Effect of Accelerated Aging on Color Change of Direct and Indirect Fiber-Reinforced Composite Restorations

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Abstract

Objectives: The aim of this study was to assess the effect of artificial accelerated aging (AAA) on color change of direct and indirect fiber-reinforced composite (FRC) restorations.

Materials and Methods: Direct (Z250) and indirect (Gradia) composite resins were reinforced with glass (GF) and polyethylene fibers (PF) based on the manufacturers’ instructions. Forty samples were fabricated and divided into eight groups (n=5). Four groups served as experimental groups and the remaining four served as controls. Color change (ΔE) and color parameters (ΔL*, Δa*, Δb*) were read at baseline and after AAA based on the CIELAB system. Three-way ANOVA and Tukey’s test were used for statistical analysis.

Results: Significant differences were found in ΔE, ΔL*, Δa* and Δb* among the groups after AAA (P<0.05). Most of the studied samples demonstrated an increase in lightness and a red-yellow shift after AAA.

Conclusions: The obtained ΔE values were unacceptable after AAA (ΔE≥ 3.3). All indirect samples showed a green-blue shift with a reduction in lightness except for Gradia/PF+ NuliteF.

Keywords: Aging; Composite Resins; Color

INTRODUCTION

The concept of applying fibers for the purpose of reinforcement in dentistry is not new. Fiber-reinforced composites (FRCs) were first introduced in the 1960s, and currently have a wide range of dental applications as in fixed partial dentures (FPDs), periodontal splints, orthodontic retainers, implant superstructure and removable partial dentures [1-3]. Nowadays, FRC-FPDs are an alternative to metal-ceramic adhesive FPDs. The performance of FRC-FPDs is affected by the composition of FRC and the adhesive strength between the fiber and composite [3]. Addition-ally, fiber surface treatment, impregnation of fibers with matrix, direction and position of fibers [4], water sorption of the matrix [5] and inherent properties of fibers and matrices [2,6] have been reported as important factors related to the mechanical properties of these restorations. Improperly saturated fibers may cause voids in FRC and enhance water sorption and consequently weaken the structure of FRC. To overcome these problems, pre-impregnated fibers were introduced [7]. The FRCs are made of at least two separate parts. The reinforcing part (fiber) provides stiffness, enhances the properties of composite and serves as a crack stopper. The matrix component (composite) supports the fibers, stabilizes their geometric orientation and allows workability [2,3]. There are several different varieties of saleable fibers such as glass fibers, carbon fibers, Kevlar fibers, Vectran fibers, and polyethylene fibers, which are added to composite materials. The most practical fibers in dental practice include glass fibers, carbon
fibers and synthetic fibers such as aramid and polyethylene types. Among the afore-mentioned types, glass and polyethylene fibers along with composite can provide excellent esthetic results [2,8]. Direct and indirect resin materials can be used for FRCs. The inherent property of resin matrix has a significant effect on mechanical properties and clinical applications of FRC-FPDs [2,3]. To date, several studies have compared the mechanical properties of FRCs [3,7,9]. With regard to previous works in the literature, only five in vitro studies have evaluated the optical properties of FRCs. The results of these studies are difficult to compare because they evaluated different materials [10-14]. Only one previous study evaluated the color stability of FRC restorations [13]. Color stability is highly important for the success of aesthetic restorations [15]. Thus, the purpose of this study was to determine the color stability of FRCs (direct and indirect) reinforced with two types of pre-impregnated (polyethylene) and non-impregnated (glass) fibers after artificial accelerated aging (AAA).

MATERIALS AND METHODS
In this in vitro experimental study, two types of fibers namely glass fibers and polyethylene fibers were incorporated into direct and indirect composite resins. The characteristics and composition of the materials according to Sharafeddin et al, [16] Hasani Tabatabaei et al, [14] and the manufactures are shown in Table 1. Classification of groups is shown in Table 2.

Preparation of samples: Two stainless steel molds were used with smooth inner surface to fabricate samples measuring 13 mm in diameter and 1mm or 3mm in height. These dimensions were chosen to enable color measurement in a spectrophotometer. The samples of the veneering composites and base composites were prepared using the shallower mold and then another deeper mold was used for the three layers of samples namely base, fiber and the veneering from the bottom to the top, respectively (Fig. 1). Forty cylinders for each veneering composite and 20 cylinders for each base composite were used. The shallower mold was overfilled with unset composite material. The composite was then gently pressed between two glass slabs in order for the excess material to leak out and prevent void formation. The base composites recommended by the manufacturers for each type of fiber were Nulite F for polyethylene fibers and adhesive C for glass fibers. The Z250 cylinders were light-cured with a light-curing unit (Blue Phase; Ivoclar Vivadent, Schaan, Lichtenstein) for 20 seconds with 1,000 mW/cm2 light intensity. The output of the curing light was tested with a radiometer (1,000 mW/cm2). Since the curing process of Gradia has two stages, the initial curing of Gradia samples was done using GC StepLight SL-I light-curing unit (GC, Tokyo, Japan) for 10 seconds based on the manufacturer’s instructions. The second curing phase of Gradia samples was done in a furnace (Labolight LV-III; GC America Inc., Alsip, IL, USA) for three minutes [17]. Base composite cylinders were light-cured according to the manufacturer’s recommendations.

In control groups, no fiber reinforcement was done in the control groups. Each polymerized veneering composite cylinder (Z250 and Gradia indirect) was placed in the deeper mold and the
residual space was filled with base composite. In G5 and G6, Z250 cylinders were placed in deeper molds and the remaining space in the molds was filled with Nulite F and adhesive C, respectively. The mold was pressed between two glass slabs and composite was light-cured for 40 seconds using the light-curing unit. In G7 and G8, Gradia indirect cylinders were placed in deeper molds and the residual space in the molds was filled with Nulite F and adhesive C, respectively. The mold was compressed between two glass slides and final curing was done using the light-curing unit [17].

In non-impregnated groups, the Nulite F polymerized composite cylinders were placed in the deeper mold. Braided fiber (BTD, NSW, Australia) was cut into 10 mm pieces, carefully impregnated with special resin recommended by the manufacturer (Resist) for 20 minutes away from light and implanted on a thin layer of Nulite F composite, cured for 20 seconds away from light and implanted on a thin layer of Nulite F composite, cured for 20 seconds, placed on polymerized adhesive C cylinder, and then covered with a thin layer of adhesive C composite. Then, the cylinders of each veneering composite were pressed on adhesive C cylinders using a glass slab. The curing in G2 and G4 was done in the same manner as in G5 and G7, respectively.

Color measurements: The color of discs was evaluated at baseline using a reflection spectrophotometer (Color-eye 7000A, GretagMacbeth, New Windsor, NY, USA) against a white background in the CIELAB color space. This color system was initially described by Pier-de-Souza et al, [18] in which L* indicates lightness, a* represents green-red (-a=green, +a=red) and b* represents blue-yellow (-b=blue, +b=yellow).

### Table 1: The characteristics and composition of materials used in this study

| Materials & Manufacturer | Composition* |
|--------------------------|--------------|
| Filtek Z250 (3M, St Paul, MN, USA) | BIS-GMA, UDMA, BIS EMA, Zirco-nium/Silicon fillers |
| GRADIA indirect (GC, Tokyo, Japan) | UDMA, silica nanofiller, glass and prepolymerized filler |
| Fiber ribbon (Angelus, Londrina, PR, Brazil) | Glass fiber |
| Fiber braided (BTD, NSW, Australia) | Polyethylene fibers |
| Nulite F (BTD, NSW, Australia) | Micro rod reinforced composite, hybrid BIS-GMA |
| Adhesive c (Angelus, Londrina, PR, Brazil) | Bis-GMA, UDMA, Silicon Dioxide highly dispersed, catalysts, pigments |
| Resist bonding (BTD, NSW, Australia) | Unfilled resin, low viscosity, Bis-GMA, UDMA, Comphorquinone |

* BisEMA: Ethoxylated Bisphenol A glycidyl dimethacrylate; UDMA: Urethane dimethacrylate

### Table 2: Classification of groups according to the type of composite and fiber/base composite (n=5 per group)

| Group | Veneering composite | Fiber/Base composite |
|-------|---------------------|----------------------|
| G1    | Z250 | Braided fiber/ Nulite F |
| G2    | Z250 | Fiber ribbon/ adhesive C |
| G3    | Gradia indirect | Braided fiber Nulite F |
| G4    | Gradia indirect | Fiber ribbon/ adhesive C |
| G5    | Z250 | No fiber/Nulite F |
| G6    | Z250 | No fiber/ adhesive C |
| G7    | Gradia indirect | No fiber/Nulite F |
| G8    | Gradia indirect | No fiber/ adhesive C |
indicates blue-yellow (-b=blue, +b=yellow). After the initial color assessment, the discs were submitted to AAA (Xenon weather-Ometer, Atlas Material Testing Technology, Chicago, IL, USA). This aging machine was employed to subject the discs to both visible and UV light in 100% relative humidity at 37°C. 300 hours of aging correspond to one year of clinical service [19]. Thus, 300 hours of aging were performed in this study, corresponding to one year of clinical use. After this time, the discs were submitted to color assessment by the same technique, which was explained earlier. The change in color that occurred during AAA was calculated using the equation: \[ \Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}, \]
where \(\Delta L^*, \Delta a^*\) and \(\Delta b^*\) are the differences in the respective values before and after aging [8]. Values of \(\Delta E \geq 3.3\) were considered clinically unacceptable [20].

The effects of veneering composite, base composite and fiber on color stability and color parameters were evaluated using three-way ANOVA. Since the interaction effect of independent variables was significant, subgroup analysis was applied using one-way ANOVA and Tukey's test. The statistical significance was set at \(P<0.05\).

RESULTS
The mean and standard deviation of color parameters (\(\Delta L^*, \Delta a^*, \Delta b^*\)) and color change (\(\Delta E\)) in each group are listed in Table 3. All the groups showed clinically unacceptable (\(\Delta E \geq 3.3\)) color change following AAA. The minimum value of \(\Delta E\) (4.42±0.37) was found in G2; whereas, the maximum value of \(\Delta E\) was found in G5 (Table 3). Three-way ANOVA indicated a significant difference in \(\Delta E\) among the groups after AAA (\(P=0.000\)). Statistical analysis using three-way ANOVA revealed significant differences in \(\Delta L^*, \Delta a^*, \Delta b^*\) among the groups (\(P=0.000\)). According to the Tukey's test results, no statistically significant difference was found in \(\Delta L^*\) between G2 and G3 (\(P=0.444\)). The Tukey's test showed no significant difference in \(\Delta a^*\) between G1 and G2 (\(P=0.096\)). This finding demonstrated that the type of fiber did not have any effect on the stability of “a” parameter of FRC restorations after AAA. According to the Tukey's test, no significant difference was found in \(\Delta b\) between G1 and G3 (\(P=0.555\)). This result confirmed that the stability of “b” parameter was not affected by the type of composite in FRC restorations. After AAA, all study samples demonstrated increased lightness except for G4, G7 and G8. All the samples in groups G1, G2, G3, G5 and G6 exhibited a red shift accompanied by a yellow shift, while samples in groups G4, G7 and G8 exhibited a green shift with a blue shift (Table 3).

DISCUSSION
The color stability of direct and indirect composites reinforced with glass and polyethylene fibers after AAA was evaluated in this study. The null hypothesis of the current study stating that the color change of the tested materials would remain within the clinically acceptable range after AAA was rejected. The \(\Delta E\) values of all groups after AAA varied from 4.42 to 15.08, which were higher than the clinically acceptable threshold [13]. Regarding the acceptable clinical color match of tooth and restorations, the \(\Delta E\) value of more than 3.5 is considered as color mismatch [14]. On this basis, all the groups showed color mismatch.

In this study, color stability was assessed by irradiation of visible and UV light on the samples using a xenon test chamber in humid environment and ambient temperature of 37°C, in order to simulate the effect of long-term clinical service [19]. The consequence of AAA on color of dental restorative materials has been evaluated in many studies [15,18-20]. Nonetheless, only one experimental study assessed the effect of AAA on color stability of FRCs [13]. The data in the current study demonstrated higher \(\Delta E\) values than those reported by Tuncdemir and Aykent.
Table 3: Mean (standard deviation) values of ∆L*, ∆a*, ∆b* and ∆E*

| Groups | ∆L*     | ∆a*     | ∆b*     | ∆E     |
|--------|---------|---------|---------|--------|
| G1     | 8.21(0.77) | 1.34(0.37)§ | 5(0.85)§ | 9.74(0.93) |
| G2     | 3.8(0.37)§ | 1.83(0.19)§ | 1.23(0.43) | 4.42(0.37) |
| G3     | 4.67(0.93)§ | 2.70(0.16) | 4.21(1.06)§ | 6.9(1.07) |
| G4     | -11.18(1.26) | -1.69(0.34) | -6.85(1.19) | 13.29(0.92) |
| G5     | 11.77(0.94) | 8.03(0.94) | 4.8(0.17) | 15.08(1.08) |
| G6     | 5.05(0.36) | 0.51(0.35) | 1.62(0.13) | 5.34(0.37) |
| G7     | -7.54(0.64) | -3.5(0.59) | -0.3(0.16) | 8.34(0.75) |
| G8     | -5.2(0.5) | -5.17(0.71) | -2.27(0.17) | 7.85(0.69) |

The similar superscript lower case letter in the same column shows no significant difference (p<0.05, Tukey’s test).

(2<ΔE<2.8) [13]. Our observations noticeably confirmed the effect of AAA on color stability of FRCs. This was in disagreement with the findings of Tuncdemir and Aykent [13]. The type of fibers and the veneering composites, preparation of samples (no-polishing or polishing, using base composite), method of AAA and color measurement instrument (colorimeter and spectrophotometer) were obviously dissimilar in our study and that of Tuncdemir and Aykent [13]. Thus, these differences might have been contributed to the discrepancy between the results of the two studies.

Color change of composite resin restorations in long-term service is a multi-factorial procedure (intrinsic and extrinsic factors). Color change of composite restorations occurs as a consequence of external factors such as dietary regimens or smoking habits, absorption and adsorption of extrinsic coloring agents, power and duration of polymerization and exposure to environmental factors. Intrinsic factors cause chemical alteration in resin matrix and at the resin-filler interface. Composition of resin matrix, fillers, loading and particle size distribution, type of photo-initiator and proportion of remaining C=C bonds are among the intrinsic factors [21-23]. The color stability of UDMA is higher than that of TEGDEMA and Bis-GMA [24,25]. Notwithstanding, the lowest ΔE was observed in G2, which included direct base composite (containing Bis-GMA and UDMA) and non-impregnated fiber.

We expected that indirect FRC with a second curing cycle would have superior color stability than direct FRC due to higher degree of conversion of composite. With regards to the results of the current study, it was surprising that the direct FRC restoration with impregnated fiber showed superior color stability. It is possible that the polymerization of indirect composite resin influenced the non-uniform composite-fiber interface [3]; thus, AAA had a worse effect on indirect FRC.

It should be borne in mind that there may be a correlation between the number of interfaces in restorations and color stability. It means that the interface is more susceptible to the effect of intrinsic and extrinsic factors in the process of aging. This correlation could not be confirmed based on the results of the current study, because the greatest color change after AAA was observed in G5 (control: Z250+no fiber/Nulite F). This result may be due to the incompatibility of the composite resin with base composite.

Our previous study [14] indicated that ΔE of the groups (similar materials as in our current study) was not significantly different before AAA. However, the color differences (4.42<ΔE<15.08) observed between groups after AAA in the current study were greater than the values reported in our previous study before AAA.
(1.91<ΔE<2.32). The group consisting of direct composite (Z250) and impregnated fibers (glass fibers) showed minimal color change both before and after AAA. This finding could be the result of superior adaptation with minimal space between the direct composite and the fibers and shows that these types of fibers do not need impregnation in resin before application. Another explanation for this result may be the chemical and structural differences that exist between fibers. A previous study [13] showed that Ribbond-reinforced composite materials exhibited greater color change than everStick-reinforced composite (polyethylene fiber) materials. Our findings were in contrast to theirs. Based on these in vitro findings, the direct composite impregnated with fiber had superior color stability compared to other FRCs.

As mentioned earlier, the non-uniform interface of fibers and indirect composite (as the result of second polymerization of indirect composite) served as a weak point susceptible to intrinsic and extrinsic factors in FRCs and resulted in the lowest color stability. However, the lowest color stability after AAA was observed in G5 (control group: Z250+Nulite F). This result may be due to structural complexity of FRCs tested in this study, which were fabricated based on the manufacturer’s instructions. Perhaps the interactions between the fibers and composites (base and veneering composites) were responsible for the observed difference and paradoxical data related to FRCs.

Analyzing the results related to the color parameters (Δa* and Δb*) of the groups before AAA [14] demonstrated that the group of Z250 composite and braided fiber exhibited a red-blue shift while other studied groups showed a green-blue shift. After 300 hours of AAA, the performance of Gradia indirect+no fiber/Nulite F, Gradia indirect+fiber ribbon/adhesive C and Gradia indirect+no fiber/adhesive C groups with regard to Δa* and Δb* remained unchanged. However, a color change occurred in other groups towards the longer wavelengths, which was in the direction of red and yellow after AAA. With the exception of G3 (Gradia indirect+braided fiber/Nulite F), all the indirect FRCs shifted to green and blue. These findings partly disagree with those of a previous study [13]. Another study [24] confirmed that all tested direct and indirect composites shifted to yellow after accelerated photo aging. Lee et al. [26] showed a red-blue shift in direct and indirect composites after thermocycling. The outcome of the above studies is controversial. Regardless of the different aging procedures, color shift difference was observed in direct and indirect composites. It seems that different aging mechanisms and variables (type of composite & method of color measurement) are involved in differences in color shift of composites in different studies. All the samples became lighter after AAA in the current study except for Gradia indirect+no fiber/Nulite F, Gradia indirect+fiber ribbon/ adhesive C and Gradia indirect+no fiber/adhesive C groups. This finding may be due to the composition of resin matrix. Bis-GMA and TEGDMA have lower color stability than UDMA due to the higher propensity for water absorption. This leads to modified susceptibility to staining, which is mainly due to change in “L” parameter rather than “a” and “b” parameters [24,25]. However, UDMA-based indirect FRCs except for G3 became darker after AAA. The reason for the discrepancies in fiber-dependent changes in CIE parameters may be the dissimilar resin compositions and filler content of base composites. In the current study, it seems that the use of Nulite F composite based on the manufacturer’s recommendation was not compatible with Gradia indirect composite.

The AAA is considered an extrinsic post-curing factor. Elevated temperature during AAA may cause an increase in the degree of conversion of composite resin. Also, UV irradiation has been related to chemical changes in the initiator system, activators and resin [22,23]. On the other
hand, UV-light exposure and xenon-arc lamps in AAA machines both produce intense light composed of UV and infrared wavelengths. This situation imitates blackbody radiation more than natural sunlight [26]. Besides, load cycling and nutritional habits may affect the color stability of FRC restorations in the clinical service, which was not evaluated in the current study. Therefore, similar assessments should be done carefully in the clinical setting and only evaluating the effect of AAA cannot be correlated to a clinical time frame.

CONCLUSION
Within the limitations of this in vitro study, the results showed that all direct and indirect FRC samples demonstrated clinically unacceptable color change after AAA. The direct composite reinforced with impregnated fibers showed better color stability than other FRCs.

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