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Color Stability of Some Aesthetic Composites after Immersion in Different Staining Solutions and UV Accelerated Aging

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ABSTRACT

Background and Objective: One of the most important factors in the success of anterior composite restorations is color stability in different situations in mouth. The aim of this study was to evaluate the color stability of four anterior composites in the staining solutions (tea and coffee) and under Ultra-Violette (UV) accelerated aging using spectrophotometer.

Methods: This in-vitro study was conducted on 80 samples of four composites (Grandio, G-aenial, Kalore and Estelite Σ quick). Samples were divided into four subgroups (5 samples in each subgroup): immersion in coffee (for 48 hours), tea (for 48 hours), distilled water (as the control group) and exposure to UV for 168 hours. The color of the samples was evaluated by the spectrophotometer before and after the various conditions using the CIE Lab system and color changes (ΔE^*_{ab}) were calculated.

Findings: In all composites, the maximum and minimum levels of color changes were related to the groups immersed in coffee and distilled water, respectively. The significant color change was observed in all subgroups (ΔE^*_{ab}>3.3), except for the control groups and tea-Estelite Σ quick (ΔE^*_{ab}=2.79). The highest color change was related to immersion in coffee-Grandio (ΔE^*_{ab}=9.084) and the lowest observed in immersion in distilled water-Estelite Σ quick (ΔE^*_{ab}=0.836). In coffee immersion subgroups of Grandio and Kalore, higher color change was seen compared to Estelite Σ quick (p=0.02 and p=0.028, respectively).

Conclusion: Long-term exposure to tea and coffee and UV accelerated aging leads to clinical color change in Grandio, G-aenial, Kalore and Estelite Σ quick composites.

Keywords: *Spectrophotometry, Composite Resins, Coffee, Color.*

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Introduction

Nowadays, resin composites are widely used due to the increased esthetic needs of patients, improved formulation and simplification of their bonding process (1). Color stability is one of the challenging subjects for composite restorations, and is one of the determinants of the longevity of aesthetic restorations. One of the main disadvantages of composites is color instability, which is compromised the success of composite restorations over the long term (2-5).

The color stability is a multifactorial issue and two groups of internal and external factors influence the color change of composites (1, 5). The external factors include nutritional habits, heat, water absorption, poor oral hygiene and surface smoothness of restoration (6, 7). The main internal factors are filler size and percentage, matrix type and percentage, coupling factor, initiator type and degree of conversion. The internal and external factors affect each other (2).

The ultraviolet radiation can be absorbed by the unreacted amines remaining in the polymer network, which leads to the production of molecules with a higher energy level. These active molecules are capable of reacting with oxygen and other aromatic groups and increasing the absorption of visible light, especially in the green-blue limit of the electromagnetic spectrum, thus leading to a yellow-red color change of the sample. Unreacted monomers are also affected by the UV radiation (8, 9).

Spectrophotometry is a suitable and reliable method for measurement of color change due to the accuracy, standardization and numerical report of color (1). According to numerous articles, the human eye is able to detect $\Delta E^*_{ab}>1$. There are controversies in various literatures regarding acceptable clinical values of ΔE^*_{ab}. The reported values range from 0.4 to 3.7. However, in most studies, the acceptable clinical values are considered as $\Delta E^*_{ab}<3.3$ (10, 11).

Several studies have evaluated the color stability of composite restorations (12-14). Mansouri et al., evaluated the water sorption and color stability of Bulk-Fill composite compared to nanohybrid composite. Their results showed that the Bulk-Fill composite yielded better color stability and similar water sorption (12). According to Brave et al., the microhybrid composites represent higher color stability than nanofill composites (13), while Reddy et al. found that the nanofill composites show the highest surface smoothness and the least color change compared to the microhybrid and hybrid composites (14).

The aim of this study was to evaluate the color stability of several new esthetic composites [Grandio, G-aenial, Kalore and Estelite Σ quick] under UV aging and immersion in several common types of drinks including tea and coffee using the spectrophotometer, which they have not been studied yet. The null hypothesis was that, there is no significant difference among the color stability of mentioned composites after UV aging and immersion in coffee, tea and distilled water. The aim of this study is to investigate the color stability of four types of anterior composites in color solutions (tea and coffee) and aging with ultraviolet rays (UV) by spectrophotometric method.

Methods

In this laboratory experimental study (ethics code: IR.KMU.REC.1395.731), 80 samples were prepared for four types of Kalore, G-aenial, Grandio and Estelite Σ quick composites, including 60 composite discs (diameter 10 mm and thickness 2 mm) for the immersion subgroups in distilled water, tea, and coffee (n=5), which were made by a metal mold. The materials used in this study are listed in Table 1.

Table 1. Composition of materials used in this study

Composite resin	Manufacturer	Filler loading (Wt%)	Type	Composition
Grandio (Gr)	Voco, Germany	87	Nano-hybrid	Matrix: Bis-GMA, TEGDEMA Filler: Bariumeboron-alumino-silicate glass (0.1-2.5 mm), Silica: 20-60 nm
G-aenial (Ga)	GC Corporation, Japan	76	Micro filled hybrid	Matrix: UDMA, Dimethacrylate Co-monomer Filler: silica, strontium, lanthanoid fluoride (16-17 nm), silica (>100 nm), fumed silica (<100 nm)
Kalore (K)	GC Corporation, Japan	82	Nano hybrid	Matrix: UDMA, Dimerhacrylat comonomer Filler: (fluoroalumino silicate glass, strontium glass, prepolymer-HDR, silicon dioxide)
Esthelite Σ quick (Es)	Tokuyama, Tokyo, Japan	82	Supra-nano filled	Matrix: bis-GMA, TEGDMA Fillers: zirconia/sílica particles

The sample size was determined according to other similar studies (15, 16). Each sample was polymerized by a light curing device (Demi L.E.D curing light, Kerr, USA) with an intensity of 600 mW/cm² for 20 seconds. The intensity of the light cure device was checked every time using a radiometer (L.E.D. radiometer, Kerr, USA).

To prepare samples of UV aging test of each composite, a rectangular cube-shaped specimen (82×2×2 mm) was prepared for the possibility of inserting the sample in UV aging test chamber. After UV aging procedure, the rectangular samples were cut to five pieces by diamond disk.

All samples were polished by a complete sequence of Sof-Lex aluminum oxide discs (3 M ESPE, St. Paul, USA) from coarse to superfine for 30 seconds (17) and then rinsed with an ultrasonic cleaner (model 2210; Branson Ultrasonics Corp., Danbury, Conn.) for two minutes to remove the remaining debris from the polishing process and any surface contamination. All specimens were kept in the incubator (Malek-Teb, Iran) for 24 hours at 100% humidity at 37°C for complete polymerization.

The samples were then prepared to measure the amount of initial color and spectral reflectance with spectrophotometer device (X-Rite Gretag Macbeth ColorEye 7000A, USA). The samples were placed on a standard white paper as a background during spectral measurement for initial color. Then, the colorimetric values of the measured spectra were calculated according to the standard CIE$L^*a^*b^*$ system and under CIE D65 illuminant and 2° standard observer. The color of the samples was recorded by three parameters: L^* (lightness), a^* (red-green) and b^* (yellow-blue).

Preparation of staining solutions:

The coffee: 3.6 grams coffee (Nescafe Classic, Nestle, Switzerland) was dissolved in 300 ml of boiled distilled water according to the proposed concentration of the manufacturer. After 10 minutes, the solution was filtered through filter paper.

The tea: two 2-gram packets of tea bags (Yellow Label Tea, Lipton, London) were boiled in 300 ml distilled water for 10 minutes (17).

The disc-shaped specimens of each composite were randomly divided into three subgroups of 5, consisting of three common types of drinks including tea, coffee and distilled water (negative control). The samples were immersed in the solutions at 50°C for 48 hours in incubator. If a person consumes an average

four cups of tea and coffee per day and an average of one minute each, 48 hours of immersion is equivalent to two years of consumption of tea and coffee (15, 18, 19). The samples were rinsed with distilled water for 30 seconds and cleaned with a soft toothbrush to remove the debris.

The accelerated aging test was carried out using the UV Accelerated Weathering Tester (Q-LAB, USA) based on the ASTM G154 protocol, so that the aging period consists of intermittent ultraviolet (8 hours) exposure with a period of condensation (4 hours) at 60°C and 100% humidity, a total of 252 hours, including 168 hours of UV radiation with an emission peak of 313 nm (20). After performing the accelerated aging and immersion in the staining solutions, the final colorimetric analysis of the specimens was performed by the spectrophotometer as mentioned before. The overall color change for each sample was calculated using the following equation: $\Delta E^*_{ab} = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$.

The data were analyzed by SPSS software version 20 using two-way ANOVA and Tukey's post hoc test; the significance level was considered to be $p<0.05$.

Results

The highest color change was related to the subgroup immersed in coffee-Grandio composite ($\Delta E^*_{ab}=9.084$) and the lowest color change observed in the subgroup immersed in distilled water-Estelite Σ quick composite ($\Delta E^*_{ab}=0.836$). All subgroups showed a clinically significant color change ($\Delta E>3.3$), except for the subgroups immersed in distilled water (control group) and in tea-Estelit Σ quick ($\Delta E^*_{ab}=2.79$). In all composites, the highest and lowest color changes were observed in the subgroups immersed in coffee and distilled water, respectively (Table 2). There was not any significant difference between all studied composites in distilled water and tea immersion and also UV aging situation, but in coffee immersion subgroup, Estelite Σ quick had significantly lower ΔE than Grandio ($p=0.020$) and Kalore ($p=0.028$).

All types of composite showed a decrease in brightness (ΔL^*). Positive values of Δa^* represent shifting to redness and negative values of Δa^* represent shifting to greenness. Positive values of Δb^* represent shifting to yellowing and negative values of Δb^* represent shifting to bluing. The values recorded for ΔL^*, Δa^* and Δb^* parameters are given in Table 3.

Table 2. Mean and standard deviation values of color change (ΔE) for composites in different methods of aging.

	Estelite Σ quick Mean±SD	G-aenial Mean±SD	Kalore Mean±SD	Grandio Mean±SD
Distilled Water	$0.836\pm0.836^{A,a}$	$1.684\pm0.922^{A,a}$	$1.127\pm0.724^{A,a}$	$1.262\pm0.373^{A,a}$
UV	$3.614\pm1.960^{A,a}$	$3.754\pm0.414^{A,a,b}$	$3.407\pm0.486^{A,a,c}$	$4.180\pm0.088^{A,a,c}$
Coffee	$4.013\pm0.707^{a,c}$	$6.438\pm1.285^{A,b,c}$	$8.935\pm4.220^{A,b}$	$9.084\pm2.631^{A,b}$
Tea	$2.792\pm1.790^{A,a}$	$4.127\pm0.758^{A,a,b}$	$6.961\pm4.215^{A,b,c}$	$6.090\pm3.363^{A,b,c}$

The same lower case in each column, means not statically significant difference. The same upper case in each raw, means not statically significant difference.

Table 3. Mean and standard deviation value of colorant parameter (ΔL, Δa, Δb) for composites studied in different methods of aging

	Estelite Σ quick Mean±SD			G-aenial Mean±SD			Kalore Mean±SD			Grandio Mean±SD		
	Δb^*	Δa^*	ΔL^*	Δb^*	Δa^*	ΔL^*	Δb^*	Δa^*	ΔL^*	Δb^*	Δa^*	ΔL^*
Distilled Water	-0.699 ±.172	0.155 ±.146	-0.3566 ±.238	-0.0456 ±.832	0.0974 ±.512	-1.391 ±1.002	-0.1096 ±1.047	0.330 ±.468	-0.998 ±.796	-0.541 ±.797	0.161 ±.147	-0.889 ±.291
UV	-1.932 ±.344	0.375 ±.167	-2.753 ±.238	3.265 ±.315	-.723 ±.059	-1.698 ±.327	2.531 ±.331	-0.845 ±.174	-2.042 ±.707	3.513 ±.354	-0.821 ±.209	-2.017 ±.604
Coffee	3.125 ±1.029	0.305 ±.385	-2.295 ±.714	4.696 ±1.342	2.835 ±4.512	-4.260 ±.478	6.943 ±3.679	-0.906 ±4.755	-5.296 ±2.318	6.811 ±2.384	1.039 ±.522	-5.848 ±1.422
Tea	1.556 ±1.738	0.700 ±.345	-2.037 ±.988	2.779 ±.630	1.740 ±1.598	-2.787 ±.665	3.833 ±1.924	1.262 ±5.925	-4.739 ±2.389	3.663 ±2.603	1.640 ±.837	-4.529 ±2.097

Discussion

According to the results of this study, the null hypothesis was rejected, all groups showed a clinically significant color change (ΔE^*_{ab}>3.3) in the immersion conditions in the staining solutions (coffee and tea) and UV aging, except for the groups immersed in distilled water (control group) and in tea-Estelite Σ quick. In all composites, the maximum and minimum levels of color change (ΔE^*_{ab}) were related to the groups immersed in coffee and distilled water respectively, which was statistically significant.

Several studies have shown composite discoloration after water immersion because of monomers degradation and fillers detachment due to the water sorption, which is different in composites (21, 22). Therefore, resin matrix characteristics and water sorption speed have influence on color changes (23).

Numerous studies have shown that tea-based color change is due to the adsorption of polar color additives to the surface that can be removed by brushing (16, 24) and in our study, there was not a significant difference between composites, which may relate to similar standard surface polish with Sof-lex disks.

While the coffee-based color change is due to both adsorption and absorption of polar color additives introduced into the composite, this absorption and penetration of colored materials into the organic phase is probably due to the compatibility of the polymer phase with the yellow pigment in the coffee. This could result a greater change in color in the immersion of coffee than tea (16).

Ertas et al. studied the degree of discoloration of several composite materials, including a posterior composite (Filtek P60), two universal composites (Filtek Z250, Quadrant) and two nanohybrid composite resins (Filtek suprime, Grandio) in immersion in various colored solutions, including wine, coffee, tea, soda and water, and reported that the color change of all types of composite materials was beyond the acceptable clinical threshold, and the color change caused by coffee was significantly higher than that of tea and distilled water (16), which is similar to the results of this study.

The remaining camphorquinone (the principal photo-initiator molecule) in composite is considered to be responsible for the color change to yellow with time. As the polymerization is not performed as 100%, the degree of composite transformation and the amount of unreacted camphorquinone are determining factors for yellowness of the composites (25-29). Therefore, the differences in the concentration and structure of photo-initiator molecules and reducing amines in a variety of composites may explain the wide range of Δa and Δb values obtained among the tested materials.

Since water plays an essential role in monomer leach, molecular displacement, and destruction of filler-matrix interfacial layers in silane and filler particles, the water absorption affects the color parameters of the restorations (28, 30). Thus, the characteristics of the matrix resin, especially the water absorption rate, play an important role in staining susceptibility. Urethane dimethacrylate (UDMA) appears to have the color stability greater than Bisphenol A-glycidyl methacrylate (BIS-GMA) because of its better polymerization and low water solubility and absorption (23, 31).

Generally, composite color change is a product of several factors mentioned above and cannot consider the effect of each factor individually (1). For example, the Estelite Σ quick composite containing the matrix resin with BIS-GMA and Triethylene glycol dimethacrylate (TEGDMA) has represented the lowest color change, while the Grandio composite with the UDMA matrix resin showed the highest color change, contrary to expectations, which can be attributed to other composite properties. In other words, the color stability is a multifactorial parameter.

The better performance of the Estelite Σ quick composite in most tested conditions of our study can be attributed to several characteristics:

The hallmark of this composite is the use of the Radical Amplified Photo Polymerization Initiator (RAP technology). The common photo-initiators include camphorquinone (CQ) /amine in which camphorquinone is used. Therefore, it is able to produce only one initiator molecule. In the RAP system, the initial stage of CQ excitation by light is the same as in conventional systems, but then the energy is transferred to the radical amplifier (RA). After transferring energy to RA, the excited CQ returns to the ground state and can produce multiple initiator radicals. Therefore, they can have high polymerization activity at the same time as conventional systems. Improved polymerization and higher degree of conversion reduce water absorption. On the other hand, remaining monomers and their unreacted double bonds oxidation lead to the production of colored lateral products (32) and also with RAP technology, initial CQ concentration is reduced, and as we know, CQ concentration have direct relation with long term discoloration (28, 29). Therefore, this composite with higher degree of conversion is expected to represent greater color stability at the same radiation time (8, 18), and in Estelite Σ quick, all above mentioned reasons may lead to lesser color changes.

In a study by Poggio et al. to compare the color stability of nanofill, nanoceramic, nanohybrids, microfill, microhybrid and supranano composites in different solutions, the color change in supranano composite was lower than others. This is consistent with the results of current study. In this study, even the control group (physiological solution) showed the color change more than clinically acceptable limit, but the color change in the present study in the control group of composites was below the clinically acceptable level. This can be resulted from the difference in control solutions (distilled water versus physiological solution), as well as immersion conditions (immersion time and solution temperature), and non-polished samples in the study of Poggio et al. and the presence of resin-rich surface on a sample that is prone to water absorption and subsequent color change (8).

However, it should be mentioned that in the present study, nano hybrid composites (Kalore, Grandio), despite the smaller size of fillers and smoother surface polishing, contrary to what was expected, showed a higher color stability compared to the microhybrid composite (G-aenial).

In the studies of Ertan et al., Poggio et al. and Al kherief et al., the color stability of the microhybrid composites were better than those of nanohybrid composites (8, 16, 33). This is consistent with results of the present study. The reason is the lower color stability of agglomerated particles (nano-clusters) in nanohybrid composites, which is less likely to be due to more water absorption of these nano-clusters compared to spread fillers in micron size (16).

In our study, all four composites showed a clinically significant change in color ($\Delta E^*_{ab}>3$) after the UV aging process. In the study of Catelan et al., all tested groups showed a color change in the clinically acceptable range after UV aging (17), which is not consistent with the results of this study. This difference can be caused by the difference in the tested composites and different preparation method (different polishing system) and different aging method.

Since the color stability is also affected by the surface roughness and the degree of conversion of the composites, it is recommended to evaluate them in further studies. Based on the results of the present study, it can be concluded that long-term exposure to some types of colored solutions (tea and coffee) is effective on the color stability of modern cosmetic restorative materials. Estelite Σ quick composite, which is a supernano composite, showed less color change compared to other composites in different conditions.

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