. It is different from that for Silux Plus. Lightness

Table 6. Color difference of six composite resins in artificial saliva.

Amelogen	0	1	2	4	8	12
D/W	0	0.424	0.586	0.736	0.638	0.732
no mucin	0	0.523	0.529	0.59	0.547	0.613
mucin	0	0.584	0.728	0.727	1.205	1.175
Conquest Crystal	0	1	2	4	8	12
D/W	0	0.344	0.495	0.499	0.544	0.556
no mucin	0	1.252	0.655	0.609	0.888	0.827
mucin	0	0.495	0.805	0.629	0.977	1.158
Ælitefil	0	1	2	4	8	12
D/W	0	0.245	0.284	0.064	0.255	0.195
no mucin	0	0.474	0.2	0.329	0.353	0.332
mucin	0	0.104	0.455	0.192	1.163	0.492
Prisma TPH	0	1	2	4	8	12
D/W	0	0.391	0.628	0.425	0.419	0.397
no mucin	0	0.832	1.029	0.92	0.57	0.776
mucin	0	0.284	0.415	0.172	0.563	0.215
Z100	0	1	2	4	8	12
D/W	0	0.548	0.206	0.266	0.245	0.35
no mucin	0	0.339	0.377	0.441	0.448	0.54
mucin	0	0.726	0.296	0.222	0.28	0.264
Silux Plus	0	1	2	4	8	12
D/W	0	0.57	0.414	0.773	1.671	1.207
no mucin	0	0.476	0.117	0.816	1.18	0.928
mucin	0	0.719	0.548	1.134	1.653	1.788

difference, ΔL^*, values for Amelogen Universal and Silux Plus were negative.

The total color difference, ΔE^*_{ab}, values for all of composite resins up to 12 weeks in distilled water, artificial saliva not contained organic mucin and in artificial saliva contained mucin at 37℃ and darkness are presented in table 6.

Discussion

As regarding the question of perceptibility, ΔE^*_{ab} values of 0~1 will be referred to as imperceptible, and values in the range of 1~2 will be referred to as just perceptible by human eye.[11] As regards unacceptible color differences, Ruyter et.al., working with the CIELAB color space, found that ΔE^*_{ab} values above approximately 3.3 were beyond acceptibility in subjective visual evaluation.[9] Of course, correlations between clinical studies and laboratory measurements are difficult to establish, because many factors are at play in the in vivo situation. In general, materials having ΔE^*_{ab} values above 7 will show a marked discoloration in the mouth, whereas materials with ΔE^*_{ab} values below 2 will be relatively color stable in the clinical situation.[3,12]

The effect of the immersion solutions on color change

Microfilled composite resins had worse color stability than fine particle blended composite resins. Microfilled composite resins have a higher content of polymer, which may undergo color change.[2,13] According to C. Ameye et. al., in vivo after 18 months extrinsic staining of composite resins was not a major problem after the introduction of the microfiller resins because they have smooth surface characteristics.[1] Whereas Leinfelder K. F. et. al., reported the opposite observation that conventional composite resins displayed better color stability than microfills.[2] According to Dijken JWV van, clinically thirty percent of patients showed extrinsic discoloration during 6 years, and the stain could easily be polished off.[12]

Figure 1~5 and table 5 show the comparative staining effects of distilled water and filtered coffee immersion procedures. From these it may be concluded that for all materials coffee produced significantly greater color changes than distilled water. Marked discoloration may be more affected by sorption of dietary colorants than by water and sunlight.[14] The small color changes occuring in distilled water may be due to the thermal effect of the immersion temperature of 50℃.[15] The BIS-GMA is not quite color stable, and tends to turn yellow. The thermal energy could be sufficient to cause enough decomposition of the BIS-GMA resin leading to discoloration. According to I. E. Ruyter, the color stability is reduced with increasing amounts of BIS-GMA because of the reduction in the degree of conversion.[13] In contrary, according to E. Asmussen, the color change decreases with increasing amount of BIS-GMA in the monomer.[4]

The effect of temperature on color changes

After immersion in distilled water, the differences in mean ΔE^*_{ab} between 4℃ and 50℃ were small. These differences in ΔE^*_{ab} are in the imperceptible region. These small differences do not seem to reflect potential changes in color difference due to geometric or spectral metamerism.[15] It has been reported that the rate of discoloration in distilled water increases with increasing temperature.[3]

While no detailed mechanism for staining has been proposed, color changes due to staining might be due to the absorption of extraneous colored materials. The increase in temperature up to 50℃ may have increased the amount of water uptake by the specimens. This can have facilitated the absorption of staining particles in coffee.[15] Another explanation that may have resulted in the surface uptake of staining particles can be that of surface porosity resulting from a dissolution of slightly soluble components of the material and air inhibition zones of unpolymerized materials.[16] The degree of conversion after polymerization will also affect the color change. Residual double bonds in materials make them less resistant to degradation.[13]

The effect of surface cleansing on color changes

After the specimens stained with filtered coffee were cleansed, the differences in ΔE^*_{ab} between the cleansed and the uncleansed were small. Data from experiment 1 showed that in hot coffee ΔE^*_{ab} values of Amelogen Universal and Prisma TPH were higher than those of Conquest Crystal, Ælitefil and Z100. Z100 and Ælitefil had better cleansibility of extrinsic coffee staining than Amelogen Universal, Conquest Crystal and Prisma TPH. This phenomenon is not fully understood, but one explanation is that according to Douglas, hydrophobic composites had less staining capacity and greater ease of stain removal.[17] According to Satou *et. al.,* staining of resins in low hydrophobic solution occurs by impregnation of staining contaminant with water sorption, and hydrogen bonding also seems to contribute to the staining process. On the other hand, in the highly hydrophobic solution, a positive relationship is observed between the contact angle of the resin and the ΔE^*_{ab} of resin. Since contact angle is an index of hydrophobicity, hydrophobic interaction may play an important role in the staining of resins.[18] Another explanation may be that according to Um, removable discoloration will be probably due to adsorption of the polar colorants from coffee at the surface of the materials. The discoloration from coffee was due to surface adsorption and absorption of colorants.[19] The less polar colorants from coffee may be penetrated deeper into the material, probably because the coffee colorants are compatible with the polymer matricies of specimens Amelogen Universal and Prisma TPH.

The effect of artificial saliva on discoloration

The resins contained an amine are discolored by reacting with oxygen, however, resins without an amine are discolored. It was thought that surface staining contributed to clinical discoloration. Resins with high water sorption are assumed to be more liable to staining by penetration.

It is not known to increase discoloration of composite resins by saliva. According to ZA Khokhar *et. al.,* the addition of chlorhexidine and saliva increased staining when used with common dietary colorants.[16] Restorative resins are susceptible to softening caused by organic acids produced in oral enviornment. Consequently, resin may be liable to pronounced surface staining. In clinical part of the study a relationship between surface staining and local oral hygiene has been demonstrated. It can be assumed that the softening and erosion by organic acid in oral enviornment imparts to the polymers a more open structure which facilitates the adsorption. Microfilled composites and fine grained composites contain a relatively large amount of TEGDMA in the monomer for viscosity control in comparison to more coarse grained composite resins. It has been stated that polymers originating from low TEGDMA content presented increased surface staining due to increased surface softening.[20] In distilled water and artificial saliva not contained mucin, five of fine particle composite resin showed ΔE^*_{ab} values less than 2 up to 12 weeks. However, Silux Plus showed higher than ΔE^*_{ab} values of fine particle composite resin. In artificial saliva contained organic mucin, Ælitefil, Prisma TPH and Z100 showed ΔE^*_{ab} values less than 1 up to 12 weeks. However, Silux Plus, Amelogen Universal and Conquest Crystal showed ΔE^*_{ab} values more than 1 up to 12 weeks. But, Conquest Crystal showed different characteristics of chromacity difference value(less chromatic) and lightness difference value(lighter) from Silux Plus and Amelogen Universal. However, the change were, in absolute values, lower than ΔE^*_{ab} of 3.3.

Increased filler levels result in better color stability, lower solubility, and lower water sorption.[9,11] The water absorbed by matrix can cause filler-matrix debonding or degradation, and there may also be some influence of the hydrophobicity of resin matrix. Air voids incorporated may lead to inhibition zones with unpolymerized material, which may result in higher water solubility.[22] Because of lower filler content and higher matrix content of microfilled composite(Silux Plus), they will exhibit greater water sorption, lower elastic moduli and higher coefficiency of thermal expansion than universal fine particle composite.

The composite resins are generally darker, more chromatic, and more opaque.[5] However, according to Power *et.al.*, recently developed posterior composite subjected to accelerated aging for several days became lighter and less chromatic color than the main body of the material, or even become whiter than initially.[23] This phenomenon is due to degradation of the surface which provides appreciable light scattering. Light scattering will occur due to the particles in composite resin. Microfilled composite resin may be less light scattered than fine particle composite resin, probably because the wave length of the light is more than the diameter of the prepolymerized microfiller particles and microfilled composite resins have less filler content than fine particle composite resins. Data from experiment 2 showed that in all of the solution after 4 weeks the specimens of Prisma TPH became lighter than intially and in artificial saliva Conquest Crystal became up to 12 weeks increasingly lighter than initially. Specimens of Silux Plus became darker than initially. Prisma TPH and Conquest Crystal tended to become less chromatic than the other material and initially. This phenomenon was also observed by Powers *et. al.*, and others and may be due to enhanced scattering by the filler particles, resulting from debonding of the resin-filler interface.[23,24] Also the difference may reflect a difference in composition of these materials. Further analytical studies of the materials may be needed how compositional differences influence the color stability.

Conclusion

1. In cold coffee, five of fine particle composite resins showed ΔE^*_{ab} values less than 2. However, in hot coffee ΔE^*_{ab} values of Amelogen Universal and Prisma TPH were higher than those of Conquest Crystal, Ælitefil and Z100.
2. Z100 and Ælitefil had better cleansibility of extrinsic coffee staining than Amelogen Universal, Conquest Crystal and Prisma TPH.
3. In distilled water and artificial saliva not contained mucin, five of fine particle composite resin showed ΔE^*_{ab} values less than 2 up to 12 weeks. However, Silux Plus showed higher than ΔE^*_{ab} values of fine parti-

cle composite resin.

4. As an immersion solution for discoloration experiment, artificial saliva not contained mucin showed similar appearance as a distilled water. However, artificial saliva contained mucin had different appearance from the others.
5. In artificial saliva contained mucin, Ælitefil, Prisma TPH and Z100 showed ΔE^*_{ab} values less than 1 up to 12 weeks. However, Silux Plus, Amelogen Universal and Conquest Crystal showed ΔE^*_{ab} values more than 1 up to 12 weeks. But, Conquest Crystal showed different characteristics of chromacity difference value(less chromatic) and lightness difference value(lighter) from Silux Plus and Amelogen Universal.

References

1. Cathy Ameye, Paul Lambrechts, & Guido Vanherle: Coventional and microfilled composite resins. Part I : Color stability and marginal adaptation. *J Prosthet Dent* 1981 ; 46 : 623−630.
2. D. C. Crumpler, H. O. Heymann, D. A. Shugars, & K. F. Leinfelder : Five-year clinical investigation of one conventional composite and three microfilled resins in anterior teeth. *Dent Mater* 1988 ; 4 : 217−222.
3. Erik Asmussen : An accelerated test for color stability of restorative resins. *Acta Odontol Scand* 1981 ; 39 : 329−332.
4. Erik Asmussen : Factors affecting color stability of restorative resins. *Acta Odontol Scand* 1983 ; 41 : 11−18.
5. John M. Powers, Joseph B. Dennison, & Patrick J. Lepeak : Parameters that affect the color of direct restorative resins. *J Dent Res* 1978 ; 57(9−10) : 876−880.
6. Wayne D. Cook & Maxwell P. Chong : Color stability and visual perception of dimethacrylate based dental composite resins. *Biomaterials* 1985 ; 6 : 257−264.
7. G. M. Brauer : Color changes of composites on exposure to various energy sources. *Dent Mater* 1988 ; 4 : 55−59.
8. John M. Powers, Joseph B. Dennison, & Andrew Koran : Color stability of restorative resins under accelerated aging. *J Dent Res* 1978 ; 57(11−12) : 964−970.
9. I. E. Ruyter, K. Nilner, & B. Möller : Color stability of dental composite resin materials for crown and bridge veneers. *Dent Mater* 1987 ; 3 : 246−251.
10. Hiroyuki Hayashi, Kazuko Maejima, Kazuhiro Kezuka, Kantara Ogushi, Atsushi Kono, & Takao Fusayama : In vitro study of discoloration of composite resins. *J Prosthet Dent* 1974 ; 32(1) : 66−69.
11. R. R. Seghi, W. T. Johnston, W. J. O'Brien : Spectrophotometric analysis of color differences between porcelain systems. *J Prosthet Dent* 1986 ; 56 : 35−40.
12. Jan W. V. van Dijken : A clinical evaluation of anterior conventional, microfiller, and hybrid composite resin fillings. A 6 year follow-up study. *Acta Odontol Scand* 1986 ; 44 : 357−367.
13. I. Eystein Ruyter & Svend A. Svendsen : Remaining methacrylate groups in composite restorative materials. *Acta Odontol Scand* 1977 ; 36 : 75−82.
14. Michael F. Burrow & Owen F. Makinson : Color change in light-cured resins exposed to daylight. *Quintessence International* 1991 ; 22(6) : 447−452.
15. M. D. Gross, & J. B. Moser : A colorimetric study of coffee and tea staining of four composite resins. *J Oral Rehabil* 1977 ; 4 : 311−322.
16. Z. A. Khokhar, M. E. Razzoog, & P. Yaman : Color stability of restorative resins.

Quintessence International 1991 ; 22(9) : 733−737.

17. W. H. Douglas & R. G. Craig : Resistance to extrinsic stains by hydrophobic composite resin system. *J Dent Res* 1982 ; 61 : 41−43.
18. N. Satou, A. M. Khan, I. Matsumae, J. Satou, & H. Shitani : In vitro color change of composite-based resins. *Dent Mater* 1989 ; 5 : 384−387.
19. Chung Moon Um & I. Eystein Ruyter : Staining of resin-based veneering materials with coffee and tea. *Quintessence International* 1991 ; 22(5) : 377−386.
20. Erik Asmussen & Erik Keith Hansen : Surface discoloration of restorative resins in relation to surface softening and oral hygiene. *Scand J Dent Res* 1986 ; 94 : 174−177.
21. H. St. Germain, M. L. Swartz, R. W. Phillips, B. K. Moore, & T. A. Roberts : Properties of microfilled composite resins as influenced by filler content. *J Dent Res* 1985 ; 64(2) : 155−160.
22. H. ØYSÆd & I. Eystein Ruyter : Water sorption and filler characteristics of composites for use in posterior teeth. *J Dent Res* 1986 ; 65(11) : 1315−1318.
23. J. M. Powers, E. R. Bakus, & A. J. Goldberg : In vitro color changes of posterior composites. *Dent Mater* 1988 ; 4 : 151−154.
24. Farid Noie, Kathy L. O'Keefe, & John M. Powers : Color stability of resin cements after accelerated aging. *The International J of Prosthodontics* 1995 ; 8(1) : 51−55.