



Physico-mechanical characterization and fracture reliability of dental resin composites for enamel restoration

Carianne Mendes de Almeida¹ · Evandro Piva¹ · Camila Gonçalves Duarte² · Henrique Timm Vieira² · Cristina Pereira Isolan¹ · Lisia Lorea Valente¹ · Eliseu Aldrighi Münchow³

Received: 2 May 2019 / Accepted: 23 August 2019 / Published online: 10 September 2019
© The Brazilian Society of Mechanical Sciences and Engineering 2019

Abstract

This study evaluated the physico-mechanical behavior and the fracture reliability of different dental resin composites indicated for the restoration of the highly mineralized enamel. The following resin composites were tested: Concept Advanced (Vigodent), Fill Magic (Vigodent), Llis (FGM), and Natural Look (DFL); Filtek Z250 (3M ESPE) was used as a universal control. All composites were fully characterized with the following tests: degree of conversion (DC), flexural strength (σ), flexural modulus (E), water sorption (WS) and solubility (SL), hardness (KMN). Scanning electron microscopy was used for morphological evaluation, whereas the topographical evaluation was carried out by profilometry (Ra) and atomic force microscopy. The color stability of the resin composites was also assessed using a digital spectrophotometer and the CIEL^{*}*a*^{*}*b*^{*} system. Data were analyzed with ANOVA as well as the Weibull analysis ($\alpha=5\%$). DC was similar among all composites (51.0–62.3%, $p=0.104$). Llis and the control showed overall greater mechanical performance and hardness than the others. Llis and Natural Look presented lower WS and SL than the others. The control acquired smoother surface than the enamel composites. Concept Advanced and Natural Look demonstrated the greatest and lowest reliability of the study, respectively, whereas Llis showed the greatest characteristic strength ($p<0.05$). Llis showed the greatest color stability of the study after 28 days of water storage ($p<0.05$). In conclusion, our findings confirmed the performance of four different resin composites indicated to restore dental enamel, showing that two of them (Llis and Natural Look) can be sufficiently strong for the restoration of stress-receiving areas, such as the posterior dentition. Most of the enamel composites were as reliable as the universal control resin composite.

Keywords FTIR · Scanning electron microscopy · Flexural strength · Water sorption and solubility · Hardness · Weibull analysis

1 Introduction

Dental esthetic restorative treatments are widely desired by patients today, with resin composites being the most common direct restorative materials used, especially due to their satisfactory mechanical and physical properties for restoring both posterior and anterior teeth [1, 2]. Of note, resin composites have evolved since their development in the early 1960s: while macrofilled composites (i.e., comprised of larger fillers of ~ 15 μm in size) were difficult to retain the requested gloss and surface smoothness after polishing, submicron-sized (< 1 μm) and nanosized (< 100 nm) composites are currently considered the state of the art of direct restoratives [3–6]. Concerning the filler phase of resin composites, it has been demonstrated that bi- or tri-modal materials (i.e., those comprised of fillers of different size)

Technical Editor: Paulo de Tarso Rocha de Mendonça, Ph.D.

✉ Eliseu Aldrighi Münchow
eliseu.munchow@ufrgs.br

¹ Graduate Program in Dentistry, School of Dentistry, Federal University of Pelotas, Pelotas, RS, Brazil

² Center of Development and Control of Biomaterials (CDC-Bio), Federal University of Pelotas, Pelotas, RS, Brazil

³ Department of Conservative Dentistry, School of Dentistry, Federal University of Rio Grande do Sul, Rua Ramiro Barcelos, 2492, Porto Alegre, RS 90035-004, Brazil

may possess greater wear resistance, hardness, and strength characteristics than monomodal composites, denoting that the quality of fillers and their related size configuration play a significant role on the overall properties of resin-based restorations [5].

The combination of resin composites to mimic the natural tooth appearance is easy to perform, and specific materials are available to restore both dentin and enamel substrates [7]. The main difference between dentin and enamel composites relies on their fillers' volume, shape, and size [5, 8], as well as on the type of pigments incorporated that could influence optical appearance of the material [9]. Although the use of enamel composites involves a smaller proportion when compared with composites indicated to restore dentin, i.e., only to coat the "body" of the restoration, the direct impact and stresses originated from oral function and mastication processes are expected to occur within the former restorative materials. Moreover, oral fluids and beverages present in the human diet may favor the discoloration of restorations in a time-progressive manner, thus affecting directly the outmost layers of the restoration [6]. Consequently, enamel composites should present satisfactory properties and wear resistance; otherwise, the restorative treatment may unavoidably fail in a short period of time. Taking this into consideration, it is extremely important to investigate the physico-mechanical characteristics and color stability of enamel composites, especially because this class of restorative material has been still poorly investigated by laboratory and clinical studies, since research is more focused on the performance of dentin resin composites [10].

Hence, the purpose of this study was to perform a full characterization on enamel resin composites by means of important physico-mechanical properties. The null hypothesis tested was that enamel resin composites would perform similarly to a thoroughly used universal resin composite (control).

2 Materials and methods

Four enamel resin composites, shade A1, were evaluated: Concept Advanced (Vigodent, Rio de Janeiro, RJ, Brazil), Fill Magic (Vigodent), Llis (FGM, Joinville, SC, Brazil), and Natural Look (DFL, Rio de Janeiro, RJ, Brazil). Filtek™ Z250 (3M ESPE, St. Paul, MN, USA) was used as control, since it is a universal composite indicated for the restoration of both dentin and enamel substrates. The manufacturer, type of composite, lot number, and chemical composition of each resin composite are illustrated in Table 1. Figure 1 shows a scheme of materials and all properties tested in the study.

2.1 Scanning electron microscopy

One disk-shaped (6 mm diameter × 1 mm thickness) sample was prepared from each resin composite using a metallic mold. Each material was inserted using a plastic spatula and properly adapted into the mold, which was covered with a mylar strip and a glass coverslip. Each sample was light-activated using a light-emitting diode (LED) light-curing unit (Radii; SDI, Bayswater, VIC, Australia) for 40 s at both the top and bottom surfaces. The irradiance of the LED was constant (900 mW/cm²). All samples were polished with wet #600-, #1200-, #1500-, #2000-, and #2500-grit abrasive SiC papers, followed by 5, 1, and 0.1 μm aluminum oxide solutions. The samples were mounted on aluminum stubs, sputter-coated with gold/palladium, and evaluated using a scanning electron microscope (SSX-550, Shimadzu, Tokyo, Japan) for qualitative analysis of their microstructure and surface morphology.

Table 1 Information regarding the manufacturer, batch number, and composition of the resin composites evaluated in the study

| Resin composites (type) | Manufacturer | Lot | Chemical composition |
|-------------------------------|--|----------|---|
| Concept Advanced (nanofilled) | Vigodent Coltene SA Indústria e Comércio, Rio de Janeiro, RJ, Brazil | 261/10 | Bis-GMA, UDMA, methacrylic acid ester, barium, and aluminum silicate |
| Fill Magic (hybrid) | Vigodent | 119/09 | Bis-GMA, Bis-EMA, UDMA, TEGDMA, photoinitiator, fillers, and pigments |
| Llis (micro-hybrid) | FGM, Joinville, SC, Brazil | 300909 | Bis-GMA, Bis-EMA, TEGDMA, CQ, co-initiator, silane, micrometric barium and aluminum silicate, oxides, and nanometric silica |
| Natural Look (micro-hybrid) | Nova DFL, Rio de Janeiro, RJ, Brazil | 11020318 | Bis-GMA, Bis-DMA, TEGDMA, silanized barium crystals, and hydrophobic amorphous silica |
| Filtek™ Z250 (micro-hybrid) | 3M ESPE, St. Paul, MN, USA | 6020A2 | Bis-GMA, UDMA, Bis-EMA, photoinitiator, zirconium glass fillers, and colloidal silica |

Bis-GMA bisphenol A diglycidyl dimethacrylate, *UDMA* urethane dimethacrylate, *Bis-EMA* bisphenol A polyethylene glycol diether dimethacrylate, *TEGDMA* triethyleneglicol dimethacrylate, *CQ* camphorquinone, *Bis-DMA* bisphenol A diethoxymethacrylate

Universal resin composite (control)



Filtek Z250
(3M ESPE)

Resin composites for dental enamel restoration



Concept
Advanced
(Vigodent)



Fill Magic
(Vigodent)



Llis
(FGM)



Natural Look
(Nova DFL)

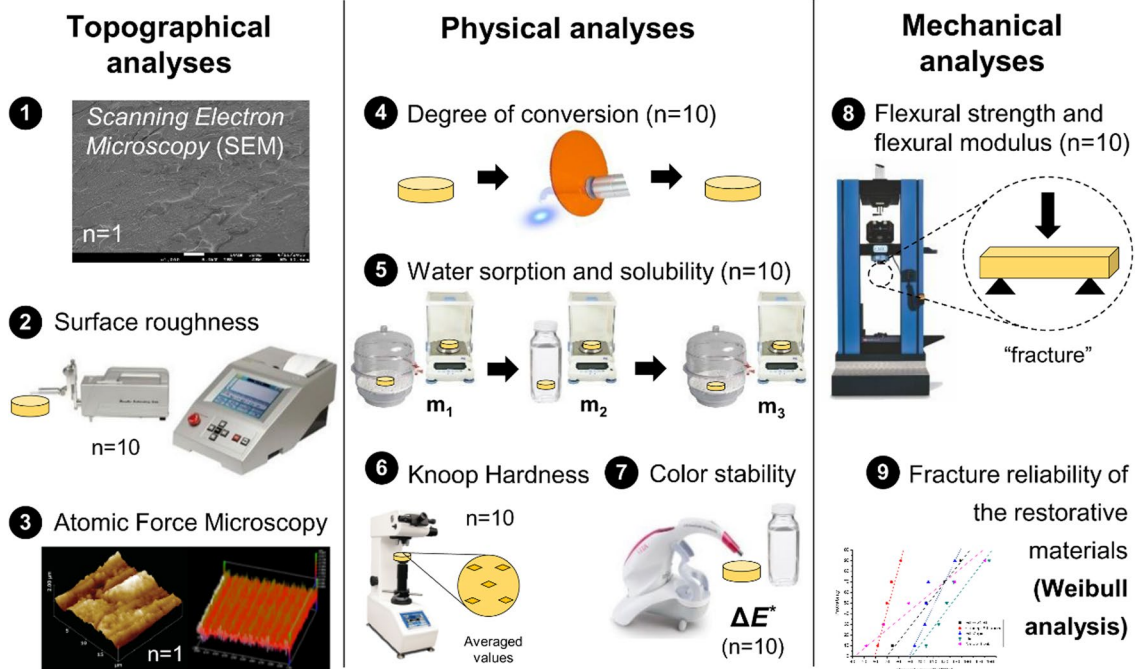


Fig. 1 Scheme demonstrating the dental resin composites and properties investigated in this study. Four enamel resin composites (Concept Advanced, Fill Magic, Llis, and Natural Look) were compared

2.2 Degree of conversion

The degree of conversion (DC) of each resin composite ($n = 10$) was evaluated using Fourier-transformed infrared (FTIR) spectroscopy (Prestige21; Shimadzu, Tokyo, Japan). The spectrometer was equipped with an attenuated total reflectance device composed of a diamond crystal, and configuration was as follows: Happ-Genzel appodization (at a range of $1750\text{--}1550\text{ cm}^{-1}$), resolution of 8 cm^{-1} , and mirror speed of 2.8 mm/s . The preliminary reading of the unpolymerized material was taken from a standard volume of material ($\sim 0.5\text{ mm}$ -thick). Light activation was performed for 40 s, and a final reading of the polymerized material was carried out. The DC was then calculated

to a universal material (control; Filtek Z250). All the materials were tested by means of topographical, physical, and mechanical analyses

as previously described [11] using the following formula (Eq. 1):

$$DC = 1 - \frac{(1635\text{ cm}^{-1}/1608\text{ cm}^{-1})_{\text{cured}}}{(1635\text{ cm}^{-1}/1608\text{ cm}^{-1})_{\text{uncured}}} \times 100\% \quad (1)$$

2.3 Flexural strength and flexural modulus

Bar-shaped samples were prepared from each resin composite using a metallic mold (25 mm length \times 2 mm width \times 2 mm thickness) ($n = 10$). The materials were placed into the mold as described before, but light-activated using a different protocol, as follows: light activation was performed for 120 s at the top

and bottom surfaces of the mold, at a fixed distance of 10 mm from the LED source [12]. Each sample was polished with abrasive SiC papers (#600 and #1200 grit) to remove surface irregularities, followed by storage in distilled water at 37 °C for 24 h. The samples were submitted to three-point bending flexural test on a mechanical testing machine (DL500; EMIC, São José dos Pinhais, PR, Brazil) at a crosshead speed of 0.5 mm/min. Data for flexural strength (σ) and flexural modulus (E) were obtained from the stress–strain curves, using the following formulas (Eqs. 2 and 3, respectively):

$$\sigma = \frac{3Fl}{2bh^2} \quad (2)$$

$$E = \frac{Fl^3}{4bh^3d} \quad (3)$$

where F is the peak load (in N); l is the span length (in mm); b and h are, respectively, the width and the thickness of the specimen (in mm); and d is the deflection of the specimen at load F during the straight line portion of the load–displacement trace. The σ and E data were expressed in MPa and GPa, respectively.

2.4 Water sorption and solubility

Ten disk-shaped samples of each resin composite (6 mm diameter \times 1 mm thickness) [11] were prepared using a metallic mold and using the same protocol described before. The samples were all placed into a desiccator containing freshly dried silica gel and calcium chloride. After 24 h, the mass of each sample was measured daily using a precision balance of 0.01 mg of readability (AUW 220D; Shimadzu Corp. Nakagyo-ku, Kyoto, Japan), until a constant mass (m_1) was obtained. Thickness and diameter of the samples were randomly measured at five points using a digital caliper to calculate their volume (V , in mm³). The samples were then immersed in distilled water at 37 °C for 7 days, removed, dried, and weighed (m_2). After weighing, the samples were dried again inside the desiccator and weighed daily to record a third constant mass (m_3), as previously described. For each sample, the WS and SL data, in $\mu\text{g}/\text{mm}^3$, were calculated using the following formulas (Eqs. 4 and 5, respectively):

$$WS = \frac{m_2 - m_3}{V} \quad (4)$$

$$SL = \frac{m_1 - m_3}{V} \quad (5)$$

2.5 Hardness

Ten disk-shaped samples of each resin composite (6 mm diameter \times 1 mm thickness) were prepared using the

foregoing metallic mold and protocol. Each sample was light-activated for 40 s on the top and bottom surfaces, polished with #600- and #1200-grit abrasive SiC papers, stored in distilled water at 37 °C for 24 h, and then submitted to the Knoop microhardness number (KMN) test using a microhardness tester (FM 700, Future Tech, Kawasaki, Japan) with 50 g of load and 15 s of dwell time. The KMN is the ratio of the load applied by the indenter to the unrecovered projected area. Five indentations were performed in each specimen, and the mean value was recorded (Fig. 1).

2.6 Surface evaluation

The same samples used for hardness testing were also evaluated by a surface profilometer (Surfcorder SE 1200, Kosaka Laboratory Ltd., Tokyo, Japan). Prior to each group analysis, the profilometer was calibrated by measuring against a reference block. The samples were rotated clockwise at random angles, and three movements of the stylus were made across the center of each sample with a cutoff value of 0.8 mm and transverse length of 4 mm. After three readings, the mean surface roughness parameter (R_a) was averaged and expressed in μm .

For 3D evaluation of the surface of each sample, atomic force microscopy (AFM) was performed in one random sample used for the surface roughness analysis. To that end, an atomic force microscope (Bruker Dimension Icon; Billerica, MA, USA) equipped with an antimony-doped silicon cantilever was used in the contact mode. The cantilever was 125 μm length and with a force constant of 42 N/m. Deflection and height-mode images were obtained simultaneously at 5 μm^2 areas, with a resolution of 512 \times 512 pixel.

2.7 Color alteration

Five disk-shaped samples (6 mm diameter \times 1 mm thickness) of each resin composite were prepared as aforementioned in Sect. 2.5. Color measurement of each sample was taken using a digital spectrophotometer (Vita Easyshade, Vita Zahnfabrik; Bad Sackingen, Germany) and the CIEL* a^*b^* color system (baseline measurement). The samples were then immersed in distilled water (pH 5.3) for 28 days. New color measurements were taken after 1 h, 1 day, 7, 14, and 28 days of water storage. The color alteration (ΔE^*) of each sample was calculated using the following formula (Eq. 6):

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (6)$$

where ΔL^* , Δa^* , and Δb^* represent the differences between the final and baseline L^* , a^* , and b^* color parameters, respectively.

2.8 Statistical analysis

All data were analyzed with the statistical program Sigma-Plot version 12 (Systat Software Inc., San Jose, CA, USA) using one-way analysis of variance (ANOVA) and Tukey's test as a post hoc test; for the color alteration, the data were analyzed with two-way repeated measure ANOVA and Student–Newman–Keuls. The level of significance was set at $\alpha=5\%$ for all analyses. The reliability of the resin composites was evaluated using the Weibull analysis, as demonstrated elsewhere [13].

3 Results

The morphology of each resin composite investigated is shown in Fig. 2, which revealed that all materials were comprised of micrometric filler particles. Among the enamel composites, Llis seems to possess smaller fillers on average, opposed by Concept Advanced and Natural Look, which seemed to be comprised of irregular and larger fillers. Llis and Fill Magic seemed to demonstrate a similar morphology to the control.

Results of degree of conversion, flexural strength, and flexural modulus are shown in Fig. 3. All resin composites produced similar degree of conversion state ($p=0.104$), ranging from 51% (Natural Look) to nearly 62% (Fill Magic) (Fig. 3a). Llis showed higher flexural strength than Concept Advanced ($p=0.003$) and Natural Look ($p=0.050$), but similar to Fill Magic and the control ($p=0.609$) (Fig. 3b). The control showed the highest modulus, although similar to Llis ($p=0.107$) (Fig. 3c). Llis showed similar flexural modulus as compared with Natural Look ($p=0.994$), which were both more rigid than Fill Magic ($p\leq 0.036$) and Concept Advanced ($p<0.001$).

Results of water sorption and solubility, hardness, and surface roughness are presented in Table 2. The control showed higher water sorption than all the enamel composites ($p\leq 0.042$). Llis and Natural Look exhibited less amount of water sorption than Concept Advanced and Fill Magic ($p<0.001$). All enamel composites demonstrated similar solubility ($p\geq 0.121$), but greater than the control ($p\leq 0.030$). The control and Llis exhibited a harder surface than the other composites ($p<0.001$). Regarding surface roughness, the control resulted in the smoothest surface of the study, which was significantly greater than all the enamel composites ($p<0.001$), which have not differed between each other ($p\geq 0.787$). As shown in Fig. 2, the AFM images confirmed the smoothest pattern of Filtek Z250 (Fig. 2e), with the least identification of grooves and valleys within the material's surface. Conversely, the AFM images for the enamel composites were considerably rougher than the control, showing depressions and protrusions indicative of an

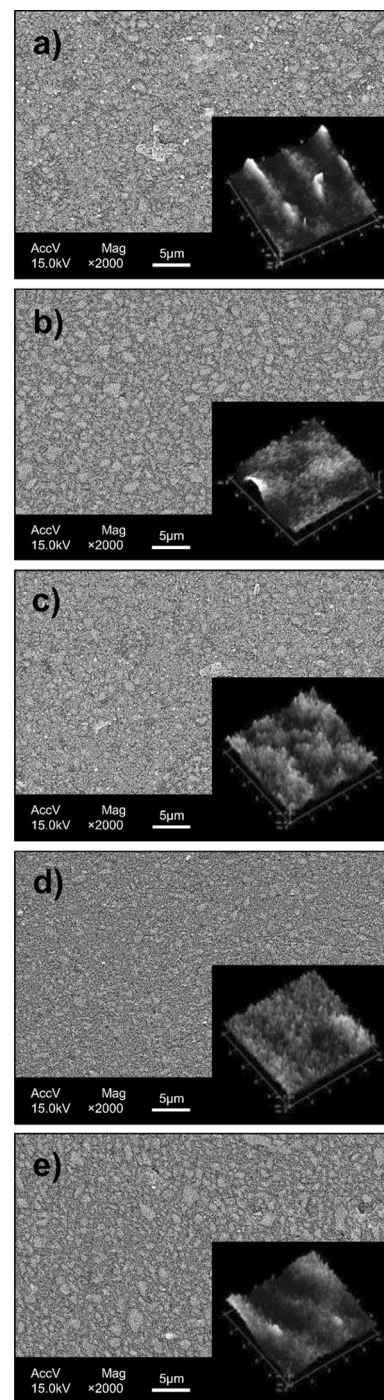


Fig. 2 Scanning electron microscopy micrographs of the resin composites evaluated: Concept Advanced (a), Fill Magic (b), Llis (c), Natural Look (d), and Filtek Z250 (e). Inset images within each SEM micrograph represent the AFM analysis for each resin composite

irregular and scratched surface (Fig. 2—images A to D), especially for Concept Advanced (image A).

Table 3 and Fig. 4 show the fracture reliability findings of this study. While Concept Advanced presented the highest Weibull modulus and reliability, Natural Look showed

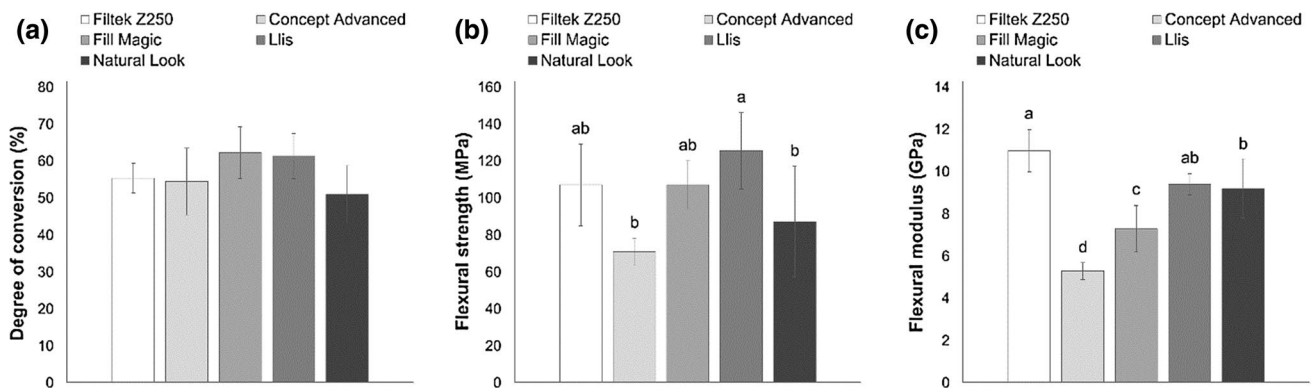


Fig. 3 Graphs showing the degree of conversion (a), flexural strength (b) and flexural modulus (c) of each resin composite tested in the study. Different letters above standard deviation bars indicate statistically significant differences among the resin composites ($p < 0.05$; ANOVA and Tukey)

Table 2 Means and standard deviation (\pm SD) of the water sorption (WS), solubility (SL), Knoop microhardness number (KMN), and surface roughness (Ra) of resin composites investigated

| Resin composites | WS ($\mu\text{g}/\text{mm}^3$) | SL ($\mu\text{g}/\text{mm}^3$) | KMN | Ra (μm) |
|------------------|----------------------------------|----------------------------------|------------------------------|----------------------------------|
| Concept Advanced | 18.6 (± 1.2) ^B | 2.8 (± 0.9) ^A | 41 (± 5) ^B | 0.28 (± 0.07) ^A |
| Fill Magic | 17.4 (± 2.2) ^B | 3.3 (± 2.0) ^A | 49 (± 8) ^B | 0.25 (± 0.07) ^A |
| Llis | 11.8 (± 1.8) ^C | 2.1 (± 1.4) ^A | 79 (± 12) ^A | 0.27 (± 0.10) ^A |
| Natural Look | 12.1 (± 1.7) ^C | 2.1 (± 0.8) ^A | 46 (± 6) ^B | 0.24 (± 0.07) ^A |
| Filtek Z250 | 22.5 (± 4.7) ^A | 0.7 (± 0.9) ^B | 92 (± 13) ^A | 0.03 (± 0.01) ^B |

Distinct letters indicate statistically significant differences among the resin composites evaluated ($p < 0.05$)

Table 3 Weibull modulus (m), characteristic strength (σ_0 , in MPa), and the correlation coefficient (r^2) with 95% confidence interval (CI), when appropriate, for the resin composites tested in the study

| Resin composites | m (95% CI) | σ_0 (95% CI) | r^2 |
|------------------|------------------------------|----------------------------------|-------|
| Concept Advanced | 11.0 (4.8–18.2) ^A | 72.5 (67.6–84.1) ^B | 0.95 |
| Fill Magic | 9.0 (4.0–14.9) ^{AB} | 103.5 (95.0–124.1) ^A | 0.78 |
| Llis | 7.0 (3.1–11.6) ^{AB} | 126.4 (113.2–159.5) ^A | 0.88 |
| Natural Look | 2.6 (1.1–4.3) ^B | 113.1 (83.8–213.4) ^{AB} | 0.96 |
| Filtek Z250 | 5.3 (2.3–8.8) ^{AB} | 113.5 (98.2–154.3) ^A | 0.96 |

Distinct letters indicate statistically significant differences among the resin composites evaluated ($p < 0.05$)

the lowest modulus of the study. The two foregoing resin composites were statistically different from each other ($p < 0.05$), and all the other composites showed similar reliability among each other. The control demonstrated an intermediary Weibull modulus, denoting its less reliable structure as compared with Concept Advanced, Fill Magic, and Llis. Llis showed the greatest characteristic strength of the study, although similar to the other composites ($p > 0.05$), except Concept Advanced ($p < 0.05$). Concerning the correlation coefficient, Concept Advanced, Natural Look, and the control displayed the highest values of the study.

Regarding the color alteration of the resin composites tested after short-term water storage, Concept Advanced

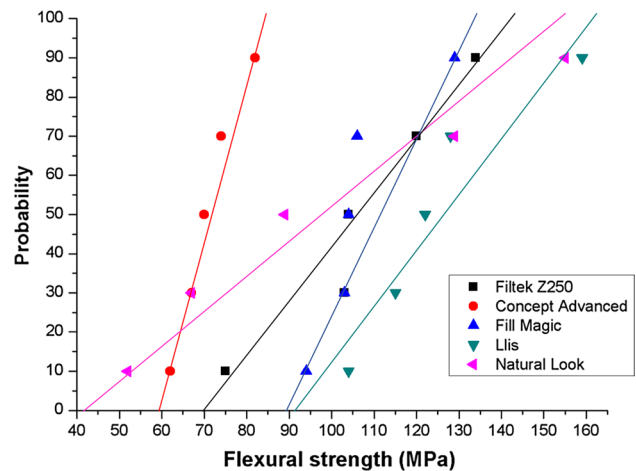


Fig. 4 Graphic representation of the Weibull analysis performed on the resin composites tested in this study. The graph shows that Concept Advanced demonstrated the most reliable structure (i.e., straightest line of the groups), opposed by Natural Look, which exhibited the least straight line results

demonstrated the greatest color change, whereas Llis showed the lowest ΔE^* values (Fig. 5). The latter resin composite was the only material that resulted in similar ΔE^* values to the threshold value (3.3) for lay people to detect a clinical color alteration, thus suggesting its greater color stability,

even after progressive water storage ($p \geq 0.112$). On the other hand, all the other composites resulted in color alteration superior to that threshold ($p < 0.05$).

4 Discussion

The major goal of this study was to compare the physico-mechanical properties and fracture reliability of enamel resin composites with that of a universal resin composite (control). According to results shown in Fig. 1 and Tables 2 and 3, the physico-mechanical performance of the enamel composites was not always similar to the control, thus rejecting the null hypothesis of study.

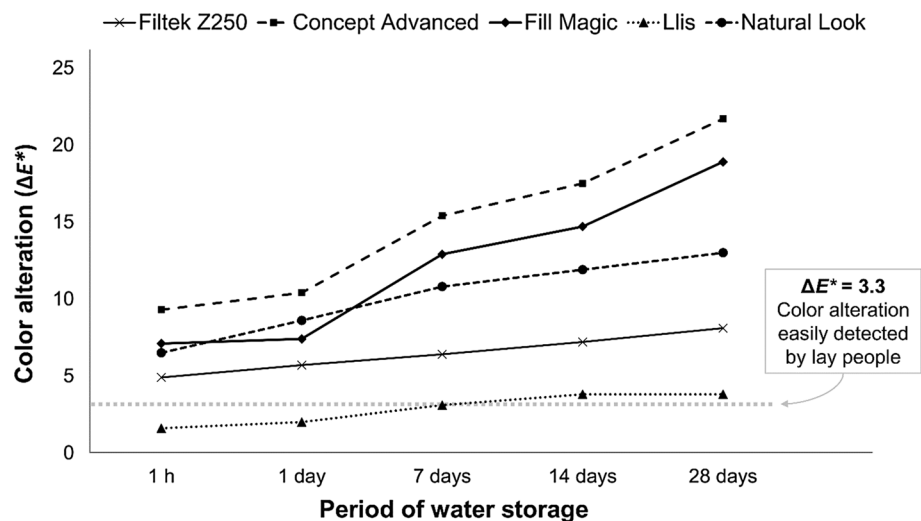
Resin composites are comprised of an organic matrix filled with inorganic fillers [4], which strongly influence on the characteristics of the material. Indeed, the type of resin monomers and the type, shape, size, and concentration of fillers have a great impact on the final properties of dental composites [5, 14]. Concerning the organic matrix of materials evaluated, all composites were comprised of bisphenol A diglycidyl dimethacrylate (Bis-GMA), as shown in Table 1, which is a very stiff monomer that contributes to the mechanical strengthening of the material. However, Bis-GMA is also very viscous, thus requiring its mixture with diluent monomers, such as triethyleneglycol dimethacrylate (TEGDMA), urethane dimethacrylate (UDMA), and bisphenol A polyethylene glycol diether dimethacrylate (Bis-EMA) [4]. One or two of the foregoing diluents are ingredients of the resin composites investigated in this study.

The degree of conversion was one of the few properties in which all composites were similar to each other (Fig. 3a), probably due to the presence of Bis-GMA in all the materials. Even with the presence of different diluents within the polymeric systems, no significant differences were observed, suggesting that the present resin composites showed little

variation about their organic and inorganic composition, which may account to their similar polymerization kinetics. This belief may be applicable only to resin composites since they are highly filled structures with inorganic particles of self-limiting nature. In fact, highly filled composites are usually associated with reduced mobility of residual monomers, affecting polymer chain formation [15]. Notably, all enamel composites evaluated may be effectively used for restorative purposes, since a similar state of cure was obtained when compared with the control. Moreover, it is known that adequate polymerization of resin composites offers optimal mechanical and physical characteristics to the restoration, as well as biological safety for the restorative treatment [16], so that the degree of conversion is an important property of dental resin-based materials.

Similarly to the degree of conversion analysis, the enamel composites have also presented similar flexural strength compared with the control (Fig. 3b); concerning modulus, the enamel materials showed inferiority as compared with the control, except for Llis (Fig. 3c). Indeed, Llis was the only composite that presented an overall similar mechanical behavior to the control. Considering that mechanical strength depends mainly on the inorganic nature of the material, factors such as the type, shape, size, and concentration of fillers may have played a significant role on the mechanical behavior of composites [5, 12, 14]. According to Table 1, all resin composites possess similar inorganic composition, varying with regard to the presence of barium, aluminum, or zirconium glass fillers, as well as silica nanoparticles. Only the inorganic nature of Fill Magic was totally unknown (not supplied by the manufacturer). Regarding the content of fillers, the exact concentration is also unknown within the enamel composites, thus limiting the use of this factor to support the results obtained. Nevertheless, as shown in Fig. 2, Llis showed a uniform morphology, presenting regular and smaller filler particles. This may have contributed

Fig. 5 Graph showing the color alteration results of the study for each resin composite tested after different periods of water storage. The gray dotted line represents the color alteration easily detected by lay people ($\Delta E^* = 3.3$), indicating the threshold for clinical perceptibility of color alteration in dentistry



to its satisfactory mechanical behavior, which was similar to the control. On the other hand, Concept Advanced, Fill Magic, and Natural Look seemed to be comprised of fillers of irregular shape and size situated within a wider range, which may have allowed stress concentration in the bulk of the materials and, consequently, reduced mechanical properties [5, 17]. Furthermore, the presence of only Bis-GMA and UDMA within Concept Advanced has probably resulted in low flexural strength and modulus characteristics, since UDMA is recognized by its low rigidity [18], originating greater deformation of the material when applied to a given force. Consequently, fatigue may occur at the tooth–restoration interface [19], affecting formation of a strong and cross-linked polymer network.

It is also important to note that for resin composites, the minimum requirement for a clinically acceptable flexural strength is 80 MPa [20]; not less important, for universal dental composites (i.e., hybrid, microhybrid), satisfactory flexural modulus usually ranges from 8.8 to 13 GPa. Taking the threshold values presented, only Llis and Natural Look achieved both requirements, and similarly to the control. These findings are important because they show that Concept Advanced and Fill Magic are not sufficiently strong to be applied as restorative materials in posterior teeth, since aggressive stress derived from mastication and oral function would fatigue the material, allowing crack propagation and, perhaps, restoration fracture to occur [21]. Concept Advanced and Fill Magic could be more interestingly applied in the anterior dentition only.

Resin composites are usually placed into tooth cavities, and considering that they will be in direct contact with the wet environment present in the mouth, water sorption and solubility tests were also performed in order to reveal the physicochemical stability of the material against hygroscopic and hydrolytic phenomena of degradation [22]. According to results shown in Table 2, Llis and Natural Look presented lower water sorption than Concept Advanced and Fill Magic. In addition, all enamel composites presented lower water sorption when compared with the control, but greater solubility. Two main factors are usually associated with the water sorption and solubility properties: “hydrophilicity” and the “network cross-link nature” of the material [22]. Resin-based materials constituted of polar groups (e.g., hydroxyl, ester, urethane, and ether linkages) are more hydrophilic than materials containing nonpolar groups. According to Venz et al. [23], typical monomers used to prepare dental resin composites may follow the subsequent order of hydrophilicity: TEGDMA > Bis-GMA > UDMA. Considering that Llis and Natural Look were the only TEGDMA-based composites (Table 1), they were supposed to undergo greater water sorption than the other materials [24]. Nevertheless, the latter composites surprisingly demonstrated the lowest water sorption of the study, which may be explained by the

positive effect that TEGDMA has on the cross-linking of the system [25], enhancing the material’s resistance to hydrolysis. On the other hand, the other resin composites were mostly UDMA based, fact that may have reduced the cross-linking of the polymer network, thus increasing water sorption and solubility [11, 22]. It is worth to mention that all composites met the requirements stated by ISO 4049:2009 [20], resulting in water sorption and solubility values lower than $25 \mu\text{g}/\text{mm}^3$ and $7.5 \mu\text{g}/\text{mm}^3$, respectively.

Enamel resin composites are commonly used as the final layer/increment of the restoration, thus denoting that they may receive the first and direct impacts originated from the mastication process. Consequently, they are more susceptible to surface wear when compared with composites used to restore dentin [26, 27]. Hardness has been thoroughly used to predict the wear of resin composites, assuming that the harder the surface, the lower the wear showed by the material, and vice versa [28, 29]. Considering that Llis was the only enamel composite as hard as the control (Table 2), it would probably resist more wear than the other composites. On the other hand, Concept Advanced, Fill Magic, and Natural Look presented, respectively, ~48, 38, and ~42% less hardness than Llis, and their softer nature would probably contribute to a worse wear resistance behavior. Hardness is mainly influenced by the organic nature and the filler content of the material [14]. Taking into consideration that Llis demonstrated greater flexural strength, modulus, and hardness than the other enamel composites, it can be inferred that the latter are constituted of less amount of fillers [30], although this information is unknown.

The superficial characteristic of each resin composite was assessed by surface roughness and AFM analyses. The real purpose of analyzing the roughness and topographical morphology of composites was not to evaluate their polishing state, but the easiness to get the material finished and polished. Therefore, samples were polished with #600- and #1200-grit abrasive SiC papers for 1 min each, followed by surface analysis. According to results shown in Table 2 and Fig. 2 [inset images within each scanning electron microscopy (SEM) micrograph], all enamel composites presented rougher surface when compared to the control, ranging from 8- to 9.3-fold increase in roughness. This result was confirmed by the AFM images, which showed that the control acquired a smoother surface as compared to the enamel composites. At a first glance, it may be suggested that the enamel composites were more difficult to polish. However, this finding should be interpreted with caution since Filtek Z250 also showed the hardest surface of the study (Table 2), which was consequently expected to be the most difficult material to get finished and polished [12]. In this way, and considering that the enamel composites were generally softer than the control, the organic matrix of the former was probably easily removed, resulting in peaks and valleys along their

surface due to smear formation, which led to the increased roughness values obtained [31, 32]. It is important to note that surface roughness analysis is not the most appropriate method to evaluate the topography of resin composites, differently from the AFM analysis, which possesses resolution far exceeding that of other stylus and optical based methods, giving information on the 2D as well as 3D nanoscale [32]. Notably, our AFM findings confirmed the surface roughness data obtained. From the clinical point of view, an interesting fact could be drawn: Over-polishing a moderately soft resin composite may increase the material's surface roughness. So that, the clinical finishing and polishing of the enamel resin composites tested here should be performed with caution and for shorter periods of time, as well as without using very abrasive products.

The Weibull statistical analysis was carried out to describe the structural homogeneity of the resin composites tested [13]. The Weibull modulus is usually used as a measure of the distribution of strengths, expressing the reliability of the material [33]. The most reliable composite tested in this study was Concept Advanced (Table 3; Fig. 4—observe the straightest line originated for Concept Advanced in the graph), which was different from Natural Look. This finding contradicts the mechanical properties presented by these two foregoing composites, in which Natural Look was stronger than Concept Advanced. One possible explanation for this duality concerns to the standard deviation values of the flexural strength data of the groups (i.e., the data used to calculate the Weibull analyses). Indeed, the Weibull analysis is an effective way to describe the resistance variation of the material [33], and considering that Concept Advanced showed a significant smaller variation of data ($SD = 7.5$) as compared with Natural Look ($SD = 29.8$), the former would also present a more reliable structure, which means its structure possesses less likelihood to fracture at small loads when compared to the latter's structure. Concerning the characteristic strength of the composites, Llis showed the strongest behavior, followed by the control and Natural Look, and, lastly, Fill Magic and Concept Advanced. The characteristic strength corresponds to the strength at a failure probability of approximately 63% [33], so that the former resin composites are definitely stronger than the latter ones, supporting more loads and stresses during function. The greatest characteristic strength of Llis may be also associated with a more uniform and homogeneous structure, perhaps free of voids and defects, which directly influences on the overall mechanical behavior of the material [34].

Lastly, the physical stability of the resin composites was also evaluated by inducing hygroscopic and hydrolytic degradation of the materials after water storage for 28 days, which was assessed by the color analysis. According to Fig. 5, Llis exhibited the greatest color stability, which may be related to its lower water sorption uptake as discussed

before. Natural Look and the control also resulted in less color alteration as compared to Concept Advanced and Fill Magic, once again due to their more chemically stable composition. Color alteration may occur even after immersion in distilled water, since degradation of the cross-links of resin-based materials due to hydrolysis may modify the transmission of light within the resin composite, thereby affecting its color perception [11, 22]. Nevertheless, the strongest and the more stable the material's structure, as demonstrated by some composites tested (e.g., Llis, Natural Look, and Filtek Z250), the lower the color alteration overtime. The color stability of the resin composites tested here could be more affected by using staining solutions (e.g., coffee, wine, soda, tea) rather than distilled water, since the former may be comprised of alcoholic and acidic composition, which directly enhances hydrolysis of resin-based dental restoratives [35, 36]. Further studies should investigate the color alteration profile of different resin composites after exposure to staining solutions, in order to verify their physical stability overtime and, consequently, the durability of dental restorative procedures.

5 Conclusions

In summary, enamel resin composites can be as strong as composites indicated to restore dentin, depending on the material selected. It seems that the more uniform the apparent morphology of fillers in the matrix, the greater the mechanical performance of the material. Conversely, some of the enamel composites tested may present insufficient strength and physical stability when compared with the universal control composite. Taken together, the present findings confirmed the performance of four different resin composites indicated to restore dental enamel, showing that two of them (Llis and Natural Look) were sufficiently strong to be placed in both posterior and anterior dentition; whereas the other two resin composites (Concept Advanced and Fill Magic) could be more satisfactorily indicated to the restoration of the anterior dentition only.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

References

- Borgia E, Baron R, Borgia JL (2019) Quality and survival of direct light-activated composite resin restorations in posterior teeth: a 5- to 20-year retrospective longitudinal study. *J Prosthodont* 28:e195–e203

2. Vural UK, Kiremitci A, Gokalp S (2019) Clinical performance and epidemiologic aspects of fractured anterior teeth restored with a composite resin: a two-year clinical study. *J Prosthodont* 28:e204–e209
3. Salerno M, Diaspro A (2015) Dentistry on the bridge to nanoscience and nanotechnology. *Front Mater* 2:1–10
4. Ferracane JL (2011) Resin composite—state of the art. *Dent Mater* 27:29–38
5. Ferracane JL, Pfeifer CS, Hilton TJ (2014) Microstructural features of current resin composite materials. *Curr Oral Health Rep* 1:205–212
6. Tanthanuch S, Kukiattrakoon B, Peerasukprasert T (2016) The effect of red and white wine on color changes of nanofilled and nanohybrid resin composites. *Restor Dent Endod* 41:130–136
7. Villarreal M, Fahl N, De Sousa AM, De Oliveira OB Jr (2011) Direct esthetic restorations based on translucency and opacity of composite resins. *J Esthet Restor Dent* 23:73–87
8. Habib E, Wang R, Wang Y, Zhu M, Zhu XX (2016) Inorganic fillers for dental resin composites: present and future. *ACS Biomater Sci Eng* 2:1–11
9. Hassan AM, Nabih SM, Mossa HM, Baroudi K (2015) The effect of three polishing systems on surface roughness of flowable, microhybrid, and packable resin composites. *J Int Soc Prev Community Dent* 5:242–247
10. Koottathape N, Takahashi H, Iwasaki N, Kanehira M, Finger WJ (2014) Quantitative wear and wear damage analysis of composite resins in vitro. *J Mech Behav Biomed Mater* 29:508–516
11. Munchow EA, Zanchi CH, Ogliari FA, Silva MG, de Oliveira IR, Piva E (2014) Replacing HEMA with alternative dimethacrylates in dental adhesive systems: evaluation of polymerization kinetics and physicochemical properties. *J Adhes Dent* 16:221–228
12. Valente LL, Peralta SL, Ogliari FA, Cavalcante LM, Moraes RR (2013) Comparative evaluation of dental resin composites based on micron- and submicron-sized monomodal glass filler particles. *Dent Mater* 29:1182–1187
13. Quinn JB, Quinn GD (2010) A practical and systematic review of Weibull statistics for reporting strengths of dental materials. *Dent Mater* 26:135–147
14. Beun S, Glorieux T, Devaux J, Vreven J, Leloup G (2007) Characterization of nanofilled compared to universal and microfilled composites. *Dent Mater* 23:51–59
15. Fugolin AP, Correr-Sobrinho L, Correr AB, Sinhoreti MA, Guiraldo RD, Consani S (2016) Influence of irradiance on Knoop hardness, degree of conversion, and polymerization shrinkage of nanofilled and microhybrid composite resins. *Gen Dent* 64:26–31
16. Gajewski VE, Pfeifer CS, Froes-Salgado NR, Boaro LC, Braga RR (2012) Monomers used in resin composites: degree of conversion, mechanical properties and water sorption/solubility. *Braz Dent J* 23:508–514
17. Dadrasi A, Fooladpankeh S, Alavi Gharahbagh A (2019) Interactions between HA/GO/epoxy resin nanocomposites: optimization, modeling and mechanical performance using central composite design and genetic algorithm. *J Braz Soc Mech Sci Eng* 41:63
18. Barszczewska-Rybarek IM (2009) Structure-property relationships in dimethacrylate networks based on Bis-GMA, UDMA and TEGDMA. *Dent Mater* 25:1082–1089
19. Xavier AM, Sunny SM, Rai K, Hegde AM (2016) Repeated exposure of acidic beverages on esthetic restorative materials: an in vitro surface microhardness study. *J Clin Exp Dent* 8:e312–e317
20. ISO 4049 (2009) Dentistry—polymer-based filling, restorative and luting materials
21. Bicalho AA, Tantbirojn D, Versluis A, Soares CJ (2014) Effect of occlusal loading and mechanical properties of resin composite on stress generated in posterior restorations. *Am J Dent* 27:129–133
22. Ferracane JL (2006) Hygroscopic and hydrolytic effects in dental polymer networks. *Dent Mater* 22:211–222
23. Venz S, Dickens B (1991) NIR-spectroscopic investigation of water sorption characteristics of dental resins and composites. *J Biomed Mater Res* 25:1231–1248
24. Liu X, Wang Z, Zhao C, Bu W, Zhang Y, Na H (2018) Synthesis, characterization and evaluation of a fluorinated resin monomer with low water sorption. *J Mech Behav Biomed Mater* 77:446–454
25. Sideridou I, Tserki V, Papanastasiou G (2002) Effect of chemical structure on degree of conversion in light-cured dimethacrylate-based dental resins. *Biomaterials* 23:1819–1829
26. Kramer N, Garcia-Godoy F, Reinelt C, Feilzer AJ, Frankenberger R (2011) Nanohybrid vs. fine hybrid composite in extended Class II cavities after six years. *Dent Mater* 27:455–464
27. Antunes PV, Ramalho AL (2018) Influence of abrasive solutions on direct restorative composites wear. *J Braz Soc Mech Sci Eng* 40:20
28. Faria AC, Benassi UM, Rodrigues RC, Ribeiro RF, Mattos Mda G (2007) Analysis of the relationship between the surface hardness and wear resistance of indirect composites used as veneer materials. *Braz Dent J* 18:60–64
29. Ramalho A, Antunes PV (2005) Reciprocating wear test of dental composites: effect on the antagonist. *Wear* 259:1005–1011
30. Wang T, Tsoi JK, Matinlinna JP (2016) A novel zirconia fibre-reinforced resin composite for dental use. *J Mech Behav Biomed Mater* 53:151–160
31. Munchow EA, Correa MB, Ogliari FA, Piva E, Zanchi CH (2012) Correlation between surface roughness and microhardness of experimental composites with varying filler concentration. *J Contemp Dent Pract* 13:299–304
32. Salerno M, Patra N, Thorat S, Derchi G, Diaspro A (2012) Combined effect of polishing on surface morphology and elastic properties of a commercial dental restorative resin composite. *Sci Adv Mater* 4:126–134
33. Prado RD, Pereira GKR, Bottino MA, Melo RM, Valandro LF (2017) Effect of ceramic thickness, grinding, and aging on the mechanical behavior of a polycrystalline zirconia. *Braz Oral Res* 31:e82
34. Warangkulkasemkit S, Pumpaluk P (2019) Comparison of physical properties of three commercial composite core build-up materials. *Dent Mater J* 38:177–181
35. DA Silva MA, Vitti RP, Sinhoreti MA, Consani RL, Silva-Júnior JG, Tonholo J (2016) Effect of alcoholic beverages on surface roughness and microhardness of dental composites. *Dent Mater J* 35:621–626
36. Munchow EA, Ferreira AC, Machado RM, Ramos TS, Rodrigues-Junior SA, Zanchi CH (2014) Effect of acidic solutions on the surface degradation of a micro-hybrid composite resin. *Braz Dent J* 25:321–326

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.