The effect of different surface treatments on bond strength between leucite reinforced feldspathic ceramic and composite resin

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ABSTRACT

Objectives: The aim of this study was to evaluate the effect of different surface treatments on the microtensile bond strength between a leucite reinforced glass–ceramic and composite resin.

Methods: Leucite reinforced ceramic blocks (n = 24) were constructed using the hot pressing technique. The blocks were assigned to 4 groups, which received the following surface treatments: G1: hydrofluoric (HF) acid and silane; G2: silane alone; G3: HF acid and silane then dried with warm air (100 °C); G4: silane alone then dried with warm air (100 °C). Unfilled resin was applied, followed by composite resin. Specimens were prepared and loaded in tension to determine the microtensile bond strength. Failure modes were classified by stereo- and scanning electron microscopy (SEM). Data was analysed using Kruskal–Wallis test followed by the Dunn’s multiple range test.

Results: The groups including the warm air step (G3 and G4) achieved a significantly stronger adhesion than G1 and G2. Pre-treatment of the surface with hydrofluoric acid in G1 resulted in significantly higher bond strength than G2. Failures were mostly cohesive in the luting resin for G1, G3 and G4 and mainly adhesive at the ceramic–resin interface in G2.

Conclusions: The method of application of silane to the ceramic surface can have a significant influence on the adhesion between the ceramic restoration and the resin cement. Enhancing the condensation reaction by drying the silane with a 100°C warm air stream significantly improved the microtensile bond strength, possibly eliminating the need for the hydrofluoric acid etching step.

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1. Introduction

Ceramic materials are among the most biocompatible materials developed for dental restorations. The combination of enamel etching with phosphoric acid and ceramic etching with hydrofluoric acid in the early 1980s, stimulated the development of resin-bonded ceramic restorations. More recent advancements in dental materials and improvements in fabrication of resin-bonded ceramic restorations provide real opportunities for achieving excellent aesthetics. However, these indirect restorations also have limitations. Possible signs of failure include de-bonding and fracture.
of the material, particularly related to cementation procedures.

Success with resin-bonded all-ceramic restorations is highly dependent on obtaining a reliable bond, which has to integrate all parts of the system into one coherent structure. The preferred manner of conditioning the fitting surface of the ceramic restoration is by etching with hydrofluoric acid, followed by the application of a silane coupling agent to achieve a high bond strength. Hydrofluoric acid works by creating surface pits via preferential dissolution of the glassy phase from the ceramic matrix. Treatment of the etched surface with silane increases the wettability and forms a covalent bond with both the ceramic and the resin cement. There are possibly three good reasons why it would be desirable to remove the hydrofluoric acid etching step from the procedure: (1) hydrofluoric acid is a highly toxic chemical, representing a potentially serious health hazard; (2) it has been reported that hydrofluoric acid etching of silica-based ceramics produces insoluble silica-fluoride salts, which can remain as by-products on the surface. If not removed, these by-products can interfere with the bond strength to the resin; (3) some glass–ceramics are currently being developed that have a very fine crystalline structure and may not benefit from hydrofluoric acid etching. Its elimination from the bonding procedure would be highly advantageous, but would only be possible if the silane bond can be shown to be adequate.

Since the 1940s, silane coupling agents have been used in industry to improve bonding between organic adhesives, ceramics and metals. But it was not until 1977 that Eames et al. suggested the use of a silane coupling agent for dental applications. The most commonly used silane in dentistry is 3-trimethoxysilylpropylmethacrylate (MPS) diluted in a water–ethanol solution. It is marketed in a pre-hydrolysed form (one bottle) or in a form where hydrolysis can occur by mixing silane and acid (two bottles). Both types of silane coupling agent are reported to perform well, even though atmospheric moisture is unfavourable to the prehydrolyzed silanes. It activates a condensation reaction that leads to polymerized siloxanes, producing oligomers, which give the solution a milky and opaque appearance.

It has been suggested that enhancing the condensation reaction of the silane could result in a significant improvement in the resin to ceramic bond. In this study, the effect of hot air treatment and silane, with and without the hydrofluoric acid etching stage, was investigated. The purpose of this study was to determine if post-silanization heat treatments can improve the microtensile bond strength of composite to ceramic to such a degree that hydrofluoric acid etching becomes redundant.

### Materials and methods

The materials employed are listed in Table 1. Twenty four 1 cm × 1 cm × 1 cm hot-pressed leucite reinforced ceramic blocks IPS Empress (Ivoclar Vivadent, Schaan, Liechtenstein) were fabricated using the lost wax technique and by pressure injection of ceramic ingots in the Empress 500 furnace (Ivoclar Vivadent, Schaan, Liechtenstein). The firing cycles followed the manufacturer’s recommendations. The specimens were ground flat for surface standardization with 400 grit followed by 600 grit wet silicon carbide paper and ultrasonically cleaned (Biosonic UC300, Whaledent, Altstätten, Switzerland) in distilled water for 5 min to remove any surface contaminants due to the silicon carbide papers. The 24 blocks were assigned at random to four groups, which received the following surface treatments:

- G1: hydrofluoric acid solution (Ultradent Porcelain Etch 9.5% Buffered, Ultradent Products, South Jordan, UT) was applied for 1 min, rinsed for 1 min, air-dried for 1 min, followed by the application of the silane coupling agent (Monobond S, Ivoclar Vivadent, Schaan, Liechtenstein) for 1 min and then air-dried for 30 s.
- G2: silane coupling agent was applied for 1 min and air-dried with compressed oil-free air for 30 s.
- G3: hydrofluoric acid solution was applied for 1 min, rinsed for 1 min, air-dried for 1 min, followed by silane coupling agent applied for 1 min, and then dried with a warm stream of air (100 °C) for 1 min.
- G4: silane coupling agent was applied for 1 min and dried with a warm stream of air (100 °C) for 1 min.

A miniature custom-made blow dryer delivering a constant flow of warm air, approximately 100 °C at 1 cm from the nozzle was utilized in the study. In all groups, following the appropriate surface treatment, a resin bonding agent (D/E Resin, Bisco, Schaumburg, IL, USA) was applied and light cured for 20 s at 600 mW/cm² (VIP, Bisco, Schaumburg, IL, USA). Following application of the adhesive, composite resin (Tetric EvoCeram, Ivoclar Vivadent, Schaan, Liechtenstein) was applied in 2 mm incremental layers, light curing each layer for 20 s, to create a 0.8 cm block. Luting procedures were carried out under a constant load of 1 kg (1249 MPa) to standardize the exerted pressure. These bonding procedures took place on a number of different occasions.

Using a low speed cutting saw (Isomet, Buehler, Lake Bluff, IL, USA), each block was longitudinally cut into a series of 1 mm thick slabs. The ceramic-composite block was then

| Table 1 – Composite resins and bonding agents used in this study. |
|-------------------|------------------|---------------------------------|
| Name              | Manufacturer      | Composition                      |
| Monobond-S (lot no. 8177) | Ivoclar Vivadent, Schaan, Liechtenstein | 3-MPS 1%, ethanol/water-based solvent, acetic acid |
| D/E Resin (lot no. 0400009669) | Bisco, Schaumburg, IL, USA | Bisphenol-A-diglycidylmethacrylate, urethane-dimethacrylate, hydroxyethyl methacrylate, Bis-GMA, ethoxylated Bis-EMA, urethane-dimethacrylate, barium glass, ytterbium trifluoride, mixed oxides, copolymers |
| Tetric EvoCeram (lot no. 555945) | Ivoclar Vivadent, Schaan, Liechtenstein | |
rotated 90° and the procedure repeated. Twelve to fourteen bars approximately 0.8 mm² in cross-section were obtained from each block.

For microtensile bond strength testing, each selected specimen was glued with cyanoacrylate (Zapit, Dental Ventures of America, CA, USA) to the Gerardeli’s jig designed to transmit purely tensile forces when mounted on a universal loading machine (Controls, Milano, Italy). The tensile load was applied at a crosshead speed of 0.5 mm/min, until failure. The load at failure in Newtons was recorded, and the specimen’s fragments cautiously removed from the fixtures with a scalpel blade. The cross-sectional area at the site of fracture was measured to the nearest 0.01 mm with a digital calliper in order to calculate the bond strength at failure in MegaPascals.

Failure modes were recorded as adhesive or cohesive and classified by stereomicroscopy at 20× magnifications (Bausch & Lomb, Rochester, USA) and scanning electron microscopy (SEM) at 200× magnifications (Philips 515, Philips, Eindhoven, Netherlands). Representative specimens from each group were rinsed with 96% ethanol, mounted on metallic stubs and gold-sputter coated (Emitech k550, Emitech, UK) prior to SEM. The cross-sectional area of those microbars was observed to assess the morphology of the debonded interfaces.

As the data was not normally distributed, Kruskal–Wallis analysis of variance on ranks followed by the Dunn’s multiple range test was applied in order to assess any statistical significance of differences among the groups. In the analyses the level of statistical significance was set at α = 0.05.

3. Results

The data for the number of pre-test failures, the number of tested specimens, the mean microtensile bond strength, standard deviation and the statistical difference are shown in Table 2 and illustrated in Fig. 1.

The mean microtensile bond strength and standard deviation in MPa were G1 = 22.8(±4.3), G2 = 18.7(±5.8), G3 = 27.8(±4.7) and G4 = 28.5(±5.7).

The Kruskal–Wallis analysis demonstrated that significant differences in microtensile bond strength existed among the experimental groups (p < 0.001). The table reports the statistically significant differences according to the Dunn’s multiple range test for post hoc comparisons. Statistically similar groups are labelled with the same letters.

G3 and G4, which employed the heat treatment step, showed the highest bond strengths, which were significantly higher than those obtained for the groups excluding heat treatment (p < 0.001). Moreover, G1 achieved significantly higher bond strengths than G2 (p < 0.05). The microscopic examination of the fracture sites showed that failures were mostly cohesive failures in the luting resin in G1, G3 and G4, and mainly adhesive (ceramic–luting resin interface) in G2.

4. Discussion

When a resin-bonded ceramic restoration is placed, many factors can play a part in the resin bond with tooth structure. Two interfaces need to be considered: the dentin–resin interface and the ceramic–resin interface. The bond strength at these interfaces has to be optimised, as a weak interface can contribute to failure of the restoration.25 Thus, a stable and durable bond between the dental materials and tooth substrate is imperative for success.26

Various methods are available for assessment of the bond strength. The shear bond test is a common testing method but often produces cohesive bulk fracture within the substrate rather than the interface due to the generation of complex stress distribution during testing and may lead to erroneous data interpretation.27,28 In this study, the bond strength at the ceramic–resin interface was assessed using the non-trimming

Table 2 – Number of pre-test failures, number of tested specimens, mean microtensile bond strength, standard deviation and statistical difference. In the significance column, the same letters represents statistically similar groups.

<table>
<thead>
<tr>
<th>Group</th>
<th>No. of pre-test failures (%)</th>
<th>No. of tested specimens</th>
<th>Mean (MPa)</th>
<th>SD</th>
<th>Significance (p &lt; 0.05)</th>
<th>Number of cohesive failures in luting resin</th>
<th>Number of adhesive failures (ceramic–luting resin interface)</th>
</tr>
</thead>
<tbody>
<tr>
<td>G1</td>
<td>Hydrofluoric acid–silane</td>
<td>0 (0%)</td>
<td>48</td>
<td>22.8</td>
<td>A</td>
<td>47 (97.9%)</td>
<td>1 (2.1%)</td>
</tr>
<tr>
<td>G2</td>
<td>Silane</td>
<td>27 (56%)</td>
<td>22</td>
<td>18.7</td>
<td>B</td>
<td>1 (4.8%)</td>
<td>20 (95.2%)</td>
</tr>
<tr>
<td>G3</td>
<td>Hydrofluoric acid–silane–heat</td>
<td>0 (0%)</td>
<td>48</td>
<td>27.8</td>
<td>C</td>
<td>48 (100%)</td>
<td>0 (0%)</td>
</tr>
<tr>
<td>G4</td>
<td>Silane–heat</td>
<td>0 (0%)</td>
<td>48</td>
<td>28.5</td>
<td>C</td>
<td>47 (97.9%)</td>
<td>1 (2.1%)</td>
</tr>
</tbody>
</table>
method of microtensile bond testing, which was introduced by Sano in 1994 in an attempt to eliminate the non-uniform stress distribution within the adhesive zone. This technique allows the loading stress to be more uniformly distributed by the testing of small-sized specimens and failure predominantly occurs at the adhesive interface. Moreover, with this variant of the method, multiple specimens can be obtained from a single block, and the variance associated with testing is usually lowered to 10–25%, providing results that are more consistent. The microtensile bond strength test was preferred to shear bond strength test because for the latter, even though more frequently used, it is not possible to determine the actual stress generated at the interface. On the downside, the microtensile bond strength test procedure is highly technique sensitive by the nature of the specimen preparation, which can result in premature failure of the specimen. However, the methodology for microtensile bond strength testing has been controversial of late regarding the number of blocks and microbars used as well as pre-test failures. In addition, microscopic analysis of the fractured surfaces should be undertaken to provide a more consistent and complete description of the fracture process and modes of failure. Emphasis can then be placed on strengthening the weakest component in the chain until failure shifts to another part of the adhesive complex.

Using a combination of hydrofluoric acid etching and conventional silane treatment produced a microtensile bond strength of 22.8 ± 4.3 MPa (G1), which was significantly higher than that of the silane bond without hydrofluoric acid etching, exhibiting a bond strength of 18.5 ± 5.8 MPa (G2). The mode of failure in G1 was predominantly adhesive, involving the ceramic–resin interface, suggesting that this interface is the weak link in the adhesive system. High incidences of pre-test failures were recorded for G2, probably due to the fragile nature of the specimens combined with poor bond strength. The addition of a heat treatment in the drying step of the silane application procedure resulted in a significant improvement in the microtensile bond strength to 27.8 ± 4.6 MPa (G3). An interesting finding was that in the absence of hydrofluoric acid etching, the highest bond strength was achieved (G4: 28.5 ± 5.8 MPa). For G1, G3 and G4, the mode of failure was distinctively different from that of G2. The majority of failures occurred as cohesive fractures of the resin. This represents the optimum that can be achieved in bonding with the resins used, since the only way the bond strength could be improved would be to increase the cohesive strength of the resins themselves.

Monticelli et al. demonstrated that drying with warm air at 38 °C increased the effectiveness of silane coupling agents when bonding ceramic to composite resin. Another study reported that heat treatment at 100 °C for 60 s following surface-roughening treatment caused a significant improvement in shear bond strength. Similarly, Shen et al. found that silane drying with a warm air stream at 45 °C enhanced the microtensile bond strength of a leucite reinforced glass–ceramic. These studies achieved the highest bond strengths when warm air was used in conjunction with another surface treatment such as hydrofluoric acid etching and abrasion. However, in this study, it was found that the highest bond strengths were achieved without hydrofluoric acid etching. When silane is applied to the ceramic surface, three structural layers form in the coupling agent with varying properties. Heat treatment at 100 °C consolidates these layers into a monolayer and increases the bond strength of the composite resin to the ceramic. It has been proposed that heat treatment of the silanated ceramic surface can improve the bond strength by eliminating water, alcohol and other by-products and complete condensation reactions by promoting siloxane bond formation. This study utilized a customized air blower delivering a constant flow of warm air at 100 °C at 1 cm distance, which may aid this elimination and increase the number of bond sites available for the reaction. Removal of water drives the silane–silica surface condensation reaction towards completion and promotes covalent silane–silica bond formation. Therefore, the heat treatment may enhance the chemical bond to the ceramic as well as within the silane compound. This may explain why enhanced microtensile bond strengths were found when warm silane was used both with and without the hydrofluoric acid etching stage. Although heat treatment may not be feasible for intra-oral repair, it could be used in the cementation stage and thereby remove the dangerous hydrofluoric acid etching stage from the procedure.

5. Conclusions

Enhancing the condensation reaction by including a drying step in the application of the silane with a 100 °C warm air stream significantly improved the ceramic to resin microtensile bond strength. Within the limitations of this study, it was found that silane plus heat treatment, but without hydrofluoric acid etching, achieved the highest microtensile bond strengths. However, further work is required to determine if similar results are produced with different glass–ceramics. In summary, modifications to the silane treatment can increase the microtensile bond strength between composite resins and leucite reinforced feldspathic ceramic. This may avoid the usage of hydrofluoric acid in the construction and possibly also the repair of ceramic restorations.

References


