

# Effect of Storage Time on Bond Strength Performance of Multimode Adhesives to Indirect Resin Composite and Lithium Disilicate Glass Ceramic

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## Clinical Relevance

Multimode adhesives provide reliable bonding to sandblasted indirect resin composites in the long term; however, the use of separate bottles of silane and bonding resin is still recommended for durable bonding to etched glass-based ceramic substrates.

## SUMMARY

**Purpose:** To investigate the bond strength performance of multimode adhesives (MMAs) to indirect resin composite and lithium disilicate glass ceramic after 24 hours or one year of water storage.

**Methods and Materials:** Thirty flat and polished plates of indirect resin composite (Epi-

cord) and thirty lithium disilicate glass ceramic plates (IPS e.max Press) were prepared. Surfaces were pretreated using sandblasting (indirect resin composite) or hydrofluoric acid (glass-based ceramic). Specimens were bonded with one of two MMAs (Scotchbond Universal [SBU] or All-Bond Universal [ABU]) or ceramic primer and hydrophobic bonding (RelyX Ceramic Primer and

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**Adper Scotchbond Multi-Purpose Bond**) as a control ( $n=10$ ). Resin cement cylinders (0.75 mm in diameter  $\times$  0.5 mm in height) were bonded to both substrate surfaces using the respective adhesives. After 24 hours or one year of water storage, bonding performance was measured by microshear bond strength (MSBS) testing. Results were analyzed using three-way ANOVA with Bonferroni post hoc tests ( $\alpha=0.05$ ).

**Results:** For indirect resin composite, significantly higher MSBS values were found for ABU after 24 hours ( $ABU > SBU = \text{control}$ ); however, no significant difference among the adhesives was observed after one year ( $p>0.05$ ). For glass-based ceramic, significantly different bond strengths were observed among the adhesives after 24 hours ( $\text{control} = ABU > SBU$ ) and one year ( $\text{control} > SBU = ABU$ ;  $p<0.05$ ).

**Conclusions:** Both MMAs tested can be considered effective alternatives for bonding to sand-blasted indirect resin composite after aging, as they showed similar bond performance to that of the control group. However, separate bottles of silane bonding resin showed higher MSBS values and more durable bonding for etched glass-based ceramic.

## INTRODUCTION

The demand for esthetic metal-free dental treatments such as inlays, onlays, veneers, and crowns has increased in recent years, which has contributed greatly to the development of adhesive cementing systems for ceramic and indirect resin composite restorations.<sup>1-5</sup> The indirect restorative procedure involves the creation of a machined or laboratory-fabricated restoration, followed by adequate conditioning of the tooth and indirect restoration, and then placement of the restoration with a resin cement.<sup>6</sup> Unlike that with the direct technique, the indirect method can be used to restore the mechanical and biological functions of the tooth with minimal intraoral polymerization shrinkage;<sup>7</sup> this can provide better control of the contact and contour of proximal restorations<sup>8</sup> and therefore enhances the marginal adaptation of the restorative material in the long term.<sup>9</sup>

On the other hand, the bond between indirect restorations and tooth structure can be challenging, since two different interfaces must be considered: that between the dentin–enamel substrate and the resin cement and that between the resin cement and

the internal surface of the indirect restoration. For these bonds to form, the tooth substrate and the internal surface of the restoration are conventionally pretreated. During pretreatment of the tooth structure, resin cements can be classified as etch-and-rinse adhesive systems, adhesives containing self-etch primers, or self-adhesive resin cements.<sup>10</sup> Among these categories of resin cements, there is a growing interest in self-adhesive resin cements because of their ease of handling, good esthetics, and suitability for indirect restorations. Such cements combine the properties of adhesiveness and cement into a single step, without the need for additional pretreatment of the tooth. Therefore, compared with conventional resin cements, self-adhesive resin cements are expected to be less technique sensitive during the luting procedure. In addition, studies have shown that self-adhesive resin cements can provide improved sealing ability<sup>11</sup> with durable bonding performance on smear-covered dentin.<sup>12</sup>

Besides significant improvements achieved in the attachment of adhesive cementing systems to tooth structures, bonding between the indirect restoration substrate and the resin cement has also been the focus of several studies.<sup>13,14</sup> Because of the differences in the composition of indirect restorative materials, different internal surface pretreatments seem crucial for their intraoral retention.<sup>15,16</sup> Pressable lithium disilicate-based glass is a ceramic composed of one glassy phase and at least one crystallized phase. Ceramic restorations involve a high degree of crystallization that enhances their mechanical properties.<sup>17</sup> Meanwhile, indirect resin composite restorations are subjected to secondary curing with light and/or heat to increase the resin conversion that enhances their wear resistance; however, this lessens the potential for chemical bonding, as the number of residual-free carbon double bonds is decreased.<sup>16</sup> Conventionally, for reliable adhesion to indirect restorations, the internal surface of the restoration is roughened. As a result, the surface area for bonding and wettability of the adhesive–cement to the restoration is increased; this allows chemical bonds to form between the ceramic or fillers and the cement.<sup>10</sup> Among the surface pretreatments used for indirect restorations, successful bonding to glass-based ceramic surfaces can be achieved by dissolving their glassy phase with hydrofluoric acid (HFA),<sup>17,18</sup> while sandblasting the surface of an indirect resin composite is recommended.<sup>19,20</sup> Both of these methods provide better mechanical and chemical interaction during silane

application prior to the luting agent.<sup>15,21</sup> A silane coupling agent is used conventionally as an adhesion promoter for silica-based materials and forms chemical bonds with the inorganic phase of the indirect restoration and the organic phase of the resin cement.<sup>21,22</sup>

Universal or multimode adhesives (MMAs) are the newest category of simplified one-bottle adhesives and are considered applicable to different substrates such as enamel, dentin, alloys, zirconia, ceramics, and composites. They have been marketed with an indication that they provide a chemical bond between indirect restoration substrates and resin cements without the need for primers or activators. This simplified strategy might be due to the presence of 10-methacryloyloxydecyl dihydrogen phosphate (MDP) monomer in their composition, which can promote adhesion to surfaces based on calcium,<sup>23</sup> metal,<sup>24</sup> or zirconia.<sup>25,26</sup> In addition, MMAs can also contain silane in the same bottle, which is expected to improve their bonding to silica-based ceramic.

The dihydrogen phosphate group from the MDP monomer is responsible for priming and bonding, while its long carboxyl chain provides the hydrophobic properties and hydrolytic stability of acidic monomers. For enamel and dentin substrates, MDP forms a strong ionic bond with calcium from hydroxyapatite that forms calcium salts with low solubility, which may be responsible for the good long-term performance of MDP-containing adhesives.<sup>23,27,28</sup> For non-silica-based substrates, such as metal or zirconia, the hydrophilic phosphate terminal end of MDP interacts chemically with the oxides on the internal surface of restorations, while the hydrophobic methacrylate terminal end copolymerizes the resin monomers of the cement.<sup>29</sup> However, for silica-based indirect restorations such as feldspathic porcelain, leucite-reinforced ceramic, or lithium disilicate glass ceramic, the reaction between silane and MDP promotes the bonding mechanism, improving surface wettability. The free silanol groups form hydrogen bonds with the hydroxyl groups of the indirect restoration. Then, cross-linkages are formed between the methacrylate groups of the cement with organofunctional groups from the silane coupling agent, as well as between the siloxane bonds and the restoration substrate.<sup>22</sup> Because of the versatility of the substrate application, MMAs may also be suitable for intraoral restoration repairs, since they could be a practical alternative to bonding different fractured substrates at the same time.<sup>30</sup> Nevertheless, few studies have investigated the long-term durability of this new

category of one-bottle adhesive<sup>31</sup> or their application with different indirect restoration substrates.<sup>32,33</sup>

Therefore, the aim of this study was to determine the bonding effectiveness of two different MMAs with their respective resin cements to indirect resin composite and lithium disilicate glass ceramic substrates after 24 hours or one year of water storage. In addition, scanning electron microscopy (SEM) was used to evaluate fracture mode patterns. The null hypothesis tested was that there would be no significant difference in bond strength among the materials assessed after 24 hours or one year of storage for both substrates.

## METHODS AND MATERIALS

A total of 60 standardized rectangular plates (15-mm wide  $\times$  6-mm long  $\times$  1.5-mm thick) were obtained for this study. Thirty of the plates were fabricated from indirect resin composite (Epicord Dentin A2, Kuraray Noritake Dental, Tokyo, Japan), while the other thirty were fabricated from lithium disilicate-based glass ceramic (IPS e.max Press MO-0 ingots, Ivoclar Vivadent AG, Schaan, Liechtenstein) by using the lost wax and hot press technique, according to the manufacturers' instructions.

For the indirect resin composite, the rectangular plates were prepared by a single increment of Epicord into a silicone mold, which was covered by a thin transparent film (KerrHawe Striproll, KerrHawe, Bioggio, Switzerland), followed by a glass slab, and then light activated using a halogen light-curing unit (intensity = 600 mW/cm<sup>2</sup>; Optilux 501, Kerr, Orange, CA, USA) for 40 seconds. The specimens were removed from the mold, and the remaining surfaces were light cured for an additional 40 seconds each.

For the lithium disilicate-based glass ceramic, rectangular wax patterns were fabricated, sprued, and attached to a muffle base with a surrounding paper cylinder. The wax patterns were invested with phosphate-based material (IPS PressVest Speed, Ivoclar Vivadent AG), and the wax was eliminated in an automatic furnace (Vulcan A-550, Degussa-Ney, Yucaipa, CA, USA) at 850°C for one hour. The IPS e.max Press ingots were then pressed into the molds in an automatic press furnace (EP 600, Ivoclar Vivadent AG).

After cooling, both substrate surfaces were wet ground with 400-, 600-, and 800-grit silicon carbide papers (Norton, Vinhedo, SP, Brazil) and ultrasonically cleaned in a water bath for five minutes to remove the remaining debris and air-dried. The

Table 1: <i>Materials Used in This Study</i>		
Material (Batch Number)	Composition	Application Technique
RelyX Ceramic Primer (N351206); 3M ESPE, St Paul, MN, USA	Ethyl alcohol, water, methacryloxypropyltrimethoxysilane	Apply RelyX Ceramic Primer. Air dry for 5 s.
Adper Scotchbond Multi-Purpose Bond (N205453); 3M ESPE, St Paul, MN, USA	Bond: Bis-GMA, HEMA, triphenylantimony	Apply adhesive for 10 s. Gentle air blow for 5 s. Light cure for 10 s.
RelyX ARC (N339863); 3M ESPE, St Paul, MN, USA	Silane-treated ceramic, TEGDMA, Bis-GMA, silane-treated silica, reacted polycaprolactone polymer, 2-benzotriazolyl-4-methylphenol, benzoyl peroxide	Dispense the cement onto the mixing pad and mix for 10 s. Insert the resin cement into the Tygon tube. Light cure for 40 s.
Scotchbond Universal (472387); 3M ESPE, St. Paul, MN, USA	Adhesive: MDP, Bis-GMA, phosphate monomer, dimethacrylate resins, HEMA, methacrylate-modified polyalkenoic acid copolymer, filler, ethanol, water, initiators, silane-treated silica	Apply adhesive for 20 s. Gentle air blow for 5 s.
RelyX Ultimate (467302); 3M ESPE, St. Paul, MN, USA	Silane-treated glass powder, 2-propenoic acid, 2-methyl-1,1'-[1-(hydroxymethyl)-1,2-ethanediyl]ester, reaction products with 2-hydroxy-1,3-propanediyl dimethacrylate and phosphorus oxide, TEGDMA, silane-treated silica, oxide glass chemicals, sodium persulfate, tert-butyl peroxy-3,5,5-trimethylhexanoate, acetate monohydrate	Dispense the cement onto the mixing pad using an intraoral tip. Insert the resin cement into the Tygon tube. Light cure for 40 s.
All-Bond Universal (1200003968); BISCO, Schaumburg, IL, USA	Adhesive: MDP, Bis-GMA, HEMA, ethanol, water, initiators	Apply adhesive. Light cure for 10 s.
Duo-Link (1200006424); BISCO, Schaumburg, IL, USA	Bis-GMA, triethyleneglycol dimethacrylate, urethane dimethacrylate, glass-filler	Dispense the cement onto the mixing pad using an intraoral tip. Insert the resin cement into the Tygon tube. Light cure for 40 s.
Abbreviations: Bis-GMA, bisphenol-A diglycidyl ether dimethacrylate; HEMA, hydroxyethyl methacrylate; MDP, 10-methacryloyloxydecyl dihydrogen phosphate; TEGDMA, triethylene glycol dimethacrylate.		

indirect resin composite specimens were sandblasted with 50-μm aluminum oxide particles (Danville Engineering Inc, San Ramon, CA, USA) for 10 seconds (air pressure, 0.552 MPa; distance from the tip, 1.5 cm). All the resin plates were then subjected to further ultrasonic cleaning in a water bath for another five minutes and were air-dried. On the other hand, the glass-based ceramic specimens were etched with 10% HFA gel (Dentsply Caulk, Milford, DE, USA) for 15 seconds, rinsed with water for 15 seconds, and air-dried.

After surface pretreatment, the plates of each substrate were randomly assigned to three groups according to the bonding materials used (n=10 per adhesive/substrate): one of two one-step MMAs with their respective adhesive resin cements (Scotchbond Universal [SBU] with RelyX Ultimate [3M ESPE, St Paul, MN, USA] or All-Bond Universal [ABU] with Duo-Link [Bisco Inc, Schaumburg, IL, USA]) or a control ceramic primer with hydrophobic bonding resin and an adhesive resin cement (RelyX Ceramic Primer and Adper Scotchbond Multi-Purpose Bond with RelyX ARC [3M ESPE]). The composition of the

materials and the application techniques used in this study are included in Table 1.

For microshear bond strength (MSBS) testing, hollow cylinders of 0.5-mm height were cut from micro-bore Tygon tubing (internal diameter, 0.75 mm; Norton Performance Plastics, Akron, OH, USA) and were used as molds for the resin cement luting procedure. The plates were randomly assigned to receive an application of each MMA or control ceramic primer with hydrophobic bonding resin onto the substrate surfaces. Without prior light irradiation, four Tygon tubes were then placed at four locations on the surface of each plate forming a centered straight line, at approximately 3.0 mm apart from the center of each Tygon tube. The resin cements were mixed according to the manufacturers' instructions and carefully inserted into the tubing on their respective adhesive and substrate surfaces. Small resin cement cylinders (approximately 0.75 mm in diameter × 0.5 mm in height) were obtained after polymerization using a halogen light-curing unit (intensity, 600 mW/cm<sup>2</sup>; Optilux 501, Kerr) for 40 seconds.

After 24 hours of water storage at 37°C, the Tygon tubing was removed carefully with a thin steel

Table 2: Mean (SD) Microshear Bond Strength of Adhesive Systems to Indirect Resin Composite and Lithium Disilicate Glass Ceramic (in MPa)<sup>a</sup>

Material	Indirect Resin Composite Substrate		Lithium Disilicate Glass Ceramic Substrate	
	After 24 h Water Storage	After 1 y Water Storage	After 24 h Water Storage	After 1 y Water Storage
RelyX Ceramic Primer and Adper Scotchbond Multi-Purpose Bond (control)	25.5 (7.9) <sup>A,a*</sup>	22.7 (2.7) <sup>A,a**</sup>	35.3 (8.5) <sup>A,b*</sup>	31.2 (5.9) <sup>A,b**</sup>
Scotchbond Universal (SBU)	26.6 (5.6) <sup>A,a</sup>	20.2 (3.1) <sup>B,a</sup>	23.9 (6.1) <sup>A,a</sup>	21.3 (5.6) <sup>A,a</sup>
All-Bond Universal (ABU)	32.7 (3.3) <sup>A,b</sup>	25.1 (2.0) <sup>B,a**</sup>	31.5 (7.0) <sup>A,b</sup>	16.9 (4.4) <sup>B,a**</sup>

<sup>a</sup> Data are presented as the mean (standard deviation) in MPa (n=10). Identical capital letters in a row within the same substrate after 24 hours and one year of water storage indicate the absence of any statistically significant difference. Identical lowercase letters in a column indicate the absence of any statistically significant difference. Comparisons within the same material and storage period between different substrates and marked with one asterisk for 24 hours and two asterisks for one year water storage are statistically significant. (Analysis of variance and Bonferroni post hoc test; significance at  $p < 0.05$ .)

cutting blade to expose the resin cement cylinder. The four resin cement cylinders obtained from each plate were further divided into two subgroups, with two cylinders being tested after a further 24 hours of water storage and the other two being tested after one year of water storage. The average MSBS value obtained from two cylinders on the same plate was considered the mean value of one sample for each storage period (n=10 per adhesive cementing system/storage period). Each plate was fixed with cyanoacrylate glue (Model Repair II Blue; Sankin Industry Co, Tokyo, Japan) to a jig in a universal testing machine (EZ Test, Shimadzu, Kyoto, Japan) in such a way that the straight line formed by the cement cylinders was perpendicular to the force. A thin wire (0.2 mm in diameter) was looped around the cement cylinder, making contact with half of its circumference, and held gently against the cement–indirect restoration substrate interface. A shear force was applied to each specimen at a cross-head speed of 1 mm/min until failure occurred. Tweezers were used to position the wire at the boundary of the cement and the indirect restoration substrates. The resin cement–indirect resin composite interface or the resin cement–lithium disilicate glass ceramic interface, the wire loop, and the center of the load cell were aligned as straight as possible to ensure the desired orientation of the shear test force.

After MSBS testing, the fractured surfaces were mounted onto brass stubs, gold coated, and observed under SEM (JSM5600, JEOL Ltd, Tokyo, Japan). The failure mode pattern of all specimens submitted to MSBS testing was evaluated using SEM micrographs at a magnification of 1000×. For the indirect resin composite and lithium disilicate glass ceramic specimens, the failure mode was determined and classified as follows: prefailure, cohesive failure in resin cement, mixed failure of adhesive and resin cement, failure between adhesive and indirect resin

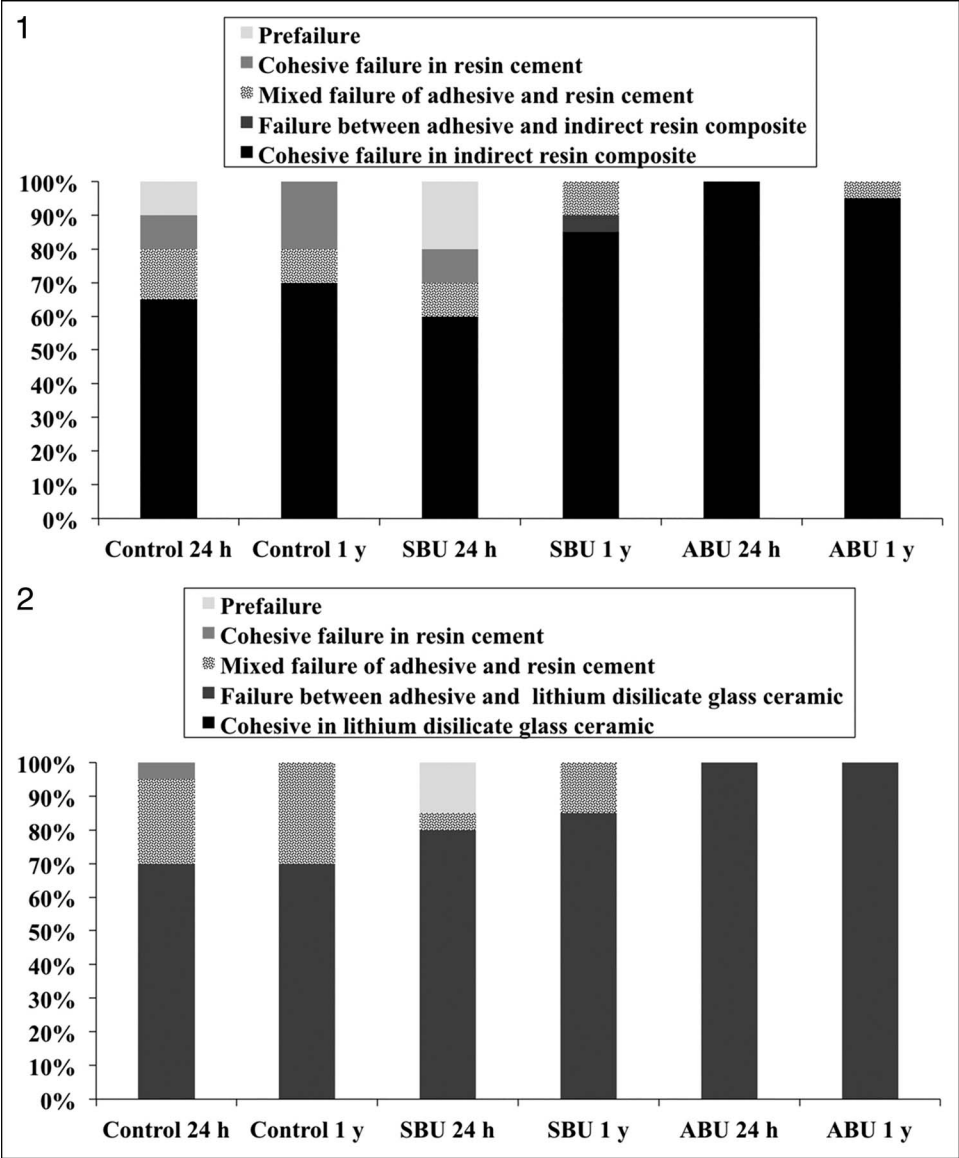
composite or lithium disilicate glass ceramic, or cohesive failure in indirect resin composite or lithium disilicate glass ceramic.

The MSBS data were statistically analyzed using three-way analysis of variance (ANOVA) with the significance level defined as  $\alpha=0.05$ ; bond strengths to indirect resin composite or glass-based ceramic were used as dependent variables, and the adhesive cementing system, storage period, and substrate were used as factors. Bonferroni post hoc tests with UNIANOVA syntax were used for multiple comparisons of significant differences in bond strength means. All statistical analyses were performed using the Statistical Package for Social Sciences (SPSS for Windows, version 16.0, SPSS, Chicago, IL, USA).

## RESULTS

The means and standard deviations of the MSBS values obtained in this study are presented in Table 2. Three-way ANOVA showed that bond strength values for indirect resin composite and lithium disilicate glass ceramic substrates were significantly influenced by the adhesive cementing system used ( $p < 0.001$ ) and by the storage period ( $p < 0.001$ ) but not by the substrate ( $p = 0.227$ ). The interactions of these three factors were not significant ( $p = 0.104$ ). On the other hand, significant statistical interaction was observed between the adhesive cementing system and the storage period ( $p = 0.005$ ) and between the adhesive cementing system and the substrate ( $p < 0.001$ ). Significant statistical interaction was not found between the storage period and the substrate ( $p = 0.458$ ).

Bonferroni post hoc tests revealed statistically significant differences between the bond strength results after 24 hours or one year of water storage for both substrates. For indirect resin composite, the ABU group displayed higher MSBS values than the



SBU and control groups after 24 hours. After one year of storage, no statistically significant differences were found among all of the materials used for this substrate, while there was a significant decrease in bond strength within the ABU and SBU groups compared with baseline ( $p<0.05$ ; Table 2).

On the other hand, for lithium disilicate glass ceramic after 24 hours, the control group showed significantly higher MSBS values than the SBU group but did not differ from the ABU group. After long-term storage, the control group showed higher MSBS values, but no statistically significant difference was found in MSBS values between the ABU and SBU groups. However, a significant decrease in bond strength was observed within the ABU group compared with the baseline value (Table 2).

Within the control group, there was a significantly higher MSBS to lithium disilicate glass ceramic as compared with indirect resin composite after 24 hours or one year of storage. On the other hand, no statistically significant difference was observed for SBU after the same storage period, regardless of the substrate used. For ABU, bonding to the indirect resin composite substrate resulted in higher bond strength values than bonding to lithium disilicate glass ceramic after one year only.

The distribution of the modes of failure is summarized in Figure 1 for indirect resin composite substrate and Figure 2 for lithium disilicate glass ceramic substrate. Representative high-magnification SEM micrographs of mode patterns are presented in Figure 3. Cohesive failure was mainly

Figure 1. Distribution (%) of failure modes after 24 hours (24 h) and one year (1 y) of water storage for the indirect resin composite substrate.

Figure 2. Distribution (%) of failure modes after 24 hours (24 h) and one year (1 y) of water storage for the lithium disilicate glass ceramic substrate.

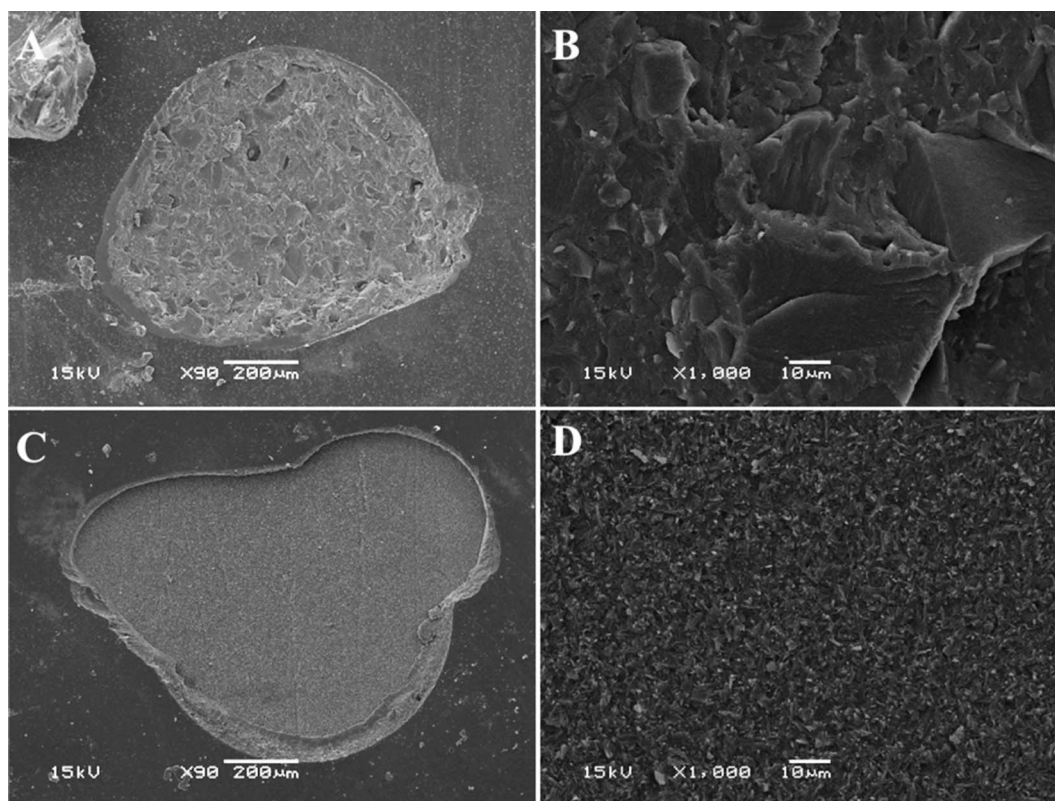


Figure 3. Scanning electron micrographs of representative main fracture patterns of the indirect resin composite substrate (A and B) and the lithium disilicate glass ceramic substrate (C and D). (A and C): 90 $\times$  magnification; (B and D): 1000 $\times$  magnification. (A) and (B) show images of cohesive failure in the indirect resin composite. (C) and (D) show images of failure between the adhesive and lithium disilicate glass ceramic.

observed in indirect resin composite for all of the materials applied (Figure 3A,B) for both storage times. For lithium disilicate glass ceramic, failure in the adhesive was predominant for both periods of storage (Figure 3C,D).

### DISCUSSION

The present study evaluated the effect of storage time on the bonding effectiveness of two MMAs to indirect resin composite and lithium disilicate glass ceramic and compared their bonding performance to a conventional method of two separate bottles of silane agent and hydrophobic bonding resin. The results obtained after 24 hours indicated that, among the materials tested, ABU provided the highest bond strength values to indirect resin composite and showed similar bond performance to the control group for the lithium disilicate glass ceramic. However, the storage time significantly affected the bond performance of both MMAs to indirect resin composite and also significantly decreased the bond strength, particularly for ABU, which was bonded to the lithium disilicate glass ceramic. Since there was a significant difference in

bond strength among the materials tested after 24 hours or one year of storage for indirect resin composite and lithium disilicate glass ceramic substrates, the null hypothesis of the current study was rejected.

In this study, the MSBS test was successfully performed for all specimens, and bonding performance was compared among the adhesive systems with two indirect restorative materials. The MSBS test was chosen because of the advantages of bonding tests with small and round bonded areas as well as the ease of sample preparation,<sup>14</sup> since it does not require cutting procedures such as those used for sample preparation in the microtensile bond strength test.<sup>34</sup>

In the current study, sandblasting with aluminum oxide particles was used as a surface pretreatment for indirect resin composite, and HFA etching was used for lithium disilicate glass ceramic. Studies have reported that sandblasting indirect resin composite surfaces can produce higher bond strength compared with acid etching by producing extensive surface roughening.<sup>35,36</sup> On the other hand, conditioning of lithium disilicate glass ceramic substrates

using HFA has been shown to be the most effective pretreatment method to increase bond strength performance, via dissolution of the glassy matrix, roughening of the surface by exposure of crystals, and subsequent enhancement of the potential for micromechanical retention.<sup>14,15,17,18</sup> Thus, the successful creation of micropores for wetting and infiltration of silane monomers, bonding resins, and resin cements was expected because of the application of these pretreatments to different substrates used in the present study.

RelyX Ceramic Primer and Adper Scotchbond Multi-Purpose Bond were used in the control group in the current study. This method uses two separate bottles of silane agent and hydrophobic bonding resin. RelyX Ceramic Primer contains 3-methacryloxypropyltrimethoxysilane diluted in an ethanol-water solution, while Adper Scotchbond Multi-Purpose Bond contains bisphenol-A diglycidyl ether dimethacrylate (Bis-GMA) as a cross-linker and hydroxyethyl methacrylate (HEMA), which has solvent-like properties.<sup>37</sup> The silane molecules from the RelyX Ceramic Primer react with water, forming three silanol groups from the corresponding methoxy groups. These silanol groups form a siloxane network with the silica surface and make covalent bridges with the hydroxyl groups on inorganic substrate surfaces.<sup>20,22</sup> Later, the monomeric ends of the silane molecules on the pretreated surfaces may react with the methacrylate groups of the bonding agent.<sup>38</sup> In this way, a linkage between the substrate, the silane agent, and the hydrophobic bonding resin can be formed.

Silane application is very effective in promoting adhesion and may even be crucial for durable bonding, particularly with silica-based materials.<sup>15,22</sup> Moreover, the subsequent application of a bonding resin as an intermediate agent facilitates the penetration of resin monomers and the resin cement into the irregularities formed by the acid etching/sandblasting pretreatment; this results in micromechanical interlocking<sup>39</sup> and ultimately enhances bond strength.<sup>40</sup> This could explain the optimal results observed for the control group that had similar bond strength values after 24 hours and one year of storage for each substrate. Interestingly, only the control group had significantly higher MSBS values for specimens bonded to glass-based ceramic than those of the indirect resin composite substrate, regardless of the storage period. The use of HFA etching as a surface pretreatment to glass-based ceramic substrate creates a honeycomb-like structure that provides additional micromechanical

retention,<sup>15,41</sup> and it could explain the higher MSBS values for specimens bonded to the glass-based ceramic. Apart from that, the control group also demonstrated significantly higher bond strength compared with ABU and SBU for lithium disilicate glass ceramic substrate after one year. The micromechanical retention created by the HFA etching pretreatment combined with the use of a separate bottle of silane agent might have enhanced the bond strength for lithium disilicate glass ceramic substrate in the present study and could explain the good results obtained for the glass-based ceramic in the control group for both storage periods.

SBU contains silane, HEMA, MDP, and Bis-GMA combined into a one-bottle solution. MDP-containing adhesives have been shown to provide a reliable bond to indirect restorative materials<sup>42,43</sup> and tooth substrates.<sup>23</sup> Thus, the presence of silane and MDP monomer was expected to contribute greatly to the bonding mechanism, by improving the wettability and forming cross-linkages with methacrylate groups.<sup>43</sup> However, in the current study, SBU showed similar MSBS values to separate bottles of silane and bonding resin for indirect resin composite substrate for both storage periods. Silane might be unstable when combined with MDP and Bis-GMA resins in a one-bottle solution.<sup>22</sup> Under acidic conditions, such as that in the presence of MDP and water, a self-condensation reaction might occur in the silanol groups of silane.<sup>22</sup> On the other hand, the presence of MDP and the retention created by air abrasion in the indirect resin composite substrate may have promoted chemical and micromechanical attachment, as SBU showed similar bond strength values to the control group for this substrate for both storage periods. Thus, the most common failure mode observed for this bonding material was cohesive within the indirect resin composite. Nevertheless, within the SBU group only, a significant decrease in MSBS was observed after one year of water storage for the indirect resin composite. A recent study reported that air abrasion may influence the durability of the SBU bond, and water could be absorbed into the primed layer as a consequence of aging and could decrease the bonding ability gained by micromechanical retention when air abrasion is performed.<sup>44</sup> Although that study included only the surface pretreatment method and its results were based on SBU bonded to air-abraded zirconia ceramic specimens,<sup>44</sup> we believe that a similar phenomenon may also have occurred in the indirect resin composite specimens in the current study. This could explain the decrease in MSBS



values within the SBU group for this substrate after the long-term storage.

On the other hand, in the present study, for SBU specimens bonded to the lithium disilicate glass ceramic substrate, HFA etching was used prior to SBU application. A recent study showed that HFA etching pretreatment of this substrate is beneficial for SBU bond performance; however, when SBU is used with or without additional silane application on lithium disilicate, a significantly higher bond strength is observed when silane is preapplied.<sup>45</sup> Therefore, this could reinforce the speculation that the silane included in one-bottle universal adhesive might not be fully effective for enhancing the chemical bonding itself. Moreover, incorporating Bis-GMA with the silane may also interfere in the silane-coupling condensation reaction with the hydroxyl groups of silica-based ceramic.<sup>46</sup> This could explain the lower MSBS values observed in the current study for SBU when compared with the control group for lithium disilicate glass ceramic for both storage periods. Nevertheless, the predominant fracture mode observed for this substrate was failure between the adhesive and lithium disilicate glass ceramic substrate, for 24 hours and one year of water storage, which indicates poor adhesion quality. In addition, no statistically significant differences were observed for SBU when both substrates were compared within the same storage period, suggesting that a similar bonding ability can be achieved regardless of the substrate or the surface pretreatment used.

ABU contains HEMA, MDP, and Bis-GMA. While MDP may improve bonding effectiveness to resin-based materials,<sup>47</sup> Bis-GMA has been reported to provide mechanical strength to adhesives by forming densely cross-linked polymers, lower polymerization shrinkage, and rapid hardening.<sup>37</sup> The combination of MDP and Bis-GMA could explain the higher initial MSBS values observed for ABU on the indirect resin composite substrate as compared with the control and SBU groups in the present study. However, water sorption can occur in HEMA-containing adhesives after long-term storage in water, causing hydrolytic degradation of the polymer, followed by elution of degradation products that results in a decrease of bond performance.<sup>48</sup> This is in agreement with the results obtained in the present study, as a statistically significant decrease in MSBS values was observed for ABU after one year of storage when compared with baseline for both substrates. However, these MSBS values did not differ from those of the SBU and control groups on

indirect resin composite after the long-term storage. In addition, all the materials used in the current study contained Bis-GMA in their formulation. Because of its high molecular weight, uncured Bis-GMA is highly viscous. This property might also have contributed to producing higher mechanical strength<sup>49</sup> and resulted in similar MSBS values for all of the materials on the indirect resin composite substrate. Moreover, the most common failure mode for both storage periods for ABU on the indirect resin composite substrate was cohesive failure in the indirect resin composite, similarly to the SBU and control groups.

For the lithium disilicate glass ceramic, ABU showed a similarly high level of bond strength to the control group after 24 hours of storage. However, a significant decrease in MSBS bond strength values was observed within the ABU group for glass-based ceramic substrate after long-term storage. Interestingly, for ABU only, the predominant failure mode was between the adhesive and the lithium disilicate glass ceramic for both storage times, suggesting that this interface was the weakest link of the bond. Although a significant decrease in bond strength was observed for ABU for both substrates after one year of storage, MSBS values were significantly lower for the lithium disilicate glass ceramic when compared with the indirect resin composite substrate. This might suggest that the total absence of silane in the ABU group compromised the long-term performance of this material, as compared with its high bond strength at baseline. This is true for both substrates, although it was more pronounced for the lithium disilicate glass ceramic.

Within the limitations of this study, after one year of water storage, MMAs showed comparable bond strength values to the control group for indirect resin composite; however, the use of separate bottles of silane and bonding resin in the control group resulted in durable bonding to the lithium disilicate glass ceramic. The use of a separate silane primer prior to the application of MMAs could be an alternative to improve their chemical bonding to lithium disilicate glass ceramic, as previously reported,<sup>45</sup> as well as to indirect resin composites. Ideally, an intermediate coupling agent should provide enhanced hydrolytic stability<sup>50</sup> and the capability to form strong bonds with different substrates at the same time. Therefore, further investigations are still required for surface pretreatment protocols and long-term interactions of this promising new category of adhesives for bonding to different restorative materials.

## CONCLUSIONS

MMA's can provide good bonding performance to sandblasted indirect resin composite after one year of water storage. However, the use of separate bottles of silane and bonding resin resulted in superior bond strength for etched glass-based ceramic substrate after long-term storage.

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## Conflict of Interest

The authors of this article certify that 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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