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Preheating Impact on the Degree of Conversion and Water Sorption/ Solubility of Selected Single-bottle Adhesive Systems

MRL Vale • FAC Afonso • BCD Borges
AC Freitas Jr • A Farias-Neto • EO Almeida
EJ Souza-Junior • S Geraldeli

Clinical Relevance

The use of preheated ethanol/water-based adhesive systems seems to facilitate solvent evaporation, which translates to improving the degree of conversion and decreasing water sorption/solubility.

SUMMARY

Objective: This study evaluated the degree of conversion (DC) and the water sorption/solubility of preheated single-bottle adhesive systems.

Methods and Materials: Five adhesive systems were tested: Adper Easy One and Adper Single Bond 2 (3M ESPE), Excite and Tetric N-Bond (Ivoclar/Vivadent), and XP Bond (Dentsply/Caulk). After storage for two hours at 25°C or 60°C, 50 samples (n=5) were prepared for all adhesive systems and stored dry in lightproof containers at 37°C for 24 hours. Fourier transform infrared/attenuated total reflectance spectroscopy was used to evaluate the DC, and water sorption/solubility was measured by means of mass loss and gain after water storage. The data were analyzed by two-way analysis of variance followed by Tukey's test ($p<0.05$).

Results: Preheated adhesive systems showed statistically significantly higher DC than those kept at 25°C. Except for XP Bond, preheated adhesive systems presented statistically sig-

Mariana R. L. Vale, undergraduate, Potiguar University (Laureate International Universities), Department of Dentistry, Natal, Brazil

Felipe A. C. Afonso, undergraduate, Potiguar University (Laureate International Universities), Department of Dentistry, Natal, Brazil

*Boniek C. D. Borges, PhD, Federal University of Rio Grande do Norte, Department of Dentistry, Natal, Brazil

Amilcar Chagas Freitas Jr, PhD, Potiguar University (Laureate International Universities), Department of Dentistry, Natal, Brazil

Arcelino Farias-Neto, PhD, Potiguar University (Laureate International Universities), Department of Dentistry, Natal, Brazil

Erika Oliveira Almeida, PhD, Federal University of Rio Grande do Norte, Department of Dentistry, Natal, Brazil

Eduardo José Souza-Junior, DDS, MS, Piracicaba Dental School, State University of Campinas – UNICAMP, Department of Restorative Dentistry, Piracicaba, Brazil

Saulo Geraldeli, PhD, University of Florida, Department of Operative Dentistry, Gainesville, FL, USA

*Corresponding author: Av. Senador Salgado Filho 1787, Natal, 59.056-000, Brazil; e-mail: boniek.castillo@gmail.com

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nificantly lower water sorption/solubility means.

Conclusions: Preheating improved the DC for all tested adhesive systems. Also, it promoted a decrease of water sorption/solubility, except for the XP Bond adhesive system.

INTRODUCTION

The popular expectation for esthetics in dentistry continues to promote the use of direct composite resins for anterior and posterior teeth, which requires the application of an adhesive system to bond to dental structures. Contemporary adhesive systems are referred to as "etch-and-rinse" or "self-etch" (etch-and-dry). These two categories are either multistep (three-step etch-and-rinse and two-step self-etch) or simplified by combining the number of steps required for the clinical application (two-step etch-and-rinse and one-step self-etch).[1] Most clinicians prefer such a combined-process simplification because the reduced number of steps in the adhesive process reduces clinical chair time for the patient.[2,3]

Successful adhesion to enamel and dentin tissue is a fundamental necessity for placement of dental materials, and that requirement is directly dependent on the quality of the dentin hybrid layer. A high degree of conversion (DC) and low water sorption/solubility of the adhesive system is fundamental to improving resistance of material degradation under *in vivo* clinical conditions. In fact, low DC of dental adhesives is associated with high water sorption/solubility as well as low bond strength values, low mechanical properties, increased permeability, and even the occurrence of phase separation.[4-6] Moreover, reduced DC can also account for continuous etching of the tooth substrate due to suboptimally polymerized acidic monomer in self-etch adhesives.[7]

A recently proposed method to increase the DC of resin-based dental materials is preheating before photoactivation.[8] Heating the material to high temperatures decreases viscosity and increases radical mobility, which favors higher DC.[8] It has been shown that preheating some single-bottle adhesive systems can improve the DC.[9]

To date, several commercially available adhesive systems have not been evaluated with regard to the effect on the DC and water sorption/solubility when the adhesive is preheated before application. Therefore, this study aimed to analyze and to compare the DC and the water sorption/solubility of preheated and non-preheated single-bottle adhesive systems. The null hypotheses were that 1) preheating would not affect the DC of the adhesive systems, and 2) preheating would not affect the water sorption/solubility of adhesive systems.

METHODS AND MATERIALS

The single-bottle contemporary adhesive systems used in this study were Adper Easy One (3M ESPE, St Paul, MN, USA), Adper Single Bond 2 (3M ESPE), Excite (Ivoclar-Vivadent, Schaan, Liechtenstein), Tetric N-Bond (Ivoclar Vivadent), and XP Bond (Dentsply/Caulk, Milford, DE, USA). The DC and sorption/solubility were evaluated for each adhesive system at different temperatures (25°C = room temperature; 60°C = preheated) (n=5). The specific compositions, manufacturers, and batch numbers of the tested materials are shown in Table 1.

Preheating of the Adhesive Systems

Each bottle of the adhesive system was kept in an oven (incubator) at 25°C or 60°C for two hours before starting the adhesive procedure.[9] Before specimen preparation, the temperature was checked with a thermometer for each adhesive system.

Specimen Preparation

For DC analysis, 50 bar-shaped samples (6 mm long × 1 mm deep × 1 mm wide) were prepared using each adhesive system and polyvinyl siloxane molds. For water sorption/solubility analyses, 50 disks (6-mm diameter by 1-mm thickness) were prepared using cylindrical polyvinyl siloxane molds. One drop of each adhesive was placed into the mold followed by solvent evaporation for 10 seconds using an uncontaminated air stream. Then, a Mylar strip was placed over the mold, and the adhesive was photoactivated for 20 seconds with a light-emitting diode (LED) unit (Coltolux, Coltène/Whaledent, Allstätten, Switzerland). The 8-mm-diameter tip of the LED covered the entire sample during photoactivation. An irradiance of 1264 mW/cm^2 was distributed over the top surface of each sample. After photoactivation, the sample was removed from the mold and stored dry in lightproof containers at 37°C for 24 hours.

DC Analysis

The DC was analyzed using Fourier transform infrared/attenuated total reflectance spectroscopy (Spectrum 100, PerkinElmer, Shelton, CT, USA) at 24°C under 64% relative humidity.[3] The absorption spectra of nonpolymerized and polymerized adhesives were obtained from the wavenumber region

Table 1: *Composition, Manufacturer, and Lots of the Adhesive Systems Used in the Study*

Adhesive System	Composition (% by Weight)	Manufacturer	Lot No.
Adper Easy One	Bis-GMA (15-25), HEMA (15-25), ethanol (10-15), water (10-15), phosphoric acid-6-methacryloxy-hexylesters (5-15), silane treated silica (8-12), 1,6-hexanediol dimethacrylate (5-10), copolymer of acrylic and itaconic acid (1-5), (dimethylamino)ethyl methacrylate (1-5), camphorquinone (1-3), 2,4,6- trimethylbenzoyldiphenylphosphine oxide (1-3)	3M ESPE, St Paul, MN, USA	436286
Excite	Phosphonic acid acrylate (<11), HEMA (<15), dimethacrylates (<53), alcohol (<20)	Ivoclar-Vivadent, Schaan, Liechtenstein	N11163
Adper Single Bond 2	Ethyl alcohol (25-30), silane treated silica (nanofiller) (10-20), bis-GMA (10-20), HEMA (5-10), glycerol 1,3-dimethacrylate (5-10), copolymer of acrylic and itaconic acids (5-10), water (5), diurethane dimethacrylate (1-5)	3M ESPE, St Paul, MN, USA	N300770BR
Tetric N-Bond	Phosphonic acid acrylate (<11), HEMA (<15), dimethacrylates (<53), alcohol (<20)	Ivoclar-Vivadent, Schaan, Liechtenstein	N76256
XP Bond	HEMA (25-50), methacrylate (10-25), tert-butyl alcohol (10-25), acrylates (10-25)	Dentsply/Caulk, Milford, DE, USA	1011000650

between 4000 cm^{-1} and 650 cm^{-1}, with 32 scans at 4 cm^{-1}. The DC (%) was calculated using the following equation:

$$DC(\%) = 100 \times (1 - [\text{R polymerized}/\text{R nonpolymerized}]) \quad (1)$$

where R represents the ratio between the absorbance peaks at 1638 and 1608 cm^{-1}. The final DC values were analyzed using two-way analysis of variance (ANOVA) considering adhesive system × temperature and Tukey's test ($p<0.05$).

Water Sorption and Solubility Measurement

Water sorption and solubility were determined according to the ISO specification 4049, except for specimen dimensions. The disks were stored at 37°C in desiccators that contained silica gel desiccant and weighed daily (24-hour intervals) on an analytical balance (Chyo Balance JK 180, Chyo Corp., Tokyo, Japan). The weighing was conducted until a constant mass (m1) (48 hours with no weight variance) was obtained. Thickness (four measurements at four equidistant points on the circumference) and diameter (two measurements at the right angles) of each specimen were measured using a digital electronic caliper (Mitutoyo Corporation, Tokyo, Japan). These mean values of the multiple measurements per specimen per dimension were used to calculate the volume (V) for each individual specimen (mm^3). Thereafter, the samples were stored in 6 mL of distilled water at 37°C for 7 days. After the immersion period, samples were weighed (24-hour intervals) after being carefully wiped with an absorbent paper. When constant weight was obtained (48 hours with no weight variance), it was recorded as m2. After this weighing, the samples were returned to the desiccators, the entire mass reconditioning cycle was repeated, and the third constant mass process (48 hours with no weight variance) was recorded as m3. The values for water sorption (WS) and solubility (WSB), in micrograms per cubic millimeters, were calculated using the following equations:

$$WS = (m2 - m3)/V \quad (2)$$

$$WSB = (m1 - m3)/V \quad (3)$$

RESULTS

Degree of Conversion

Two-way ANOVA showed statistically significant differences between materials and temperatures ($p<0.05$). Multiple comparisons among the groups are listed in Table 2. All five adhesive systems exhibited higher mean DC values at 60°C than they did at 25°C. The Adper Easy One had the highest mean value, and XP Bond provided the lowest mean value, regardless of the adhesive temperature condition.

Water Sorption

Two-way ANOVA showed statistically significant differences in the interaction for materials × temperatures ($p<0.05$). Multiple comparisons among the groups are listed in Table 3. With the exception of XP Bond, the remaining four adhesive materials

Table 2: *DC Mean Percentages (Standard Deviations) for the Adhesive Systems at Different Temperatures*[a]

Materials	Temperature	
	25°C	60°C
Adper Easy One	83.8 (3.4) aB	89.3 (2.7) aA
Excite	72.4 (1.9) bB	79.9 (2.0) bA
Adper Single Bond 2	65.1 (2.0) cB	81.8 (2.2) bA
Tetric N-Bond	71.8 (3.6) bB	79.9 (4.0) bA
XP Bond	42.7 (3.3) dB	63.0 (2.1) cA

[a] *Different uppercase letters in rows and lowercase letters in columns indicate statistically significant differences* ($p<0.05$).

demonstrated lower water sorption at 60°C than at 25°C. Adper Easy One and Adper Single Bond 2 provided the highest mean values at 25°C. XP Bond produced the highest values at 60°C.

Solubility

Two-way ANOVA showed statistically significant differences in the interaction materials × temperatures ($p<0.05$). Multiple comparisons among the groups are listed in Table 4. Except for XP Bond, all materials showed lower solubility at 60°C than at 25°C. Adper Easy One showed the highest mean at 25°C. On the other hand, XP Bond showed the highest mean at 60°C.

DISCUSSION

The first null hypothesis tested in this study, that higher temperature would not affect the DC of adhesive systems, was rejected as increased temperature provided higher DC to all adhesive systems tested (See Table 2). As a matter of fact, the DC of resin-based materials is known to be incomplete at room temperature.[10] Higher temperatures are known to reduce viscosity and enhance radical mobility, hence resulting in additional polymerization and higher conversion.[8] The collision frequency of unreacted active groups and radicals increases at higher curing temperatures that are below the glass transition temperature.[10] Further, as the material temperature rises, additional free volume builds up. This, in turn, provides increased mobility to trapped radicals, resulting in further conversion.[10] Therefore, the results obtained in the present investigation appear to be relevant for improving the clinical performance of single-bottle adhesive systems, as optimal physical and mechanical properties of adhesive systems are dependent on a high DC.[5,6] It has been shown in a previous study[9] that preheated Single Bond did not enhance DC. It is possible to argue that the 50°C temperature may have been the reason as explained earlier. In addition, the 10-second air application used to evaporate the solvent can substantially decrease the temperature of the adhesive, leading to reduction in DC. For this reason, the temperature of 60°C was used in the present study. However, further analyses are needed to confirm these assumptions.

Table 3: *Water Sorption Means (Standard Deviations) (µg/mm) for the Adhesive Systems at Different Temperatures*[a]

Materials	Temperature	
	25°C	60°C
Adper Easy One	336.3 (36.0) Aa	238.1 (45.5) Bb
Excite	254.2 (25.6) Ab	198.6 (65.9) Bbc
Adper Single Bond 2	370.7 (55.9) Aa	146.2 (27.1) Bc
Tetric N-Bond	221.2 (26.6) Ab	144.7 (21.6) Bc
XP Bond	244.5 (25.5) Bb	460.5 (27.2) Aa

[a] *Different uppercase letters in rows and lowercase letters in columns indicate statistically significant differences* ($p<0.05$).

Table 4: *Water Solubility Means (Standard Deviations) (µg/mm³) for the Adhesive Systems at Different Temperatures*[a]

Materials	Temperature	
	25°C	60°C
Adper Easy One	270.2 (9.6) Aa	159.2 (37.6) Bb
Excite	130.0 (11.8) Ac	74.6 (7.5) Bc
Adper Single Bond 2	133.5 (16.3) Ac	60.7 (4.1) Bc
Tetric N-Bond	82.7 (7.4) Ad	65.4 (3.8) Bc
XP Bond	184.3 (49.5) Bb	324.9 (13.7) Aa

[a] *Different uppercase letters in rows and lowercase letters in columns indicate statistically significant differences* ($p<0.05$).

Simplified dental bonding agents are composed of a mixture of hydrophilic primers and hydrophobic adhesive resins dissolved in acetone, ethanol, water, or some combination of these solvents.[3] Acetone has a lower boiling temperature (56.5°C) and a higher vapor pressure (200 mm Hg) than ethanol (78.3°C and 43.9 mm Hg) and water (100°C and 17.5 mm Hg).[11] This means that when adhesive systems are heated to high temperatures, such as 60°C, faster solvent evaporation might occur with acetone-based systems because this temperature is higher than the boiling temperature of acetone (56.5°C).[9] The faster solvent evaporation can impair ideal monomer infiltration to dentin leaving a thin adhesive layer that is more susceptible to polymerization inhibition by oxygen. As a result this can be expected to result in decreased bond strength values. For these reasons, only alcohol-based adhesive systems were tested in this study.

Adper Easy One had the highest DC among the adhesive systems in this study, regardless of the temperature. This finding can be explained by its photoinitiator species, which differs from those in other adhesive systems. The formulation of current adhesive systems is usually based on hydrophobic initiators (eg, camphorquinone [CQ]) and co-initiators (eg, ethyl-4-[dimethylamino]benzoate).[12] Most likely they are distributed preferentially to the hydrophobic domains, jeopardizing the overall polymerization of the adhesive.[5] Thus, the inclusion of an alternative hydrophilic photoinitiator, such as trimethylbenzoyldiphenyl phosphine oxide (TPO), with higher reactivity to hydrophilic domains than CQ, has been shown to improve the overall DC of experimental solvent-based adhesive resins.[6] Because Adper Easy One contains CQ and TPO, it is likely that such a combination accounted for the observed higher DC. As an assumption and based on the 350-425 nm absorption range of TPO,[13] the use of a polywave LED with an additional peak around 400 nm could improve the DC for Adper Easy One, although the present study used monowave LED (350-425 nm band). Nevertheless, further analyses should be performed to investigate this possibility.

The concomitant study of physical properties of adhesive systems can contribute to meaningful interpretation of the results. In this study, the benefits of preheating single-bottle adhesive systems on DC, water sorption, and solubility were investigated. Although an increased temperature of single-bottle alcohol-based adhesive systems could enhance the DC for the five materials tested, it decreased the water sorption and solubility for XP Bond alone. This finding requires rejection of the second null hypothesis that preheating would not affect the water sorption and solubility of adhesive systems tested in this study. The presence of residual solvent after air-drying solvated monomer mixtures and the degree of hydrophilicity play a great role in their water sorption and solubility behavior.[14] Fast solvent evaporation is facilitated by its high vapor pressure,[15] but complete evaporation is difficult to achieve and is hampered by the short clinical air-blowing time (10 seconds).[16] XP Bond has tert-butyl alcohol (2-methyl-2-propanol) as a solvent, which consists of a C4 body with an alcohol group surrounded by three methyl groups. This makes it totally miscible with water and polymerizable resins.[17] Thus, although ethanol and tert-butyl alcohol have similar vapor pressures, the latter presents better stability toward chemical reaction with monomers.[16] This may have caused a higher solvent retention in XP Bond samples, so that a higher adhesive temperature was able to increase the DC but not to facilitate the solvent evaporation. Higher amounts of remaining solvent in the adhesive layer may result in more voids, and hence, increased permeability and nanoleakage.[18] In the presence of high water sorption, macromolecular polymer chains undergo a relaxation process as they swell to absorb the water. Initially, the presence of water softens the polymer by swelling the network, and reducing the frictional forces between the polymer chains.[19] After the relaxation process, unreacted monomers trapped in the polymer network are released at a rate that is controlled by the swelling and relaxation capacities of the polymer.[20] Thus, a high amount of permeability will facilitate fluid transport in and out of the network, leading to enhanced water uptake and elution.[21] These affirmations may explain why XP Bond presented the lowest DC, regardless of the temperature, and increased water sorption and solubility when it was preheated. To improve the DC of XP Bond, the use of prolonged evaporation times (60 seconds), with or without air stream, has been observed to be effective.[3]

On the other hand, it is possible that the temperature of 60°C could facilitate ethanol evaporation, which would explain the decreased water sorption and solubility obtained for all other preheated ethanol-based adhesive systems. In fact, Adper Easy One showed the highest water sorption and solubility when it was maintained at room temperature (25°C). It is possible that a higher amount of solvent remained in this adhesive at 25°C. It should also be considered that the extent of solvent retention in polymer networks depends on resin polarity. The resin polarity influences the number of hydrogen bonding sites, and the attraction between the polymer and the solvent. The higher the formation of hydrogen bonds between solvent and monomers, the harder it is to volatilize the solvent.[22] It is likely that a high number of hydrogen bonds between water and self-etching monomers of Adper Easy One could have remained at room temperature, favoring solvent retention. In addition, inclusion of relatively high concentrations of acidic monomers and water permits ionization of those monomers and solubilization of calcium and phosphate, but at the same time it makes the polymers of one-bottle self-etching adhesive systems very hydrophilic.[23] These observations may justify the high sorption and solubility of Adper Easy One at room temperature.

The method used in this study to warm the adhesive systems may not be feasible clinically. However, single-dose adhesive systems can be properly warmed in a few minutes by means of a device used for resin-based composite application, that is, Calset Compule Heater (AdDent Inc, Danbury, CT, USA).

The use of preheated ethanol/water-based adhesive systems seems to facilitate solvent evaporation, which translates to improving the DC and decreasing water sorption/solubility. However, further studies evaluating the impact of preheating other etch-and-rinse and self-etching systems on the DC, water sorption, and solubility are necessary. Also, physical properties and bond strength tests of preheated adhesive systems to tooth or composite should be performed.

CONCLUSION

Preheating single-bottle adhesive systems to 60°C improved the DC of the five adhesive materials tested in this study. Therefore, the first hypothesis that preheating would not affect the DC adhesive system was rejected. The water sorption and solubility for four of the five preheated adhesive materials was also improved, with the exception of the adhesive system containing tert-butyl alcohol. Thus, the second hypothesis that the preheating would not affect the water sorption/solubility adhesive systems was also rejected.

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Conflict of Interest

The authors of this manuscript 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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