

Analysis of color stability and degree of conversion of different types of resin composites

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Abstract: Resin composites containing surface pre-reacted glass (S-PRG) have been introduced to reduce demineralization and improve remineralization of the tooth structure. However, water diffusion within the material is necessary for its action, which can impair its overall physicochemical properties over time, including color stability. This study aimed to evaluate the color stability and related degree of conversion (DC) of four resin composites. Discs (6 x 4 mm, n = 5/group) of microhybrid (MH), nanofilled (NF), nanohybrid (NH), and S-PRG-based nanohybrid (S-PRG-NH) composites with two opacities (A2/A2E and A2O/A2D) were prepared. Color (CIELab and CIEDE2000) was evaluated with a spectrophotometer after aging in grape juice (2 x 10 min/10mL/7days). The DC was analyzed by using Fourier transform infrared spectroscopy before and after light-curing. Data were statistically analyzed by using two-way analysis of variance and post-hoc least significant difference tests ($p < 0.05$). In the color stability analysis, the interaction between filler type and opacity was significant (CIELab, $p = 0.0015$; CIEDE2000, $p = 0.0026$). NH presented the highest color stability, which did not differ from that of MH. The greatest color alteration was observed for S-PRG-NH. S-PRG fillers also influenced DC ($p < 0.05$). The nanohybrid resin composite presented favorable overall performance, which is likely related to its more stable organic content. Notwithstanding the benefits of using S-PRG-based nanohybrid resins, mostly in aesthetic procedures, professionals should consider the susceptibility of such resins to color alteration, probably due to the water-based bioactive mechanism of action.

Keywords: Color; Composite Resins; Dental Restoration Failure; Materials Testing; Polymerization.

Introduction

Resin composites are often indicated to replace lost tooth structures because of their ability to mimic the shape and color of natural teeth, resulting in a nearly imperceptible restoration.¹ In addition, physical and chemical properties allow resin-based restoration to endure daily oral challenges and preserve its integrity, luster, and color stability over the years.² However, there is no single ideal material for all clinical



situations; therefore, it is necessary to know the characteristics and behaviors of each material for better-informed decisions.³⁻⁵

As established in the literature, the composition of materials,⁶ filler size, and their distribution modulate the interaction of a resin composite with light⁷ by having a direct impact on the material roughness.³ Nanosized composites have shown reduced surface roughness when compared to their macro and microsized counterparts.⁸ This is important because increased surface roughness may lead to increased staining,⁶ particularly in patients with frequent intake of staining beverages, such as coffee,⁹ wine,¹⁰ and grape juice,¹¹ whose low pH can also harm the integrity of the surface.¹² Thus, to improve the performance of resin composites over time, the industry has been implementing modifications in the concentration, amount, type, and size of fillers.¹³⁻¹⁵

The optical properties of resin composites have been a significant factor, as their interaction with light plays an important role in mimicking tooth structures. Therefore, their opacity and/or translucency has enhanced layering techniques, enabling a higher level of reproducibility of natural dental aspects. The level of translucency of these materials is also mainly regulated by their composition and configuration of inorganic fillers.¹⁶ Most resin composites on the market are available in different opacities generally referred to as dentin (opaque) and enamel (translucent) resins. The variation in composition, as well as the optical interaction with light and pigments, can influence the color stability of these materials.

Additionally, another major concern with resin composite restorations is the frequent occurrence of dental caries around restorations. To overcome this clinical problem, ion-releasing composites have been developed, such as bioactive glass composites or xerogel-based multionic systems, including surface pre-reacted glass (S-PRG) fillers.¹⁷⁻²⁰ These fillers consist of a pre-reacted fluorosilicate filler with a polyacrylic acid-treated surface layer. They combine core particles containing glass ionomer in a resin matrix.^{14,18,20} As a consequence of the release of fluoride, sodium, boron, aluminum, silicate, and

strontium, these resins can reduce demineralization and improve remineralization.¹⁸⁻²² In addition, because of the release of boron and fluoride, they have antibacterial properties, thereby reducing the risk of carious lesions.^{18,20,21}

Composites containing S-PRG fillers have great structural strength and good physical, chemical, and mechanical properties,^{18,20,23} and a beyond-acceptable degree of conversion (DC). However, owing to the presence of S-PRG fillers, a certain amount of water should be diffused within the material to allow ion release into the environment,^{19,24,25} which could influence the optical properties of this resin composite over time, including color stability.^{24,26}

To allow the incorporation of bioactive materials and as an attempt to address other drawbacks, the organic matrix of resin composites has gone through a lot of interesting changes in recent years.^{5,27-29} High- and low-molecular-weight monomers are therefore balanced to safeguard inorganic fillers, preserving other important clinical features, such as viscosity and DC.³⁰ Special attention should be given to the DC of resin composites, as inadequate or insufficient conversion of monomers jeopardizes the mechanical properties and favors staining due to the presence of unreacted residual monomers.³¹⁻³³ Considering that S-PRG fillers partially depend on the aqueous environment, it is still unclear how their presence can interfere in color appearance.^{3,34,35}

Given that unsatisfactory esthetic appearance has been considered the main reason for the replacement of anterior restorations³⁶ and the inclusion of ion-releasing fillers requires some level of water diffusion within the material,³⁷ the optical properties of different composite resins should be further investigated. Therefore, this study aimed to evaluate the color stability and related degree of conversion (DC) of four resin composites (microhybrid, nanofilled, nanohybrid, and S-PRG-based nanohybrid) in two different opacities (A2/A2E and A2O/A2D). The first null hypothesis stated that no difference in color stability would be detected among the types of resin composites evaluated, regarding their type and opacity. The second null hypothesis stated that there was no difference in DC among the tested resins.

Methodology

Experimental design

This *in vitro* study analyzed two factors: a) resin composites classified into four levels according to the size and type of filler particles: microhybrid (MH), nanofilled (NF), nanohybrid (NH), and nanohybrid with S-PRG (S-PRG-NH); and b) opacity at two levels: translucent/enamel (T/E) and dentin/opaque (D/O). A2 color was set for all specimens. For color assessments, two response variables were used: color alteration measured by a spectrophotometer and calculated based on the CIELab (ΔE_{ab}^*) and CIEDE2000 (ΔE_{00}) equations after aging in grape juice. For the DC, the absorbance of the materials before and after light-curing by Fourier transform infrared spectroscopy (FTIR) was the response variable. The experimental unit consisted of a specimen of resin composite.

The technical specifications of each material are displayed in Table 1.

Sample size calculation

In the color assessments, the effect size for the CIELab and CIEDE2000 equations was estimated to be 0.779 and 0.741, respectively, based on the findings from a pilot study. Consequently, the total sample size was estimated to be $n = 4/\text{group}$ and $n = 5/\text{group}$. However, considering that the same specimens were used to calculate the color alteration using both equations, the final sample size used was $n = 5/\text{group}$.

For the DC analysis, the effect size was estimated to be 0.791; resulting in a total sample size of $n = 4/\text{group}$. However, to account for potential losses, a sample size of $n=6/\text{group}$ was selected to be used in this study.

All sample size calculations were performed using the G*Power 3.1 software (Aichach, Germany) considering an $\alpha=0.05$ and power $(1-\beta) = 0.8$.

Specimen preparation

Forty discs (6 mm in diameter and 2 mm in thickness) were prepared and randomized into eight groups ($n = 5/\text{group}$) by a blinded trained operator. The material was inserted into a Teflon mold in increments

of 2 mm, covered with a polyester strip, and pressed against glass plates to prevent the formation of air bubbles and to remove excess material. The upper glass plate was removed, and the resin composite was light-cured using a light-curing unit (Radii-Cal LED, 1,000mW/cm²; SDI, Bayswater, Victoria, Australia). The intensity was monitored with a radiometer (Demetron; Kerr, Middleton, WI, USA) for each of the five specimens. The bottom surfaces of the discs were marked with a scalpel blade, stored in deionized water for 24 hours, and protected from light at 37°C.

The top surface was polished with sequential aluminum oxide discs (Sof-lex, Pop-on, 3M ESPE, St. Paul, USA): medium (10 s), fine (10 s), and extra fine grit sizes (10 s). After polishing, the specimens were placed in an ultrasonic bath for 5 minutes for elimination of debris.

The tests were performed in accordance with the ISO guidelines 4049.³⁸

Cycling protocol

This study followed the protocol proposed by Svizero et al.¹¹ The artificial aging process was performed with two daily immersion cycles of 10 minutes in 10 mL of undiluted grape juice (Suco de Uva Integral UniSabor, Indústria do Sucos 4 Léguas, Caxias do Sul, RS, Brazil) for 7 days. The grape juice had a pH of 3.47 and was kept at room temperature ($23 \pm 2^\circ\text{C}$). After each immersion, the specimens were washed and stored in deionized water at 37°C. The juice and water were replaced in each cycle. After 7 days, the specimens were subjected to an ultrasonic bath with deionized water for 5 minutes, dried with absorbent paper, and their final color was measured.¹¹

Colorimetric assessment by CIELab and CIEDE2000 systems

Color was assessed on the top surface of the specimens using a spectrophotometer (Easy Shade Advance Vita; Vita Zahnfabrik, Bad Säckingen, Baden-Württemberg, Germany) on a flat matte white standardized acrylic background under standardized lighting. Three measurements for each specimen were performed by a single blinded operator, and their average was calculated. Color readings were conducted at two different time points: after polishing

(at 24 hours; Δ_{baseline}) and after aging in grape juice (at 7 days; Δ_{final}). The spectrophotometer was calibrated for each specimen.

In CIELab (Equation 1) and CIEDE2000 (Equation 2), color systems used are based on three main parameters: L^* refers to luminosity ($L^* = 0 = \text{black}$; $L^* = 100 = \text{white}$), a^* indicates the chroma on the red-green axis ($a^* > 0 = \text{red}$ and $a^* < 0 = \text{green}$), and b^* the chroma on the yellow-blue axis ($b^* > 0 = \text{yellow}$; $b^* < 0 = \text{blue}$). CIEDE2000 sought to enhance the blue and gray color performance with a more specific equation, where $\Delta L'$, $\Delta C'$ and $\Delta H'$ represent luminosity, chroma, and hue, respectively. $\Delta R = RT (\Delta C' \times \Delta H')$ refers to the interaction between chroma and hue in the blue region. SL , SC , and SH are weighting functions that adjust the total color difference in L^* , a^* , and b^* coordinates. KL , KC , and KH are parametric factors that serve as correction terms for experimental conditions.³⁹

$$\Delta E_{\text{ab}}^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$$

Equation 1: CIELab

$$\Delta E_{00} = \left[\left(\frac{\Delta L'}{K_L S_L} \right)^2 + \left(\frac{\Delta C'}{K_C S_C} \right)^2 + \left(\frac{\Delta H'}{K_H S_H} \right)^2 + RT \left(\frac{\Delta C'}{K_C S_C} \right) \left(\frac{\Delta H'}{K_H S_H} \right) \right]^{1/2}$$

Equation 2: CIEDE2000

The 50:50 acceptability and perceptibility thresholds were adopted according to Ghinea et al.⁴⁰ The ΔE_{ab}^* values corresponding to 50% acceptability and perceptibility were 3.46 and 1.80, respectively. The ΔE_{00} values corresponding to 50% acceptability and perceptibility were 2.25 and 1.30, respectively.

Degree of conversion

The DC analysis was measured using FTIR (IRPrestige-21, Shimadzu, Tokyo, Japan) associated with an attenuated total reflectance (ATR) device.⁴⁰ Forty-eight discs were prepared ($n = 6/\text{group}$). For the initial reading of the unpolymerized material, the resin composite was inserted into the same Teflon mold ($6 \text{ mm} \times 2 \text{ mm}$) on the ATR crystal.

All materials were light-cured (Radii-Call LED, SDI, Bayswater, Victoria, Australia) at $1,000 \text{ mW}/\text{cm}^2$, according to the manufacturers' instructions (Table 1). The final reading (polymerized material) was conducted 3 minutes after light-curing.

The readings were conducted in the absorption mode within the spectral range of 4000 to 650 cm^{-1} and included 32 scans at a resolution of 4 cm^{-1} . DC was calculated based on changes in the intensity of aliphatic ($1636 \text{ cm}^{-1}/1638 \text{ cm}^{-1}$) and aromatic bonds (1608 cm^{-1} for NF and NH, and 1715 cm^{-1} for MH and S-PRG-NH), according to Equation 3.

Table 1. Classification of resin composite materials based on type of particles, commercial name, composition, opacity, and light-curing time.

Classification	Commercial name	Composition	Opacity	Light-curing	% Filled																				
Microhybrid	Gradia Direct – GC	UDMA, DMA, silica pre-polymers, silicon dioxide, fumed silica, silica glass, fluorine aluminum silicate glass.	A2	10 s	73.0wt%																				
			A2O	20 s		Nanofilled	Filtek Z350 - 3M ESPE	BisGMA, BisEMA, TEGDMA, silane-treated ceramic, silane-treated silica, silane-treated zirconia oxide, polyethylene glycol diethanedimethacrylate, BHT and pigments.	A2E	20 s	78.5wt%	A2D	20 s	Nanohybrid	Spectra Smart – Dentsply	Glass powder, silica, colloidal hydrophobe, DMA, benzophenone III, EDAB (photoinitiator), concentrate FluBlau, camphorquinone, BHT butylated hydroxytoluene, yellow iron oxide, red iron oxide, black iron oxide and titanium dioxide	A2	20 s	Information not disclosed by the manufacturer	A2O	20 s	Nanohybrid (S-PRG)	Beautiful II – Shofu	BisGMA, TEGDMA and Giomer technology (pre-activated glass particle with fluorine, strontium, sodium, boron, aluminum and silicate ions)	A2
Nanofilled	Filtek Z350 - 3M ESPE	BisGMA, BisEMA, TEGDMA, silane-treated ceramic, silane-treated silica, silane-treated zirconia oxide, polyethylene glycol diethanedimethacrylate, BHT and pigments.	A2E	20 s	78.5wt%																				
			A2D	20 s		Nanohybrid	Spectra Smart – Dentsply	Glass powder, silica, colloidal hydrophobe, DMA, benzophenone III, EDAB (photoinitiator), concentrate FluBlau, camphorquinone, BHT butylated hydroxytoluene, yellow iron oxide, red iron oxide, black iron oxide and titanium dioxide	A2	20 s	Information not disclosed by the manufacturer	A2O	20 s	Nanohybrid (S-PRG)	Beautiful II – Shofu	BisGMA, TEGDMA and Giomer technology (pre-activated glass particle with fluorine, strontium, sodium, boron, aluminum and silicate ions)	A2	10 s	83.3 wt%	A2O	10 s				
Nanohybrid	Spectra Smart – Dentsply	Glass powder, silica, colloidal hydrophobe, DMA, benzophenone III, EDAB (photoinitiator), concentrate FluBlau, camphorquinone, BHT butylated hydroxytoluene, yellow iron oxide, red iron oxide, black iron oxide and titanium dioxide	A2	20 s	Information not disclosed by the manufacturer																				
			A2O	20 s		Nanohybrid (S-PRG)	Beautiful II – Shofu	BisGMA, TEGDMA and Giomer technology (pre-activated glass particle with fluorine, strontium, sodium, boron, aluminum and silicate ions)	A2	10 s	83.3 wt%	A2O	10 s												
Nanohybrid (S-PRG)	Beautiful II – Shofu	BisGMA, TEGDMA and Giomer technology (pre-activated glass particle with fluorine, strontium, sodium, boron, aluminum and silicate ions)	A2	10 s	83.3 wt%																				
			A2O	10 s																					

*BisGMA: Bis-phenol A di-Glycidylmethacrylate, UDMA: Urethanedimethacrylate, TEGDMA: Triethyleneglycoldimethacrylate, BisEMA: Ethoxylatebisphenol A dimethacrylate, EDAB: dimethylaminoethylbenzoate, BHT: 2,6-di-tert-butyl-p-cresol, DMA: dimethacrylate.

$$DC = \left(1 - \frac{R_{\text{cured}}}{R_{\text{uncured}}}\right) \times 100$$

Equation 3: Degree of conversion

Statistical analysis

Data were entered into Microsoft Excel spreadsheets (Excel 2016; Microsoft, Redmond, USA) and analyzed for normal distribution and homogeneity using Sigma Plot software (Systat Software, Inc., San Jose, USA).

For color alteration, the data were subjected to two-way analysis of variance (ANOVA) and post-hoc least significant difference (LSD) tests. Two-way ANOVA and post-hoc LSD tests were applied for DC analysis. The significance level was set at 5% for all tests.

Table 2. Means and standard deviations of ΔE values of resin composites analyzed by the CIELab and CIEDE2000 equations.

Resin composites	T/E	D/O
CIELab		
Microhybrid	3.64 (1.30) b	3.64 (0.58) b
Nanofilled	7.24 (0.99) d	5.09 (0.44) c
Nanohybrid	2.63 (0.44) ab	2.47 (0.72) a
Nanohybrid (S-PRG)	8.24 (0.86) d	9.49 (1.22) e
CIEDE2000		
Microhybrid	2.39 (0.86) bc	2.11 (0.40) b
Nanofilled	4.62 (0.58) d	3.14 (0.32) c
Nanohybrid	1.65 (0.27) ab	1.27 (0.48) a
Nanohybrid (S-PRG)	5.43 (0.65) e	6.15 (0.84) e

n = 5/group. T/E: translucent/enamel; D/O: dentin/opaque. Lowercase letters indicate statistical difference between type of materials and opacities.

RESULTS

Colorimetric assessment by CIELab and CIEDE2000 systems

Both homogeneity (p = 0.164 for CIELab and p = 0.135 for CIEDE2000) and normality (p = 0.370 for CIELab and p = 0.656 for CIEDE2000) of the data were tested. Statistically significant differences were found for the type of fillers (CIELab and CIEDE2000, p = 0.0001) and an interaction between the type of filler and opacity (CIELab, p = 0.0015; CIEDE2000, p = 0.0026) was observed. No significant difference was found for the opacity factor (CIELab, p = 0.348; CIEDE2000, p = 0.0645). The mean ΔE values for the CIELab and CIEDE2000 systems are presented in Table 2. Table 3 shows the mean value of each analyzed parameter in a descriptive way.

Based on the CIELab equation, among the T/E resin composites, NH presented the lowest ΔE*_{ab}, which did not differ from that of MH. The greatest ΔE*_{ab} was observed for S-PRG-NH, which did not differ from NF; however, both differed from MH and NH.

For the D/O resin composites, all groups were different. The lowest ΔE*_{ab} was also noted for NH, which differed from MH, NF, and S-PRG-NH. The highest ΔE*_{ab} was observed for S-PRG-NH, which differed from all others.

Nonetheless, despite these results, all ΔE*_{ab} values of both T/E and D/O composites were classified as clinically notable (> 3.46), except for NH, whose ΔE*_{ab} was the only one classified as clinically perceptible (> 1.8).

Regarding the CIEDE2000 equation, favorable outcomes were also detected for T/E composites in the case of NH and MH, which did not differ between

Table 3. Descriptive means of color coordinate values for the analyzed resin composites.

Variable	T/E					D/O				
	ΔL	ΔC	ΔH	Δa	Δb	ΔL	ΔC	ΔH	Δa	Δb
MH	-3.40	-1.15	-0.45	0.00	-1.18	-3.02	-1.84	-0.05	-0.28	-1.83
NF	-6.13	-3.69	2.51	-1.07	-3.68	-4.27	-2.94	0.77	-0.89	-2.76
NH	-2.43	-0.92	-0.51	0.16	-0.94	-1.69	-1.71	-0.25	-0.05	-1.71
S-PRG-NH	-7.71	-2.35	0.47	-0.52	-2.29	-9.07	-2.98	0.86	-0.57	-2.70

MH: microhybrid; NF: nanofilled; NH: nanohybrid; NH-S-PRG: nanohybrid (S-PRG); T/E: translucent/enamel; D/O: dentin/opaque.

them. The more perceptible changes were attributed to S-PRG-NH, which differed from those of the other materials. NF presented intermediate values, which differed from all groups.

Among the D/O composites for CIEDE2000, a performance similar to that of CIELab was verified. NH presented the lowest ΔE_{00} , which differed from those of the other materials. MH and NF presented intermediate values of ΔE_{00} , differing from each other and from both NH and S-PRG-NH. The S-PRG-NH material showed the most perceptible results and differed from all materials.

According to the established thresholds, all ΔE_{00} values of T/E were classified as clinically unacceptable (> 2.25), except for NH, whose ΔE_{ab}^* values were classified as clinically perceptible (> 1.3). For D/O opacity, the materials were classified as clinically

unacceptable (> 2.25), except for MH and NH materials, which were within the acceptable range (< 1.3).

Degree of conversion

The homogeneity ($p = 0.002$) and normality ($p = 0.109$) of the data were assessed. Statistically significant differences were found between the evaluated materials, opacities, and their interaction ($p < 0.0001$). The DC values are listed in Table 4 and representative spectra of FTIR analyses before and after light-curing are presented in Figure.

In the case of T/E specimens, NF and NH presented higher DC compared to MH and S-PRG-NH ($p < 0.05$). Among O/D specimens, S-PRG-NH and MH showed a lower DC than did other materials with the same opacity ($p < 0.05$). Significant differences between the opacities were observed only for NF. For this material, the translucent version showed a higher DC than the opaque version ($p < 0.05$).

Table 4. Degree of conversion (%) and standard deviation of the tested groups.

Resin composites	Opacity	
	T/E	D/O
Microhybrid	43.06 (3.49) cd	44.72 (2.57) cd
Nanofilled	59.36 (4.85) a	46.47 (6.07) c
Nanohybrid	55.43 (2.85) ab	49.53 (2.65) bc
Nanohybrid (S-PRG)	43.96 (2.67) cd	39.32 (0.79) d

N=6/group. T/ E: translucent/ enamel; D/O: dentin/ opaque. Lowercase letters indicate statistical difference between type of materials and opacities.

Discussion

Discoloration of restorative materials occurs for different reasons, such as DC, titratable acidity, food colorant absorption and penetration,^{1,24,32} and the size and amount of inorganic filler.⁴⁰ The tooth substrate and surface properties of the materials can also be included, as well as the type of polishing performed.^{3,6,9} In the present study, for both CIELab and CIEDE2000

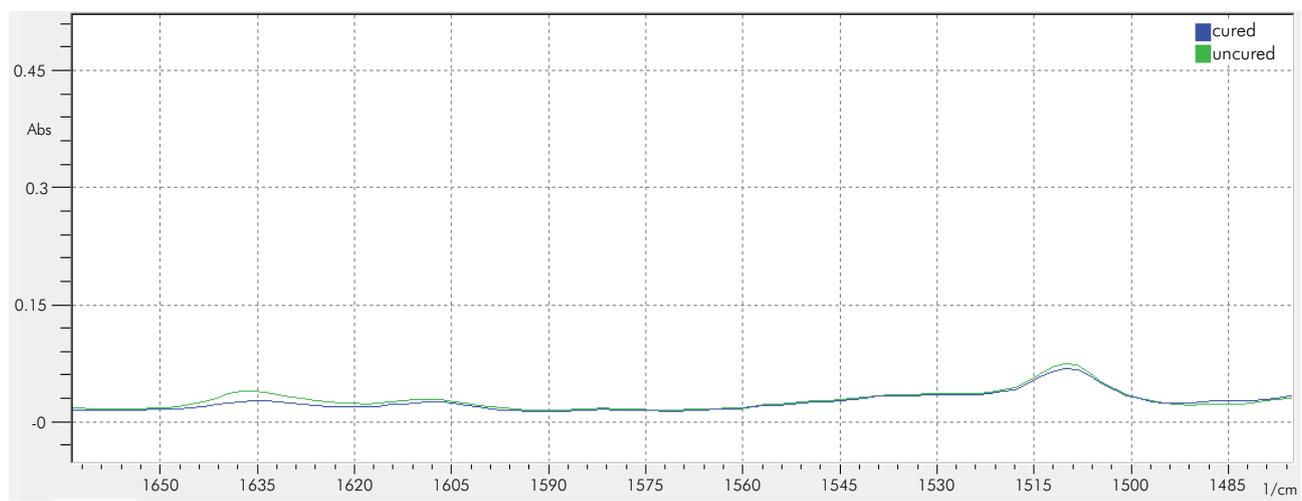


Figure. Representative FTIR spectra of a resin composite before and after light-curing.

equations, S-PRG-NH showed the highest values for color alteration, followed by NF, MH, and NH, which presented the lowest ΔE values. Regarding opacities, NF and S-PRG-NH showed difference in terms of color stability for CIELab, which was noted only for NF in the case of CIEDE2000. Therefore, the first null hypothesis was rejected.

In clinical practice, layering of different colors and opacities of resin composites is recommended in order to achieve excellent aesthetic outcomes that closely resemble natural teeth. The overall discoloration of this material can result from extrinsic factors (*e.g.*, pigments from food and beverages) or intrinsic factors, such as the composition of the organic matrix, size and amount of the inorganic matrix, photoinitiators, and DC.^{24,40} The opacity and translucency of these materials can also be influenced by their composition, which is mainly determined by the difference in refractive index between the organic and inorganic matrix and by the amount and size of the filler.⁴⁰ This difference between the opacities could influence their response to discoloration; however, most materials showed no difference in color change between the two opacities (T/E and D/O) in this study, except for NF (CIELab and CIEDE2000) and S-PRG-NH (CIEDE2000).

This alteration was evident for all evaluated parameters, particularly in the case of NF-T/E compared to NF-D/O, which explains the difference of ΔE between their opacities. Although all evaluated resins showed a decrease in luminosity (ΔL), this alteration was more evident in S-PRG-NH, which was the most affected parameter. The presence of S-PRG particles in resin composites based on this technology leads to a more whitish appearance, affecting the perception of luminosity of these materials. The greater opacity of D/O group also contributed to the fact that the decrease in ΔL values became more evident for this material. In addition, both resins showed an increase in hue (ΔH) values, in contrast to MH and NH, whose values decreased. In general, the yellow-blue axis (Δb) was affected, in line with the bluish coloration of the substance used to age the specimens.

The mechanism of action of S-PRG fillers depends on water diffusion,³⁷ which requires higher amounts of hydrophilic monomers and higher water sorption as compared to materials not subjected to this

technology.^{20,24,25} Previous studies have shown a positive correlation between water sorption and staining,^{24,25} which could explain the higher staining observed for S-PRG-based composites. Nonetheless, as water is absorbed, expansion and plasticization of the resin and hydrolysis of the silane bonds occur, which can generate microcracks and the release of residual monomers.¹¹ This could also favor staining and compromise the aesthetic appearance of the restoration.^{28,32} Moreover, as silane bonds are degraded, the filler particles of the resin can start to detach from the organic matrix, contributing to the increase in their staining potential due to increased roughness and pigment retention.^{28,32,37}

Studies have shown that the solubility of BisGMA-based composites increases when associated with triethylene glycol dimethacrylate (TEGDMA) and decreases with urethane dimethacrylate (UDMA).^{24,29} These findings could explain the findings of the present study, in which greater staining was seen for BisGMA and TEGDMA-based composites (S-PRG-NH and NF), indicated mainly by a greater change in the ΔL^* parameter,²⁹ corroborating the findings of this study. The high water sorption promoted by the association of BisGMA and TEGDMA, combined with the mechanism of S-PRG fillers, could explain the staining of the materials shown in this study. The manufacturers did not specify the dimethacrylates in the tested NH and MH resins, except for the UDMA diluent used in MH. The presence of UDMA in the organic matrix could explain the higher resistance of MH to staining, but the lack of this information precludes a more robust interpretation of the results.

In this case, limitations can also be extended to other properties, and the balance of the main properties needs to be considered for the appropriate recommendation of each material.⁴

The color stability of resin composites has also been strongly associated with their DC.³¹ In this study, S-PRG-NH and MH presented the lowest DC, while NH and NF materials showed the best percentages of monomer-to-polymer conversion. A higher opacity can also influence the passage of light through the material, thus decreasing DC and leading to greater susceptibility to discoloration.³¹ The impact of this property was only observed for

NF, which showed lower values for its D/O version compared to T/E. Therefore, the second null hypothesis was also rejected. Incomplete light-curing generates a greater amount of residual monomers, which are easily degraded, resulting in greater susceptibility to pigmentation.³³

Differences in the organic matrix composition of methacrylate and the size, type, and volume of particles can affect the depth of light curing and scattering, and consequently, the DC.³⁰ Hence, the volume and size of fillers in the composition of the S-PRG-based resin composite used in this study could have influenced the DC results. Ilie and Fleming³⁴ compared different materials, including one with S-PRG fillers similar to this study, and the poor performance was attributed to the presence of the filler.³⁵ This technology shows larger particles when compared with other technologies, and it is associated with more filler percentage, hindering light penetration into deep layers, thus decreasing the DC.³⁴ S-PRG-NH presented a percentage of inorganic fillers of 83.3wt% against 73.0wt% of MH and 78.5wt% of NF. The percentage of fillers in the NH resin was not provided by the manufacturer.

Additionally, it is known that TEGDMA increases the DC,³⁵ which could have contributed to the higher values observed for NF. The similar performance of S-PRG-NH, which also contains TEGDMA monomer, can be limited by these fillers, as mentioned above. Nevertheless, the presence of TEGDMA can impair the mechanical properties³⁵ because the monomers used in the formulation of these composites are strongly related to the staining potential and DC of the material. Future studies addressing the physicochemical properties of these resins should be conducted.

Therefore, based on the analyses in the present study, MH and NH could be used as an excellent aesthetic treatment alternative. In contrast, S-PRG-NH should be indicated in specific situations. Gordan

et al. evidenced optimal performance of posterior restorations using Beautifill II.¹⁹ In particular, in Class II situations, this 13-year follow-up proved its clinical effectiveness. In a recent study, Toz-Akalin et al. conducted a 2-year follow-up and demonstrated how this material containing low-shrinkage organic monomers and reduced amount of S-PRG can yield more interesting results under service conditions.⁵ Therefore, as reported in other studies, clinical circumstances, such as daily brushing¹⁰ and polishing protocols,¹² may also play important roles in the longevity of restorative materials. Future studies should investigate these parameters, given that repolishing can re-establish the original color of the restorative material and achieve clinically acceptable levels of aesthetic longevity.¹²

An equilibrium between the *in vitro* and *in vivo* performance of the materials promotes robust interpretation, aiding professionals to choose materials and techniques in different circumstances and assuring more precise benefits to the patients.

Conclusions

Despite the limitations of this study, it can be concluded that the nanohybrid resin composite presented favorable overall performance, which is likely related to its more stable organic content. However, the use of S-PRG-based nanohybrid resins in aesthetic procedures should take into account their greater susceptibility to color alteration, probably due to the water-based bioactive mechanism of action. The opacity of the material may also affect the color stability and the DC of some materials.

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References

1. Villavicencio-Espinoza CA, Giacomini MC, Narimatsu MH, Magalhães AC, Atta MT, Wang L. Adapted three-step restorative technique: recovering dental substrate compromised by complex erosive wear in a young patient. *Oper Dent*. 2020 Sep;45(5):457b-466. <https://doi.org/10.2341/18-204-S>

2. Choi JW, Lee MJ, Oh SH, Kim KM. Changes in the physical properties and color stability of aesthetic restorative materials caused by various beverages. *Dent Mater J*. 2019 Feb;38(1):33-40. <https://doi.org/10.4012/dmj.2017-247>
3. Tanthanuch S, Kukiattrakoon B, Siriporananon C, Ornprasert N, Mettasilthikorn W, Likhitpreeda S, et al. The effect of different beverages on surface hardness of nanohybrid resin composite and giomer. *J Conserv Dent*. 2014 May;17(3):261-5. <https://doi.org/10.4103/0972-0707.131791>
4. He L, Hao Y, Zhen L, Liu H, Shao M, Xu X, et al. Biomineralization of dentin. *J Struct Biol*. 2019 Aug;207(2):115-22. <https://doi.org/10.1016/j.jsb.2019.05.010>
5. Toz-Akalin T, Öztürk-Bozkurt F, Kusdemir M, Özsoy A, Yüzbaşıoğlu E, Özcan M. Clinical evaluation of low-shrinkage bioactive material Giomer versus nanohybrid resin composite restorations: a two-year prospective controlled clinical trial. *Oper Dent*. 2023 Jan;48(1):10-20. <https://doi.org/10.2341/21-155-C>
6. Barakah HM, Taher NM. Effect of polishing systems on stain susceptibility and surface roughness of nanocomposite resin material. *J Prosthet Dent*. 2014 Sep;112(3):625-31. <https://doi.org/10.1016/j.prosdent.2013.12.007>
7. Duc O, Di Bella E, Krejci I, Betrisey E, Abdelaziz M, Ardu S. Staining susceptibility of resin composite materials. *Am J Dent*. 2019 Feb;32(1):39-42. <https://doi.org/10.29328/journal.jcad.1001006>
8. Oliveira GU, Mondelli RF, Charantola Rodrigues M, Franco EB, Ishikiriama SK, Wang L. Impact of filler size and distribution on roughness and wear of composite resin after simulated toothbrushing. *J Appl Oral Sci*. 2012;20(5):510-6. <https://doi.org/10.1590/S1678-77572012000500003>
9. Zhao X, Zanetti F, Wang L, Pan J, Majeed S, Malmstrom H, et al. Effects of different discoloration challenges and whitening treatments on dental hard tissues and composite resin restorations. *J Dent*. 2019 Oct;89:103182. <https://doi.org/10.1016/j.jdent.2019.103182>
10. Schroeder T, da Silva PB, Basso GR, Franco MC, Maske TT, Cenci MS. Factors affecting the color stability and staining of esthetic restorations. *Odontology*. 2019 Oct;107(4):507-12. <https://doi.org/10.1007/s10266-019-00421-x>
11. Svizero NR, Góes AR, Bueno TL, Di Hipólito V, Wang L, D'Alpino PH; SvizeroNda R. Micro-sized erosions in a nanofilled composite after repeated acidic beverage exposures: consequences of clusters dislodgments. *J Appl Oral Sci*. 2014;22(5):373-81. <https://doi.org/10.1590/1678-775720130658>
12. Liebermann A, Langwieder B, Brauneis M, Eichberger M, Stawarczyk B. Impact of thermocycling on mechanical properties and discoloration of veneering composite resins after storage in various staining media. *J Prosthet Dent*. 2021 Jun;125(6):940-5. <https://doi.org/10.1016/j.prosdent.2020.03.030>
13. Sabbagh J, Ryelandt L, Bachérius L, Biebuyck JJ, Vreven J, Lambrechts P, et al. Characterization of the inorganic fraction of resin composites. *J Oral Rehabil*. 2004 Nov;31(11):1090-101. <https://doi.org/10.1111/j.1365-2842.2004.01352.x>
14. Kooi TJ, Tan QZ, Yap AU, Guo W, Tay KJ, Soh MS. Effects of food-simulating liquids on surface properties of giomer restoratives. *Oper Dent*. 2012;37(6):665-71. <https://doi.org/10.2341/11-419-L>
15. Soares CJ, Faria-E-Silva AL, Rodrigues MP, Vilela AB, Pfeifer CS, Tantbirojn D, et al. Polymerization shrinkage stress of composite resins and resin cements - What do we need to know? *Braz Oral Res*. 2017 Aug;31 suppl 1:e62. <https://doi.org/10.1590/1807-3107bor-2017.vol31.0062>
16. Prodan DA, Gasparik C, Mada DC, Miclăuş V, Băciuş M, Ducea D. Influence of opacity on the color stability of a nanocomposite. *Clin Oral Investig*. 2015 May;19(4):867-75. <https://doi.org/10.1007/s00784-014-1315-1>
17. Hyun HK, Ferracane JL. Influence of biofilm formation on the optical properties of novel bioactive glass-containing composites. *Dent Mater*. 2016 Sep;32(9):1144-51. <https://doi.org/10.1016/j.dental.2016.06.011>
18. Fujimoto Y, Iwasa M, Murayama R, Miyazaki M, Nagafuji A, Nakatsuka T. Detection of ions released from S-PRG fillers and their modulation effect. *Dent Mater J*. 2010 Aug;29(4):392-7. <https://doi.org/10.4012/dmj.2010-015>
19. Gordan VV, Blaser PK, Watson RE, Mjör IA, McEdward DL, Sensi LG, et al. A clinical evaluation of a giomer restorative system containing surface pre-reacted glass ionomer filler: results from a 13-year recall examination. *J Am Dent Assoc*. 2014 Oct;145(10):1036-43. <https://doi.org/10.14219/jada.2014.57>
20. Kitagawa H, Miki-Oka S, Mayanagi G, Abiko Y, Takahashi N, Imazato S. Inhibitory effect of resin composite containing S-PRG filler on *Streptococcus mutans* glucose metabolism. *J Dent*. 2018 Mar;70:92-6. <https://doi.org/10.1016/j.jdent.2017.12.017>
21. Bergantin BT, Di Leone CC, Cruvinel T, Wang L, Buzalaf MA, Borges AB, et al. S-PRG-based composites erosive wear resistance and the effect on surrounding enamel. *Sci Rep*. 2022 Jan;12(1):833. <https://doi.org/10.1038/s41598-021-03745-3>
22. Spinola MD, Moecke SE, Rossi NR, Nakatsuka T, Borges AB, Torres CR. Efficacy of S-PRG filler containing varnishes on enamel demineralization prevention. *Sci Rep*. 2020 Nov;10(1):18992. <https://doi.org/10.1038/s41598-020-76127-w>
23. Nomura R, Morita Y, Matayoshi S, Nakano K. Inhibitory effect of surface pre-reacted glass-ionomer (S-PRG) eluate against adhesion and colonization by *Streptococcus mutans*. *Sci Rep*. 2018 Mar;8(1):5056. <https://doi.org/10.1038/s41598-018-23354-x>
24. Gonulol N, Ozer S, Sen Tunc E. Water sorption, solubility, and color stability of Giomer restoratives. *J Esthet Restor Dent*. 2015;27(5):300-6. <https://doi.org/10.1111/jerd.12119>

25. de Lima Navarro MF, Pascotto RC, Borges AF, Soares CJ, Raggio DP, Rios D, et al. Consensus on glass-ionomer cement thresholds for restorative indications. *J Dent.* 2021 Apr;107:103609. <https://doi.org/10.1016/j.jdent.2021.103609>
26. Khokhar ZA, Razzoog ME, Yaman P. Color stability of restorative resins. *Quintessence Int.* 1991 Sep;22(9):733-7.
27. Fugolin AP, Paula AB, Dobson A, Huynh V, Consani R, Ferracane JL, et al. Alternative monomer for BisGMA-free resin composites formulations. *Dent Mater.* 2020 Jul;36(7):884-92. <https://doi.org/10.1016/j.dental.2020.04.009>
28. Ferracane JL. Hygroscopic and hydrolytic effects in dental polymer networks. *Dent Mater.* 2006 Mar;22(3):211-22. <https://doi.org/10.1016/j.dental.2005.05.005>
29. Sarafianou A, Iosifidou S, Papadopoulos T, Eliades G. Color stability and degree of cure of direct composite restoratives after accelerated aging. *Oper Dent.* 2007;32(4):406-11. <https://doi.org/10.2341/06-127>
30. Knezević A, Tarle Z, Meniga A, Sutalo J, Pichler G, Ristić M. Degree of conversion and temperature rise during polymerization of composite resin samples with blue diodes. *J Oral Rehabil.* 2001 Jun;28(6):586-91. <https://doi.org/10.1046/j.1365-2842.2001.00709.x>
31. Schneider LF, Pfeifer CS, Consani S, Pahl SA, Ferracane JL. Influence of photoinitiator type on the rate of polymerization, degree of conversion, hardness and yellowing of dental resin composites. *Dent Mater.* 2008 Sep;24(9):1169-77. <https://doi.org/10.1016/j.dental.2008.01.007>
32. Silva EM, Dória J, Silva JJ, Santos GV, Guimarães JG, Poskus LT. Longitudinal evaluation of simulated toothbrushing on the roughness and optical stability of microfilled, microhybrid and nanofilled resin-based composites. *J Dent.* 2013 Nov;41(11):1081-90. <https://doi.org/10.1016/j.jdent.2013.08.013>
33. Samra AP, Pereira SK, Delgado LC, Borges CP. Color stability evaluation of aesthetic restorative materials. *Braz Oral Res.* 2008;22(3):205-10. <https://doi.org/10.1590/S1806-83242008000300003>
34. Ilie N, Fleming GJ. In vitro comparison of polymerisation kinetics and the micro-mechanical properties of low and high viscosity comomers and RBC materials. *J Dent.* 2015 Jul;43(7):814-22. <https://doi.org/10.1016/j.jdent.2015.04.009>
35. Emami N, Söderholm K. Young's modulus and degree of conversion of different combination of light-cure dental resins. *Open Dent J.* 2009 Oct;3(1):202-7. <https://doi.org/10.2174/1874210600903010202>
36. Demarco FF, Collares K, Coelho-de-Souza FH, Correa MB, Cenci MS, Moraes RR, et al. Anterior composite restorations: A systematic review on long-term survival and reasons for failure. *Dent Mater.* 2015 Oct;31(10):1214-24. <https://doi.org/10.1016/j.dental.2015.07.005>
37. Yoshihara K, Nagaoka N, Maruo Y, Sano H, Yoshida Y, Van Meerbeek B. Bacterial adhesion not inhibited by ion-releasing bioactive glass filler. *Dent Mater.* 2017 Jun;33(6):723-34. <https://doi.org/10.1016/j.dental.2017.04.002>
38. International Organization for Standardization. ISO 4049. Dentistry — Polymer-based filling, restorative, and luting materials. Geneva: International Organization for Standardization; 2009.
39. Ghinea R, Pérez MM, Herrera LJ, Rivas MJ, Yebra A, Paravina RD. Color difference thresholds in dental ceramics. *J Dent.* 2010;38 Suppl 2:e57-64. <https://doi.org/10.1016/j.jdent.2010.07.008>
40. Collares FM, Portella FF, Leitune VC, Samuel SM. Discrepancies in degree of conversion measurements by FTIR. *Braz Oral Res.* 2013;27(6):453-4. <https://doi.org/10.1590/S1806-83242013000600002>