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Laboratory Research

Characterization of Contemporary Conventional, Bulk-fill, and Self-adhesive Resin Composite Materials

C David • CE Cuevas-Suárez • GC de Cardoso • CP Isolan
RR de Moraes • WLO da Rosa •EA Münchow • AF da Silva

Clinical Relevance

Bulk-fill and self-adhesive flowable resin composites are restorative materials with wide application potential in dental practice. Therefore, it is essential to know their properties, aiding in the careful decision by the dental practitioner to meet the requirements of each clinical case.

SUMMARY

Objective: To evaluate the physical and biological properties of different types of flowable resin composites and their bonding ability to dentin, comparing the performance of self-adhesive and bulk-fill materials with a conventional control.

Methods and Materials: Four flowable resin composites were tested: two self-adhesive (Y-flow [SA_YF]; and Dyad Flow [SA_DF]); one bulk-fill (Filtek Bulk Fill Flow [BF]); and one conventional composite (Opallis Flow [OF]). The microshear bond strength (μSBS) to dentin (bovine samples)

Carla David, Biopathological Research Group, Faculty of Dentistry (GIBFO), University of the Andes Mérida, Mda, Venezuela; Graduate Program in Dentistry, School of Dentistry, Federal University of Pelotas, Pelotas, RS, Brazil

Carlos Enrique Cuevas-Suárez, Dental Materials Laboratory, Academic Area of Dentistry, Autonomous University of Hidalgo State, Pachuca, Hgo, Mexico

Gabriela Cardoso de Cardoso, Graduate Program in Dentistry, School of Dentistry, Federal University of Pelotas, Pelotas, RS, Brazil

Cristina Pereira Isolan, Graduate Program in Dentistry, School of Dentistry, Federal University of Pelotas, Pelotas, RS, Brazil

Rafael Ratto de Moraes, Graduate Program in Dentistry, School of Dentistry, Federal University of Pelotas, Pelotas, RS, Brazil

Wellington Luiz Oliveira da Rosa, Graduate Program in Dentistry, School of Dentistry, Federal University of Pelotas, Pelotas, RS, Brazil

Eliseu A Münchow, Department of Conservative Dentistry, School of Dentistry, Federal University of Rio Grande do Sul, POA, RS, Brazil

*Adriana Fernandes da Silva, Graduate Program in Dentistry, School of Dentistry, Federal University of Pelotas, Pelotas, RS, Brazil

*Corresponding author: Gonçalves Chaves St, 457/503, Centro 96015-560, Pelotas, Brazil; e-mail: adrisilvapiva@gmail.com

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was investigated at 24 hours and 6 months of storage. The materials were also characterized by degree of conversion, cross-link density, water contact angle, color stability, and cell viability (ISO 10993-5/2009) analyses. Data were analyzed using Analysis of Variance and Tukey tests (α=0.05).

Results: The μSBS values were higher for control specimens at 24 hours, whereas the resin-dentin bonds were similarly distributed among the groups after aging. Adhesive failure was the most frequent pattern observed at both time intervals. SA_YF was the only material that increased the bond strength over time. Degree of conversion increased in the following order: SA_YF (28.6±1.4%) < BF (49.7±0.8%) < OF (60.0±2.0%) = SA_DF (63.6±2.3%). Cross-link density was similar among all materials. The self-adhesive composites were more hydrophilic than the other types, with BF showing the lowest water contact angle and the greatest color alteration. All resin composites had a biocompatible behavior.

Conclusion: Chemical composition appeared to be an influential factor affecting the physico-mechanical and biological behavior of the materials tested.

INTRODUCTION

Dental resin composites are tooth-colored restorative materials with broad clinical applicability in dentistry. Despite their overall similar chemical composition (ie, a methacrylate-based organic matrix filled with silica and glass particles of varying sizes), resin composites may differ with regard to their viscous state, ranging from highly viscous materials (eg, packable composites) to those with moderate viscosity (regular composites) or low viscosity (flowable composites).[1,2] Regular resin composites have notably been the most traditional materials used in daily practice over the last few decades, while the clinical application of flowable materials has gained popularity in modern dentistry due to their versatility and applicability in several procedures. For instance, flowable resin composites are indicated as lining materials in deep cavities,[3] as restorative materials in deciduous teeth[4] or Class I/Class V cavities of permanent teeth,[5] as sealers of pit and fissure surfaces,[6] and as the material for repairing defective resin composite[7,8] or amalgam[9] restorations.

At present, the development of flowable resin composites has shifted from manufacturing traditional materials that require prior application of adhesive systems to materials with self-adhesive properties (self-adhesive composites) or expanded polymerization depth (bulk-fill composites).[10,11] Relative to self-adhesive composites, they share the chemistry of self-etching bonding agents due to the presence of functional acidic monomers, allowing the material to create its own pathway toward the hybridization of dental substrates, without any surface pre-treatment with adhesives.[12] Equally important are the bulk-fill composites that are also associated with a simpler and less technique-sensitive restorative protocol since they can be applied in the tooth cavity with the use of thicker resin increments (eg, 4-5-mm thick) when compared with the thicknesses required when using conventional composites (<2 mm).

Although there is widespread clinical application of flowable self-adhesive/bulk-fill resin composites in modern dentistry, there is still unclear evidence regarding their overall performance. Characterization and comparative analyses of these materials could help clinicians to understand their laboratory and clinical behavior. Hence, this study aimed to evaluate the physical and biological properties of different types of flowable resin composites, and their bonding ability to dentin, with the aim of comparing the performance of self-adhesive and bulk-fill materials with that of a conventional control. The null hypothesis was that the resin composites would not differ among them, irrespective of their classification.

METHODS AND MATERIALS

Materials

Four flowable resin composites were tested in this study: two self-adhesive materials, namely Y flow SA (Yller, Pelotas, RS, Brazil, SA_YF) and Dyad Flow (Kerr Orange,CA, USA, SA_DF); one bulk-fill material (Filtek Bulk Fill Flow, 3M ESPE, St Paul, MN, USA, BF); and one conventional material (Opallis Flow, FGM, Joinville SC, Brazil, OF), serving as the control. Table 1 provides information on the manufacturer, chemical composition, color, batch number, viscous category, polymerization depth, and polymerization protocol for each resin composite according to the manufacturers' directions of use.

Study Design and Sample Size Calculation

This research followed the CRIS Guidelines for *in vitro* studies.[13] A lab assistant did the randomization and allocation of the specimens and groups. A trained operator conducted all experiments in the laboratory. The analysis and interpretation of the data were carried out by two blinded researchers who had no contact with the experimental analyses. The response variables tested in this *in vitro* study were as follows:

Table 1: *Specifications of the Resin Composites and Universal Adhesive Tested in the Study*

Material (Manufacturer)	Chemical Composition	Color	Batch Number	Flow Category	Polymerization	
					Depth	Time
Yflow SA (Yller, Pelotas, RS, Brazil)	Inorganic fillers, acid monomers (MDP and GPDM), methacrylate monomers, pigments, initiators and stabilizers	A2	3193	Low	2 mm	40 s
Dyad Flow (Kerr Orange, CA, USA)	GPDM, prepolymerized particles, barium glass fillers, colloidal silica nanoparticles, nano-sized ytterbium trifluoride fillers	A2	4910830	Low	2 mm	40 s
Filtek Bulk Fill Flow (3M ESPE, St. Paul, MN, USA)	Bis-GMA, UDMA, Bis-EMA 6, Procrylat, zirconia and silica particles, ytterbium trifluoride	U	N976617	Low	4 mm	20 s
Opallis Flow (FGM Joinville, SC, Brazil)	Silanized inorganic fillers (barium-aluminum silicate and silica nanoparticulate dioxide), methacrylic monomers, TEGDMA, Bis-EMA, Bis-GMA, camphorquinone, co-initiators, stabilizers and pigments	A2	101018	Average	2 mm	40 s
Single Bond Universal (3M ESPE, St. Paul, MN, USA)	MDP, dimethacrylate resins, HEMA, polyalkenoic acid copolymer, fillers, ethanol, water, initiators, silane	—	90424A	—	—	20 s

Abbreviations: 10-MDP, 10-methacryloxydecyl dihydrogen phosphate; GPDM, glycerol phosphate dimethacrylate; Bis-GMA, bisphenol A glycidyl methacrylate; UDMA, urethane dimethacrylate; Bis-EMA, ethoxylated bisphenol A glycidyl methacrylate; TEGDMA, tryethyleneglycol dimethacrylate; HEMA, 2-hydroxyethyl methacrylate.

bond strength to dentin (n=20) and failure mode after bond strength testing (n=20), degree of conversion of C=C bonds (n=3), crosslink density (n=3), water contact angle (n=3), color stability (n=3), and cell viability (n=6). The primary response variable was the microshear bond strength, and the sample size calculation was based on a comparative study design of 4 independent groups, an average difference in shear bond strength of 11.5 MPa, an average standard deviation of 6.5, $\alpha = 0.05$, and test power of 0.8.[14]

Bond Strength to Dentin

Eighty bovine incisors were used, after all blood and soft tissue were removed, and thereafter stored in 0.5% chloramine/water solution at 4°C for 1 week. Then, the teeth were stored in distilled water at 4°C for no longer than two months until use. The roots of the teeth were sectioned at the cement-enamel junction, and the middle dentin was exposed by using an orthodontic grinder. The teeth were then fixed in cylindrical plastic molds with self-curing acrylic resin and the buccal surface of each tooth was ground with 180-grit SiC abrasive paper to obtain flat dentin surfaces. Subsequently, the specimens were polished with 600-grit SiC abrasive papers for 60 seconds to provide a uniform and standardized smear layer, and they were randomly allocated into four groups according to the restorative material (n=20): SA_YF, SA_DF, BF, and OF.

The resin composites were applied by using elastomeric molds with two cylindrical orifices of 1.2 mm in diameter each. In groups BF and OF, the materials were placed only after the application of a universal bonding agent (Single Bond Universal, SBU, 3M ESPE) according to the manufacturer's instructions. The bonding agent was vigorously rubbed on the dentin surface using a microbrush for 10 seconds, then the mold was placed on the adhesive layer; the bonding agent was photo-activated with a light-emitting diode (LED) curing unit (Radii Cal, SDI, Bayswater, VIC, Australia) through the orifices of the mold for 20 seconds. Next, the resin composites BF and OF were applied in the respective orifices and photo-activated with the LED for the recommended time (Table 1). In groups SA_YF and SA_DF, no bonding agent was applied so that each resin composite was applied as follows: the dentin surface was dried with absorbent paper, the elastomeric mold was placed in position, each material was inserted into the respective orifice, and photo-activation of materials was performed with the LED for the recommended time (Table 1).

All the restored samples were allocated into two subgroups, according to the period of storage in distilled water at 37°C: 24 hours (immediate testing) or 6 months (long-term testing) (n=20). After each time interval, the microshear bond strength (μSBS) to dentin of each restored specimen was evaluated in a universal testing machine (DL500, EMIC, São José dos Pinhais, PR, Brazil) at a crosshead speed of 1 mm/min. To that end, the samples were mounted on the testing machine with the resin-tooth interface placed parallel to the stainless-steel wire 0.2 mm in diameter. A shear force was applied until failure. The μSBS values were calculated in MPa and considering the restoration area. Fractured specimens were observed at 40× magnification under a stereomicroscope to determine the failure mode as cohesive, adhesive, or mixed failures.

Degree of Conversion (DC)

The DC of the resin composites (n=3) was assessed using a Fourier transform infrared spectrometer (FTIR Spectrometer Prestige 21, Shimadzu, Japan) equipped with an attenuated total reflectance (ATR) device. Standardized quantities of each material were dispensed on the ATR crystal. Infrared spectra were obtained before and after photo-activation of the materials with the LED described earlier, which had an irradiance of 1000 mW/cm^2. The DC was determined by observing the ratio of the absorbance intensity of the aliphatic C=O (peak height at 1638 cm^{-1}) to the absorbance intensity of the aromatic C=C (1608 cm^{-1}) used as an internal standard.[15]

Crosslink Density

Crosslinking was indirectly measured by the percentage change in microhardness of the materials before and after storing the specimens in 100% ethanol for 24 hours (n=3). Briefly, three specimens of each resin composite (5 mm diameter × 2 mm thickness) were prepared using an elastomeric mold. The top and bottom surfaces of each specimen were photo-activated with the LED for 10 seconds. The initial (dry VHN) and final (wet VHN) Vickers Hardness Numbers of each specimen were obtained by using a microindenter (FM 700; Future-Tech Corp, Japan) with a Vickers indenter, under a load of 200 g for 15 seconds. Three indentations were performed for each specimen and the results were averaged. The ratio between ethanol-wet VHN and dry VHN (%) was used as an indication of crosslinking percentage.[16]

Water Contact Angle

The water contact angle formed on the surface of the resin composites (n=3) was measured using an Optical Tensiometer (Theta Lite TL101, Biolin Scientific Inc, Finland) with a sessile drop method. Standardized drops of distilled water (5 μL) were dispensed directly onto the surface of specimens that were made of each resin composite. The specimens were prepared as demonstrated earlier. Immediately after dispensing the drop, a dynamic reading in real time was taken of the right and left contact angles formed with the material surface. The One Attension software (Biolin Scientific Inc) was used at 20 frames per second for 20 seconds. The contact angle (°) was recorded as the mean between the right and left readings (n=3).

Color Stability (ΔE_{00})

Three cylindrical specimens (5 mm diameter × 2 mm thickness) were prepared as previously mentioned. CIELab color parameters were measured with a VITA Easyshade Spectrophotometer (Zahnfabrik, BadSäckingen, Germany) against white ($L^*=93.1$, $a^*=1.3$, $b^*=5.3$) and black ($L^*=27.9$, $a^*=0$, $b^*=0$) backgrounds. The measurements were made immediately after polymerization (first reading) and after storing the specimens in distilled water at 37°C for 6 months (second reading). The color alteration (ΔE_{00}) of the resin composites after storage was calculated based on the CIEDE2000 equation:[17]

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

where, ΔL', ΔC', and ΔH' are the differences in brightness, chroma, and hue between two sets of color coordinates; RT is the rotation function that explains the interaction between differences in chroma and hue in the blue region; SL, SC, and SH indicate the weighting functions used to adjust the total color difference to variation in perceived magnitude, with variation in the location of difference in the color coordinate between two color readings; and KL, KC, and KH represented the correction terms for the experimental condition. The perceptibility threshold was established at $\Delta E_{00} = 0.8$, whereas the acceptability threshold was set at $\Delta E_{00} = 1.8$.[18]

Cell Viability

The cytotoxicity test was performed according to ISO 10993-5: 2009,[19] using mice fibroblast cells (L929) cultured at a density of 2×10^4 cells in 96-well plates containing DMEM (Eagle's Medium Modified by Dulbecco) supplemented with 10% L-glutamine, 10% fetal bovine serum (FBS), penicillin (100 U/mL), and streptomycin (100 U/mL). The cells were incubated at 37°C in 95% air and 5% CO_2 for 24 hours, and the cell viability ratio was evaluated by the MTT assay. Cylindrical shaped specimens were prepared (5×1 mm, n=3). The specimens were placed in 24-well plates with 1 mL DMEM and stored at 37°C and pH 7.2 under static conditions. After 24 or 48 hours, 200 μL of each specimen was transferred to the 96-well plate containing the precultured cells, and the plate was incubated for another 24 hours. As a control, a group containing only fibroblast cells in DMEM was used. After incubation, DMEM was removed and an MTT solution was placed in each well. After 4 hours of incubation at 37°C in a condition of darkness, the blue formazan precipitate was extracted from the mitochondria using 200 μL/well of dimethyl sulfoxide (DMSO) on a shaker at 150 rpm for 5 minutes. The absorption was determined using a spectrophotometer at a wavelength of 540 nm.

Statistical Analysis

For statistical analysis, the statistical method was based on adherence to the normal distribution model and equality of variances. For the microshear bond strength, a one-way analysis of variance (ANOVA) was used to detect significant differences between the materials for each storage period. The effect of aging condition was analyzed using a two-sample Student's-t test. For the properties of crosslink density, degree of conversion, and color stability, one-way ANOVA was used. Two-way ANOVA analysis was used to evaluate the effect of the material and the incubation time on cell viability. All the multiple comparisons were performed using the Tukey *post-hoc* test. For all tests, $p<0.05$ was considered statistically significant. The statistical analyses were carried out using SigmaPlot 12 software (Systat Inc, San Jose, CA, USA).

RESULTS

The results for the bond strength to dentin and fracture pattern of the resin-dentin bonds are shown in Figure 1. At immediate testing (24 hours of water storage), the control group (OF) demonstrated higher bonding performance than the other groups ($p \leq 0.003$), which did not differ among them ($p \geq 0.214$). In the long-term test (6 months of water storage), all groups performed in a similar manner to each other ($p \geq 0.181$). When the immediate μSBS values of the same resin composite group were compared to the long-term values, the resin-dentin bonds were stable over time for SA_DF, BF, and OF, whereas for SA_YF the μSBS values were statistically increased after 6 months of water storage ($p=0.03$). Regarding the failure pattern obtained for each group in the two storage time intervals, all groups demonstrated a predominance of adhesive failures, with the control (OF) showing nearly 20% of mixed fracture and the BF and SA_DF groups exhibiting approximately 30% of pre-test failures after 6 months of water storage (Figure 1B).

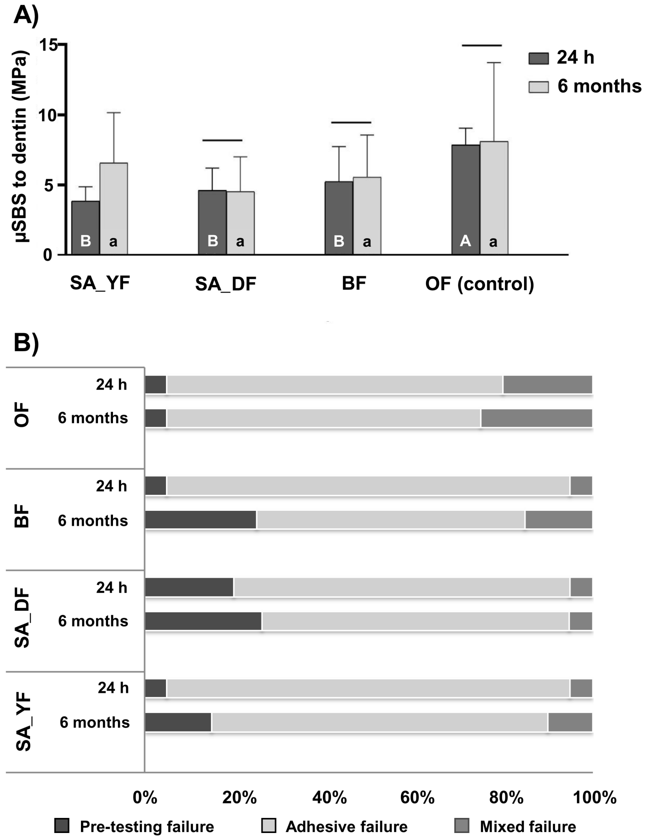

Figure 1. *(A): Graphs showing microshear bond strength to dentin. The same uppercase and lowercase letters indicate no statistically significant differences between resin composites at 24 hours and 6-month time intervals, respectively* ($p<0.05$)*. Bars under the same horizontal line indicate statistically similar bond strength values when immediate and long-term time intervals were compared* ($p>0.05$)*. (B): Failure mode of resin composites tested at immediate (24 hours) or long-term (6 months) time intervals. Abbreviations: SA_YF, Y-flow Self-Adhesive; SA_DF, Dyad Flow Self-Adhesive; BF, Filtek Bulk Fill Flow; OF, Opallis Flow.*

Table 2 shows the results for the degree of conversion, crosslink density, and water contact angle of the resin composites tested in the study. Relative to the degree of conversion, SA_YF had the lowest conversion of C=C double bonds, followed by BF. These resin composites resulted in lower polymerization levels than the SA_DF and control materials ($p<0.001$), which did not differ between each other ($p=0.129$). All materials showed statistically similar crosslink density values ($p=0.620$). BF demonstrated the least hydrophilic behavior of tested materials, followed by the control, which all resulted in higher water contact angle values when compared with the self-adhesive materials ($p \leq 0.001$); SA_YF and SA_DF did not differ between each other ($p=0.202$).

Table 2: *Results (Mean and Standard Deviation) for the Cross-Link Density (CLD), Degree of Conversion (DC), and Water Contact Angle (WCA) of Groups Tested*[a]

Group	DC (%)	CLD (%)	WCA (°)
SA_YF	28.6 (1.4) C	83.1 (2.2) A	60.9 (2.3) C
SA_DF	63.6 (2.3) A	89.7 (9.8) A	57.3 (3.4) C
BF	49.7 (0.8) B	82.9 (9.0) A	70.7 (2.0) A
OF (control)	60.0 (2.0) A	86.3 (5.3) A	65.0 (2.3) B

Abbreviations: SA_YF, Y-flow Self-Adhesive; SA_DF, Dyad Flow Self-Adhesive; BF, Filtek Bulk Fill Flow; OF, Opallis Flow.

[a] *Mean (standard deviation); distinct uppercase letters in the same column indicate statistically significant differences among the groups (*p*<0.05).*

The results for the color alteration of materials after water storage for 6 months are shown in Figure 2A. BF demonstrated higher ΔE_{00} values than the other resin composites ($p \leq 0.001$), which did not differ among each other ($p \geq 0.677$). The control (OF) was the only resin composite that resulted in color alteration values below the 50% acceptability threshold (ΔE_{00}=1.8). Lastly, the results for the cell viability assay are presented in Figure 2B. According to the statistical analysis, cell viability was significantly influenced by both material and incubation time ($p<0.001$), and a significant interaction between these two variables was also observed ($p<0.0001$). At the 24-hour time interval, cell viability was statistically similar among the different resin composites ($p>0.05$), whereas it increased in the following order at the 48-hour time interval: SA_YF < SA_DF < OF (control) = BF. While the BF and OF materials displayed similar biocompatibility at both time intervals investigated, cell viability was significantly reduced at 48 hours for the self-adhesive resin composites.

DISCUSSION

The present study compared the effects of three contemporary resin composites with low viscosity to that of a flowable conventional restorative material.

Overall, the materials tested in this study performed adequately in terms of physical and biological properties. However, our main goal was to investigate the bonding ability to dentin of the different types of flowable resin composites, since this analysis helps to predict their clinical performance.[20] At the immediate test, the two self-adhesive materials resulted in resin-dentin bonds similar to those of the bulk-fill material (Figure 1A), in spite of their different degree of conversion values. Here, we may assume that the similar bonding mechanism of all the three foregoing resin composites had an influence on their similar bonding ability. Indeed, SA_YF and SA_DF are composed of GPDM and 10-MDP monomers, which create the necessary acidic environment for etching dentin; the flowable nature of the materials may also allow resin infiltration into the etched dentin, creating an adequate hybrid layer. The other resin composites had a similar bonding mechanism, in which both the bulk-fill and the conventional control were applied after application of a universal bonding agent based on 10-MDP. It is of the utmost importance to understand that the self-adhesive resin composites created resin-dentin bonds

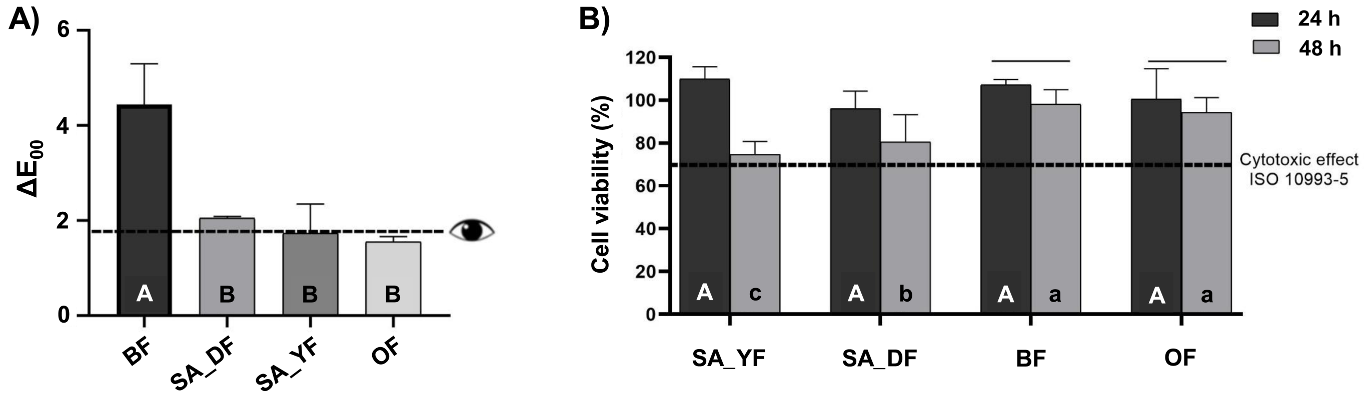

Figure 2. *(A): Graphs showing color alteration results (ΔE00). For color alteration, the same uppercase and lowercase letters indicate no statistically significant differences between resin composites at 24 hours and 6-month time intervals among resin composites (*p*<0.05); value indicated by dotted line at ΔE = 1.8 corresponds to 50% visual acceptability under clinical conditions, which was used as a threshold for visually detectable color changes. (B): Cell viability results. For cell viability, the same uppercase and lowercase letters indicate no statistically significant differences among resin composites at 24-hour and 48-hour time intervals, respectively (*p*<0.05). Bars under the same horizontal line indicate statistically similar cell viability between different incubation time intervals (*p*>0.05). Abbreviations: SA_YF, Y-flow Self-Adhesive; SA_DF, Dyad Flow Self-Adhesive; BF, Filtek Bulk Fill Flow; OF, Opallis Flow.*

that were statistically similar to the bonds derived from the bulk-fill material, confirming the bonding ability of the former. Nevertheless, the conventional control resulted in stronger resin-dentin bonds, probably due to the considerably high conversion of monomers of this group in particular.

Notably, the higher the level of polymerization of the restorative material, the better its cohesiveness and mechanical strength during fracture. A higher number of mixed failures also occurred in the control, which suggested a more densely packed structure that had a positive influence on the bond strength results. More importantly, OF seemed to contain a larger amount of filler particles, since it is a material with average flowability, differing from the other materials that have a less viscous consistency (low viscosity category). Here, the higher the level of filler loading, the lower the negative effects of polymerization stress,[21] and consequently, the better the quality of resin-dentin interfaces. This would perhaps explain the better bonding performance of the control over the self-adhesive and bulk-fill resin composites in the immediate test time interval.[22]

It is known that the degree of conversion of resin-based polymer systems may increase after *in situ* polymerization,[23] therefore the resin-dentin bonds may also become stronger with time.[24] This was especially true for SA_YF, which showed increased bond strength values after long-term water storage, possibly due to the gain in the conversion of monomers, which were previously low (<30%) at immediate testing, strengthening the hybrid layer during aging simulation. Moreover, SA_YF is a HEMA-free material, and considering that HEMA can inhibit the adhesion mechanism of acidic monomers,[25,26] one may suggest that hybridization with SA_YF formed a stable adhesive interface with strengthening potential. An aspect that deserves further discussion relates to the presence of 10-MDP in the resin-dentin bonds created with the application of SA_YF, BF, and OF materials, but not with application of SA_DF. The 10-MDP monomer is one of the most important acidic ingredients responsible for the formation of stable hybrid layers over time,[27] since it is capable of creating a stable 10-MDP-calcium salt with low solubility, thus forming a nano-layered bonding interface between the resin phase and hydroxyapatite.[28] Remarkably, this nano-layer has greater resistance to hydrolysis, as suggested elsewhere.[29]

The degree of conversion varied widely among the resin composites, with the two self-adhesive materials exhibiting the lowest (SA_YF) and the highest (SA_DF) polymerization states of the materials tested in the study (Table 2). Remarkably, the conversion of monomers is an intrinsic property relying on chemical features such as the type of resin monomers, type/concentration of initiation system, and rheological characteristics of materials.[30] Bearing this in mind, something related to the chemistry of SA_YF might have played a role in the ~2.2 times lower DC values, when compared with the SA_DF counterpart. Here, both self-adhesive composites were composed of functional acidic monomers, which are usually characterized by having only one polymerizable methacrylate group per molecule, thus limiting the material to acquiring an extended level of polymerization.[31] However, one may understand that SA_YF and SA_DF are both composed of GPDM (Table 1), which is an acidic monomer that has two polymerizable methacrylate groups, ie, a factor that may have positively influenced polymerization of the materials.[32] Conversely, while SA_DF resulted in high conversion of monomers, SA_YF yielded values below the 30% level. We may assume that the presence of 10-MDP, which is an acidic monomer with only one methacrylate group available for polymerization, contributed to the reduced polymerization level of SA_YF.

Despite the differences in polymerization levels reached by each resin composite, the materials did not differ in terms of their crosslink density. Overall, this property relates to the amount of chemical reactions that link various polymer chains together, playing an important role in the physico-mechanical behavior of resin-based polymer systems.[33] The method used in our study to determine the material crosslink density was to evaluate the percentage reduction in hardness after immersion in ethanol, which provided an indirect estimation only. Nevertheless, it may add to understanding of the crosslink state of the materials tested.[34,35] It is noteworthy that the crosslink density of the contemporary resin composites was similar to that of the control, suggesting that all materials shared a similar stability when exposed to a hydrolytic medium. Thus, we may assume the materials may undergo hygroscopic and hydrolytic degradation in the same manner, which is interesting for confirming the clinical suitability of the self-adhesive and bulk-fill materials when compared with the conventional control.[33]

The characteristic of wettability was also investigated in our study, since this surface property may be positively correlated to the bonding ability of resin composites.[36] Although the materials displayed water contact angle values lower than 90°, suggesting their hydrophilic behavior,[37] the bulk-fill composite was the least hydrophilic of the materials tested in the study, followed by the control and then by the self-adhesive materials (Table 2). Of note, the presence of functional acidic monomers (eg, GPDM or 10-MDP) in the self-adhesive resin composites may have increased their hydrophilicity, since both acidic monomers have a hydrophilic moiety (eg, pendant hydroxyls) capable of increasing wettability properties.[32] Despite the similar water contact values obtained when comparing the two self-adhesive composites, SA_DF showed the lowest values, probably due to the more hydrophilic nature of GPDM when compared with 10-MDP.[31,32] Indeed, considering that SA_YF is composed of both GPDM and 10-MDP, one may infer that hydrophilicity would be slightly lower in the aforementioned resin system.[7] Despite the importance of hydrophilicity for adhesion purposes, the higher the hydrophilic behavior of a resin system, the more hydrolytically unstable it will be.[38] Here, we did not evaluate the hygroscopic and hydrolytic properties of the resin composites which, therefore, deserves further investigation. However, we may assume that the bulk-fill resin composite would demonstrate a slightly greater resistance to hydrolysis due to its higher water contact angle values. This may be explained by the procrylat-based composition of BF (Table 1) since this ingredient is a proprietary monomer analog to Bis-GMA, showing an extended molar mass but still having properties of low viscosity and low water solubility, thereby contributing to a less hydrophilic polymer system, as verified in our findings.

The present study also characterized the resin composites by evaluating their color stability after long-term (6 months) water storage. Color plays an important role in obtaining optimal esthetics in resin composite restorations, and color stability may depend on several compositional features, such as the type of resin matrix, the size and shape of filler particles, the depth of polymerization achieved by the material, and the nature of pigments present for making the restorative tooth-colored.[39] As shown in Figure 2A, the control group (OF) was the only resin composite that resulted in $\Delta E_{00} \leq 1.8$ (ie, below the 50% acceptability threshold),[18] thereby representing the most physically stable material. One may suggest that silane functionalization of the inorganic fillers found in OF created a strong chemical interaction between resin matrix and filler phase, resulting in a material with greater color stability.[40] Considering that information on the silanization of fillers for the other resin composites was not supplied (Table 1), we may assume these materials would undergo faster discoloration than the control. One interesting finding of our study was that the bulk-fill resin composite exhibited the greatest color alteration, which was at least two times higher than that of the other materials. Indeed, earlier in the article we inferred that the bulk-fill material would offer greater resistance to hydrolysis due to its more hydrophobic behavior, but according to the color alteration results, BF suffered from color instability to a greater extent, suggesting an extended degradation profile compared to the other materials. Notably, aspects such as the universal color shade of BF and its characteristic of increased depth of polymerization (ie, 4 mm), which differed from the other resin composites, could have played a role in the overall absorbance and reflection of light within the bulk-fill composite, making the material optically less stable after long-term wet storage.[41] Equally as important, the self-adhesive composites showed ΔE_{00} values close to the 1.8 threshold and statistically similar to the control, indicating their suitability as restorative materials, especially for creating restorations with durable esthetics.[18]

With regard to cell viability, all resin composites demonstrated a non-toxic behavior, resulting in cell proliferation levels above 70% (Figure 2B), thus reaching the requirement for a biocompatible material.[19] It is important to highlight that resin composites are expected to release uncured monomers, oligomers, or other non-reacted ingredients (eg, initiators, stabilizers, pigments) after polymerization, posing a risk for cell toxicity.[42-44] Notwithstanding, cell viability was similarly distributed among the resin composites at the 24-hour time interval, indicating their clinical safety at earlier stages. One may consider this result an advantage for the self-adhesive composites, since they represent the most recent advancements in the development of resin-based restorative materials, and due to their biocompatible properties, dental practitioners could more readily begin to consider the clinical use of these restoratives. However, SA_YF and SA_DF created a considerably lower cell viability scenario at the 48-hour time interval when compared with the bulk-fill and control materials, probably due to the more acidic behavior of the former.[45] It is important to highlight that despite the reduction in cell viability, the self-adhesive composites have still performed as non-toxic materials, and the reduction in their biocompatibility may be a consequence of the release of unreacted acidic monomers, which may result in an initial acidic medium, although without an intense cytotoxic potential.[46] It is worth mentioning that the polymerization of dimethacrylate-based resins is never complete, so that some cytotoxic reaction may occur over time due to the release of unpolymerized monomers.[43]

The present study evaluated the mechanical, physical, and biological properties of different contemporary flowable resin composites, and according to our findings, the restorative materials performed mostly different among them, thus leading to the rejection of the null hypothesis.

CONCLUSIONS

Within the limitations of this *in vitro* study, it could be concluded that flowable resin composites of different categories, ie, self-adhesive, bulk-fill, or conventional materials, performed differently among each other, depending on the properties investigated. The chemical composition of materials appeared to be an influential factor on their physico-mechanical and biological behavior. Overall, the self-adhesive flowable resin composites showed promising results.

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Conflict of Interest

The authors of this article certify that they have no proprietary, financial, or other personal interest of any nature or kind in any product, service, and/or company that is presented in this article.

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