

Original Research Article

Effect of water storage and hydrophobic adhesive layer application on the bond strength of all-in-one adhesives

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Received for publication: February 23, 2013. Accepted for publication: April 26, 2013.

Keywords: dentin-bonding agents; dental bonding; dentin.

Abstract

Introduction: To prevent the rate of water absorption and degradation of exposed collagen and the resin matrix on the hybrid layers, the use of an additional layer of hydrophobic resin on all-in-one adhesives is one of the approaches to improve the bond strength and infiltration of monomers in demineralized dentin. **Objectives:** To compare the microshear bond strength of different self-etching adhesive systems, and to evaluate the effect of the application of a hydrophobic adhesive layer on all-in-one adhesive systems after a storage period of 48 h and 30 days in distilled water at 37°C. **Material and methods:** Bovine incisor crowns were polished to expose flat dentin surfaces. The crowns were randomly distributed into 14 groups (n = 12) according to the adhesive system [Clearfil SE Bond (CSEB), AdheSE (ASE), Adper Scotchbond SE (SSE), Adper Easy Bond (EB), and Go! (GO)], and storage time. In two groups (indicated as GO+B and EB+B), a layer of a hydrophobic adhesive was applied on all-in-one adhesive systems. After 48h and 30 days

of storage in distilled water at 37°C, microshear bond strength was determined. The data were statistically analyzed by ANOVA and Tukey's test ($\alpha = 5\%$). **Results:** After 48h, EB, EB+B, CSEB, and GO+B showed higher bond strength values. The application of a hydrophobic layer did not influence EB and increased GO bond strength values. After 30 days, CSEB, SSE, and EB+B showed the highest bond strength values. Comparing groups of all-in-one adhesives with and without a hydrophobic adhesive-resin layer, the bond strength values showed no significant difference. **Conclusion:** The application of a hydrophobic adhesive-resin layer increased bond strength values only at 48h. With respect to bond degradation over time, only EB showed a statistically significant decrease of bond strength after 30 days.

Introduction

Currently, the etch-and-rinse and self-etching approaches are the two strategies of resin-based adhesive systems to accomplish the bonding to enamel and dentin. Despite the relative ease of application and less-sensitive technique related to the application of simplified self-etching adhesive systems, their bond strength is not yet considered effective in enamel or dentin. Many authors reported lower bond strength values for all-in-one adhesives when compared to two-step self-etching adhesives [2, 3, 19, 26, 29]. Moreover, many problems are associated with their use. These adhesives, even when polymerized to form adequately the hybrid layer, are porous structures that can act as semipermeable membranes, allowing the bi-directional flow of water through the adhesive layer if it is not coated with a hydrophobic adhesive-resin layer [27].

The literature also reported a high amount of nanoleakage when these adhesive systems were used [28] and a greater decrease in bond strength over time when compared with conventional adhesives such as those in which a layer of hydrophobic adhesive-resin was applied [3]. There is also evidence that small nanoscale defects in the adhesive layer seen in nano infiltration tests can act as channels for water passage [28].

The degradation of the hybrid layer may be due to a number of factors. Among these, the most important is the incomplete penetration and infiltration of monomers in the demineralized dentin [13], the heterogeneous distribution of monomers through the hybrid layer,[11] the inadequate or insufficient polymerization [14, 21], and the degradation and hydrolysis of both the resin component and the non-hybridized exposed collagen [5].

Different clinical approaches have been proposed to improve the bond strength and infiltration of

monomers in demineralized dentin in order to reduce the rate of water absorption and degradation of exposed collagen and the resin matrix [5]. Among the most commonly described approaches are the use of an additional layer of hydrophobic resin [23, 24] the application of multiple layers of simplified adhesives,[12, 16] enhanced solvent evaporation [12], increased polymerization time [6], and the use of MMPs (matrix metalloproteinases) inhibitors [7].

The objectives of the present study were to compare the microshear bond strength of different self-etching adhesive systems and to evaluate the effect of the application of a hydrophobic adhesive layer on all-in-one adhesive systems after a storage period of 48 h and 30 days in distilled water at 37°C.

Material and methods

Bovine incisors were selected, cleaned, and stored into 0.5% chloramine T solution, under refrigeration, until the preparation of the specimens. The crowns were separated and their buccal surfaces polished under constant irrigation to expose flat dentin surfaces.

Each crown was embedded in a PVC cylinder with acrylic resin (Jet, Artigos Odontológicos Clássico Ltd, Sao Paulo, Brazil) and randomly divided into 14 groups (n = 12), according to the adhesive system and storage time. A standard smear layer was prepared on the dentin surfaces using 600-grit SiC paper for 1 min under water irrigation. The self-etching adhesives [Clearfil SE Bond (CSEB), AdheSE (ASE), Adper Scotchbond SE (SSE), Adper Easy Bond (EB), and Go! (GO)] were applied according to each manufacturer's recommendations. The adhesive systems and their form of application are shown in table I. In two additional groups, all-in-one adhesives received an additional layer of hydrophobic resin (Scotchbond Multi Purpose, 3M ESPE) (EB+B and GO+B).

Table I – Bonding procedure for the adhesive systems investigated

Type	Code	Adhesive	Bonding procedure
Two-step self-etching adhesive	CSEB	Clearfil SE Bond (Kuraray, Osaka, Japan)	1. Apply primer for 20 s 2. Dry with mild air stream 3. Apply bond 4. Dry with gentle air stream 5. Light cure for 10 s
	ASE	AdheSE (Ivoclar Vivadent, Amherst, NY, USA)	1. Apply primer for 30 s 2. Gently air dry 3. Apply bond 4. Gently air dry 5. Light cure for 10 s
	SSE	Adper Scotchbond SE (Adper SE Plus, 3M ESPE, St. Paul, MN, USA)	1. Apply liquid A so that a continuous red-colored layer is obtained on the surface 2. Apply liquid B into the entire wetted surface of the bonding area for 20 s. The red color will disappear. 3. Thoroughly air dry for 10 s 4. Apply a second coat of liquid B to the entire surface 5. Lightly air dry for 10s 6. Light cure for 10 s
All-in-one self-etching adhesive	EB	Adper Easy Bond (Adper Easy One, 3M ESPE)	1. Apply adhesive for 20 s 2. Gently air dry until liquid does not move anymore 3. Light cure for 10 s
	GO	Go! (SDI, Victoria, Australia)	1. Remove excess water 2. Apply adhesive on the surface and leave in place for 20 s 2. Air dry for 5 s leaving a glossy surface 3. Light cure for 10 s
	B	Hydrophobic adhesive (Scotchbond Multi Purpose – 3M ESPE)	1. Apply one coat after all-in-one adhesives EO e GO 2. Light cure for 15 s

Transparent cylindrical matrixes with an internal diameter of 0.7 mm and height of 1 mm (Tygon tubing R-3603, Saint-Gobain Performance Plastics, Maime Lakes, FL, USA) were positioned onto the dentin surface treated with the described adhesives. A composite resin (shade A2, Amelogen Plus, Ultradent Products Inc., South Jordan, UT USA) was carefully inserted into each matrix and light-cured for 20 s at 1000 mW/cm² (LED curing Bluephase, Ivoclar Vivadent, Schann, Lietchenstein). The specimens were stored into distilled water at 37°C for two different periods: 48 h (early bond strength) and 30 days (delayed bond strength).

The microshear bond strength tests were performed with a universal testing machine (Kratos, São Paulo, SP, Brazil) at a cross-head speed of 0.5 mm/min until fracture. The specimens were carefully aligned to allow that the load be applied

as close as possible to the bonding interface at the cylinder bases with aid of a stainless steel wire-loop (0.2 mm diameter).

After the microshear bond strength tests, the fractured interfaces were examined in a light microscope at x57 magnification (SZX9, Olympus, Tokyo, Japan) to determine the failure mode, classified as adhesive, cohesive, or mixed.

Data were statistically analyzed using ANOVA and Tukey's test with a significance level of 5%.

Results

The results for the microshear bond strength to dentin determined for the adhesives tested as a function of storage time in distilled water at 37°C are shown in table II.

Table II - Means, standard deviations, and coefficients of variation for microshear bond strength (MPa) to dentin

Group	Microshear bond strength to dentin (MPa)	
	48 h	30 days
CSEB	27.13±6.32 (23%) ^{ab}	28.61±6.15 (21%) ^a
ASE	14.73±3.81 (26%) ^e	17.85±3.33 (19%) ^{cde}
SSE	21.22±4.72 (22%) ^{bcde}	23.48±6.05 (26%) ^{abc}
EB	28.70±6.27 (22%) ^a	20.28±4.74 (23%) ^{cde}
EB+B	27.55±6.06 (22%) ^{ab}	22.51±4.80 (21%) ^{abcd}
GO	15.55±3.71 (24%) ^{de}	16.18±3.37 (21%) ^{de}
GO+B	23.16±6.68 (29%) ^{abc}	17.18±3.76 (22%) ^{cde}

Values followed by same superscript letters are statistically similar ($p > 0.05$)

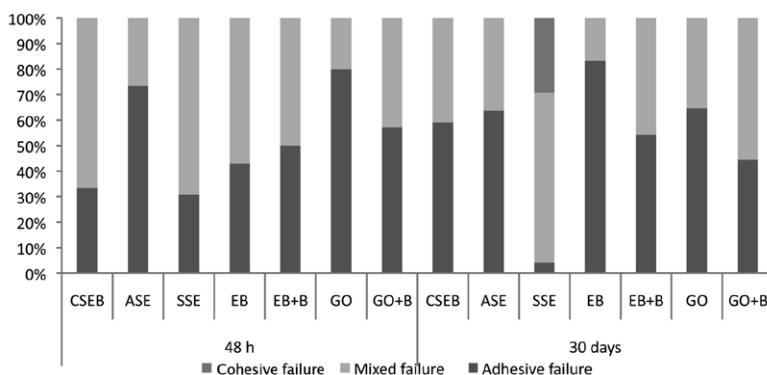
For the 48 h storage period, EB showed the higher bond strength values, statistically similar to EB+B, CSEB, and GO+B. On the other hand, ASE, GO and SSE showed lower microshear bond strengths. It can also be noted that all-in-one and two-step self-etching adhesives behaved differently. Regarding the two-step self-etching adhesives, CSEB showed higher bond strength values, while ASE showed lower values. As for the all-in-one adhesives, EB showed the highest bond strengths, while GO presented one of the lowest values. When comparing groups of all-in-one adhesives applied according to manufacturers' recommendation to those in which a layer of hydrophobic adhesive was applied, the bond strength values did not change or increase.

For the 30-day storage period, CSEB showed the higher bond strength values, statistically similar to those of groups SSE and EB+B. Conversely, GO, GO+B, ASE, EB, and EB+B presented lower microshear bond strength values. Again, for the two-step self-etching adhesives, CSEB demonstrated higher bond strength values, while ASE demonstrated lower values. As for the all-in-one adhesives, EB

showed higher bond strength values, while GO presented one of the lowest values. Comparing the groups of all-in-one adhesives applied according to manufacturers' recommendation to those in which a layer of hydrophobic adhesive was applied, the bond strength values showed no statistically significant difference for both EB and GO.

Regarding the bond strength degradation over time, it can be observed that only EB showed a statistically significant decrease in the bond strength values after 30 days of storage in distilled water. Even if a statistical difference could not be noted for the two other two groups (EB+B and GO+B), a decreasing trend in the bond strength values was observed. On the other hand, it was also possible to notice that some groups (CSEB, ASE, SSE, and GO) presented a tendency towards increasing the bond strength values over time, even though no statistically significant difference was observed.

The results of failure analysis performed after the microshear bond strength tests are shown in figure 1. All groups showed predominantly adhesive and mixed failures. Only SSE after 30 days storage showed cohesive failure in resin (29.2%).

**Figure 1** - Frequency of failure mode

Discussion

The results of the present study indicated that the all-in-one and two-step self-etching adhesives behaved differently. Other studies obtained similar results, indicating that the all-in-one adhesives presented lower bond strength values when compared to two-step self-etching adhesives [2, 3, 26]. One possible explanation for this fact would be that, as all-in-one adhesives can create thin layers [16], their polymerization can be diminished by the presence of oxygen [25].

Clearfil SE Bond showed high bond strength values in various studies and is considered the gold standard when comparing different self-etching adhesive systems [2, 19, 26]. The good performance of this adhesive appears to be related to some factors. Its pH of ≈ 2.0 allows a lower dissolution of smear plugs while maintaining hydroxyapatite crystals [30] and facilitates the penetration and polymerization of monomers in the underlying dentin to form an adequate hybrid layer. Also, the presence of photoinitiators in both components (acidic primer and adhesive) increases the efficiency of monomer polymerization and facilitates the solvent evaporation [22]. It has also been suggested that the high hydrolytic stability of MDP and its chemical interaction with the dental tissues can contribute to a long-term durability of the bond strength [15].

Also a two-step self-etching adhesive, AdheSE showed lower microshear bond strength values when compared to Clearfil SE Bond. This could be possibly explained because its self-etching capacity is based on the phosphonic acid acrylates. These monomers have a pH of ≈ 1.4 [1] with greater capacity to dissolve the smear layer, smear plugs, and weak bonds to dentin. Their highly hydrophilic properties also contribute to their behavior as a semipermeable membrane, [8] leading to the degradation of the resin-dentin bonds when stored in water [1].

Among the two-step self-etching adhesives, Adper Scotchbond SE is noteworthy. Its composition and especially the fact that aqueous primer alone does not etch the tooth tissues led to controversies regarding the classification of this adhesive, but it can be categorized as a two-step self-etching system since the second application of the adhesive can be considered equivalent to the application of the hydrophobic resin in traditional two-step self-etching adhesives [19].

The results of some *in vitro* studies indicate that the use of a hydrophobic layer on all-in-one adhesives could maintain or improve their

performance [4, 10, 20]. In the present study, corroborating the aforementioned results, the use of an additional layer of hydrophobic resin also indicated good results. Reis *et al.* [23] showed that the use of a hydrophobic adhesive layer prevented the decrease in bond strength of three all-in-one adhesives after six months of water storage. The use of a hydrophobic resin layer on single-step self-etching adhesives was also tested in a randomized clinical trial of 18 months in non-cariou cervical lesions [24]. The results demonstrated that the retention rates of restorations after 18 months were significantly higher for groups in which a layer of hydrophobic adhesive was applied.

There are several possible explanations for this increase in bond strength. This layer of hydrophobic resin seems to limit the diffusion of water through the hybrid layer, which could have happened relatively quickly without the presence of this hydrophobic layer [27], inhibiting polymerization and weakening the adhesive-composite interface. This additional layer may also have decreased the removal of non-polymerized monomers and oligomers of the hybrid layer. Zones of hydrophilic low-polymerized phases that allow the movement of water have been identified in the hybrid layer of self-etching adhesives [28]. In both cases, this additional hydrophobic layer would enable all-in-one adhesives to simulate two-step self-etching adhesives, increasing the thickness of the adhesive layer [4], which could reduce the polymerization stress [9] and improve the stress distribution during the tests. These two factors may have contributed to the higher values of bond strength observed in the groups with the presence of an additional layer of hydrophobic resin.

Considering the single-step self-etching adhesives, the good performance of EB, comparable to CSEB at 48 h with and without the additional layer of hydrophobic adhesive, can be explained by its pH around 2.4, similar to the performance of CSEB (≈ 2.0).

Another important concern is the degradation of the adhesive layer over time. This degradation is the result of the interaction of polymers with water in dentinal fluid and saliva, which has a plasticizing effect and separates the polymer chains and reduces the mechanical properties of the material [1, 17, 18]. In the present study, after 30 days of storage in distilled water at 37°C, a decrease of the bond strength values occurred for the adhesives tested. However, only group EB showed a statistically significant decrease. In the EB+B group, with application of a hydrophobic

adhesive layer, the bond strength values after 30 days of storage were statistically similar to those for two-step adhesives. This demonstrates that the hydrophobic adhesive layer was effective in decreasing the high permeability attributed to all-in-one adhesives. However, it must be noted that the GO+B group obtained lower values of bond strength. These values were not statistically different from those observed for the group without the hydrophobic adhesive layer.

The performance of the adhesive systems in terms of bond strength changed over time, indicating that the results of bond strength tests in the short term may not accurately reflect their behavior in the long term, in particular for dentin. However, it should be noted that the storage period of 30 days, used in this study, can still be considered short for this type of test and that further studies are needed to evaluate the bond strength of these and other self-etching adhesives systems with longer storage periods.

Conclusion

It can be concluded that, for the storage period of 48 h, the application of a hydrophobic adhesive layer on the single-step self-etching adhesives led to an increase in the bond strength values only for GO group, which presented a bond strength value similar to that of CSEB. For the storage time of 30 days, the bond strength values showed no significant difference for both EB and GO when a hydrophobic adhesive layer was applied. With respect to the degradation of strength over time, only EB showed a statistically significant decrease of bond strength after 30 days.

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