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Water Sorption and Solubility of total and Self Etch Adhesive Systems

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ABSTRACT

Objectives: To evaluate water sorption and solubility of total and self-etch adhesive systems at different time intervals.

Materials and Methods: Three adhesives were selected in this study: one and two steps self-etch adhesives (Xeno V and AdheSE) and two steps total etch (Excite). Forty eight discs were prepared, twelve discs for each group as follow:

GI: Xeno V (X);

GII: AdheSE bonding agent mixed with their respective primer in a 1:1 volume ratio (SEBP).

GIII: AdheSE bonding agent alone (SEB).

GIV: Excite (E). Adhesives were polymerized in a teflon mold (5mm in diameter and 2mm in thickness). Specimens stored in vials contain white silica and weighed daily using an analytical balance ± 0.001g until a constant mass (M1) obtained. Specimens of each group were immersed in deionized water for different time intervals (one day, one week, and one month)(n=4) and mass (M2) was recorded. Specimens were replaced into vials for dryness and weighed daily until reached a constant mass (M3). Water sorption (W_{sp}) and solubility (W_{sl})in µg/mm^{3}were calculated according to the formula: W_{sp}= (M2-M3)/V; W_{sl}= (M1-M3)/V. Data analyzed using ANOVA and Tukey tests at 95% confidence intervals.

Results: There were significant differences between adhesives and time intervals for both parameters (water sorption and solubility).

Conclusions: Water sorption and solubility was adhesive system and storage time dependent.

Key words: Water sorption, Solubility, HEMA, Bonding agents.

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INTRODUCTION

Adhesive systems should provide adequate marginal seal and retention of resin composite restorations to dental structures.Effective bonding of resin-based materials to dentin and enamel was achieved with help of technological advances. Existence of water is unavoidable when bonding to dentin thus dentin adhesives should be compatible with wet dentin substrates. Currently,the most commonly used systems are etchand rinse and self-etching were used friendlywhich reduces the number of steps of the bonding protocol[1].Water soluble methacrylate monomer termed as 2-hydroxyethylmethacrylate (HEMA) is frequently used in dentin adhesives. It improves the ability to soak adhesive and resin penetration. It improved ability to mix hydrophilic and hydrophobic components within adhesive[2].

Chemical degradation of adhesives is usually caused by oxidation and hydrolysis, which are processes that require the presence of water. When a solvent enters the polymer network, it causes an expansion of the structure, facilitates extraction of monomers that did not react and promotes the dissolution of linear polymer chains[4]. This expansion is facilitated when the density of crosslinks is low. A solvent has an effect on the water sorption and solubility of adhesives. Malacarne*et al*., 2009[5] suggest that the concentrations of ethanol (5%or 15%) may have increased the water sorption and solubility of solvated adhesives by interfering with their optimal macromolecular packing density.Improvement in solvent evaporation may lead to formation of a highly cross linked polymer with reduced water sorption and solubility[6].
Absorbed water exists in two distinct forms: “unbound water” that is filling the free volume between the chains and the Nano-pores created during polymerization[11], and “bound water” the water molecules attracted to polar groups forms hydrogen bonds[12].

It is well known that durability of dentin bond strength and quality of the seal produced by bonding agents decrease with time both *in vitro* and *in vivo*[7]. Water sorption within resin-dentin interfaces has been quoted as one of the dominant factors involved in adhesion degradation[8]. In addition it has been reported that reduction in mechanical strength[9]and the modulus of elasticity [10] increased with the hydrophilicity of the copolymer blends following water storage. The properties of adhesive materials are affected by sorption and solubility. The new adhesive systems can be attributed to their ability to decrease or eliminate postoperative sensitivity, improve marginal seal, reduce microlekage and enhance the flow of resin into fissure. The development of functional monomers with strong and stable chemical affinity to hydroxyapatite is adirection to continue for improvement of dental adhesion[3]. The hypothesis to be tested in this study that water sorption and water solubility will not affected by the type of adhesive system or storage time intervals. The purpose of this study was to evaluate thewater sorption and solubility of total and self-etch adhesive systems at different time intervals.

MATERIALS AND METHODS

Threedental adhesive systems were used in the study: one step self-etch adhesive (XenoV, Dentsptly), two stepsself-etch adhesive (AdheSE, IvoclarVivadent) and two steps total etch adhesive (ExciTE, IvoclarVivadent)(Figure 1). The composition and manufacturers of each adhesive are shown in (Table 1).Forty eight discs were prepared representing four main groupsaccording to the type of adhesive systems twelve specimens each as follow:

Group I: Xeno Vone step self-etchadhesive (X).**Group II:** AdheSE two stepsself-etchadhesive, where AdheSE bonding agent mixed with the respective primer in a 1:1 volume ratio (SEBP). **Group III:** AdheSEtwo steps self-etchadhesive,AdheSE bonding agent used alone for specimens preparation without primer (SEB).**Group IV:** Excite two steps total etch adhesive (E).

Then each group divided into three subgroups of four specimens according to storage periods in deionized water (one day, one week and one month) (n=4).For specimen preparation, the adhesives systems were dispensed drop by drop into a Teflon mold with 5mm in diameter and 2mm in thickness until it was filled (Figure 2). The mold was placed ona celluloid strip on a glass slide, another strip was placed over the adhesive and covered with a glass slide, a quartz-tungsten-halogen light-curing unit with an output of 550 mW/cm^2 was used to polymerize the specimens for 40 sec the intensity of light curing unit was monitored periodically after every three specimens by radiometer (Figure2,3).

The cured specimens were removed from the mold,and immediately weighed using a sensitive electronic balance (A& DGX-200 company, Limited, Japan) with an accuracy of 0.001g. The specimens were then placed into the individual vials containing white silica inside incubator at 37°C. The specimens were weighed dailyuntil a constant mass (M1) was obtained as specimen dry weight. Specimens were immersed individually in a labeled bottle with 5ml of deionized water; the bottles were sealed until specified storage period. At the end of each storage period the specimens were removed from the bottles, and initially dried with absorbent paper for 15 sec.,weighted and mass (M2) was recorded as specimen wet weight. To obtain full dryness specimens wereplaced into the individual vials containing white silica. The specimens were weighed daily until they were reached a constant mass and (M3) was obtained. The average of water sorption (W_{sp}) and solubility (W_{sl}) in µg/mm^{3}for each group were measured and calculated using the following formula[24,33]:

W_{sp}= (M2-M3)/V
W_{sl}= (M1-M3)/V
V is the specimen volume in cubic millimeters.

Data analyzed using one way ANOVA and Tukey tests at 95% confidence intervals.

RESULTS

For difference in water sorption between adhesives the result of analysis revealed that there was a significant difference in the water sorption value between the adhesives at each time interval $p<0.05$ (Table 2) and (Figure 4).Table (2) showed thatat all interval the X adhesive had significantly the highest water sorption value than other adhesives, while SEB(AdheSE bonding agent) the lowest oneexcept at one day there was no significant difference betweenSEB and E. Generally the water sorption value of the adhesives were seen in thefollowing order (X>SEPP=E>SEB). The statistical analysis of water sorption values between different time intervals for each adhesive revealed that the storage periods had a significant effect on the water sorption of the three adhesives(X, E and SEB adhesives) ($p<0.05$).One month storage time of these adhesives showedsignificantly highervalues than one day. Table (2) showed the details of differences between time intervals. At regarding, water solubility of adhesive systems result showed that there was a significant difference in water solubility between adhesive materials at one week and one month period ($p<0.05$)(Table 3) and (Figure 4). While after one dayof storage interval the result showedno significant difference in the water solubility value between the four adhesives ($p>0.05$).Table (3) showed the value of water solubility for all groups at each storage interval. X had significantly more water solubility than other groups at one week and one month, however there was no significant differences in solubility between SEBP and SEB at all storage intervals. The storage periods had a significant effect on the water solubility of the Xand E adhesives. One month storage time result in more water solubility than one day and one week.

Table 1: Composition of dental adhesive resins used in this study.

Dental adhesives			Manufacturer	Components
Two-step total etch	Excite	E	IvoclarVivadent AG, Liechtenstein	Phosphonic acid acrylate, bis-GMA, HEMA, methacrylates, silicon dioxide, ethanol, catalysts and stabilizers.
Two-step self-etch	AdheSE	SE	IvoclarVivadent AG, Liechtenstein	AdheSE primer: dimethacrylate, phosphoric acid acrylate, Initiator and Stabilizer in an aqueous solution. AdheSE Bond : HEMA,dimethacrylate, silicon dioxide, Initiators and Stabilizers
All in one self-etch	Xeno V	X	Dentsply GmbH, Germany	Bifunctional acrylate, Acidic acrylate, Functionalized phosphoric acid ester, Water, Tertiary butanol , Initiator , Stabilizer.

Figure 1: Bonding systems

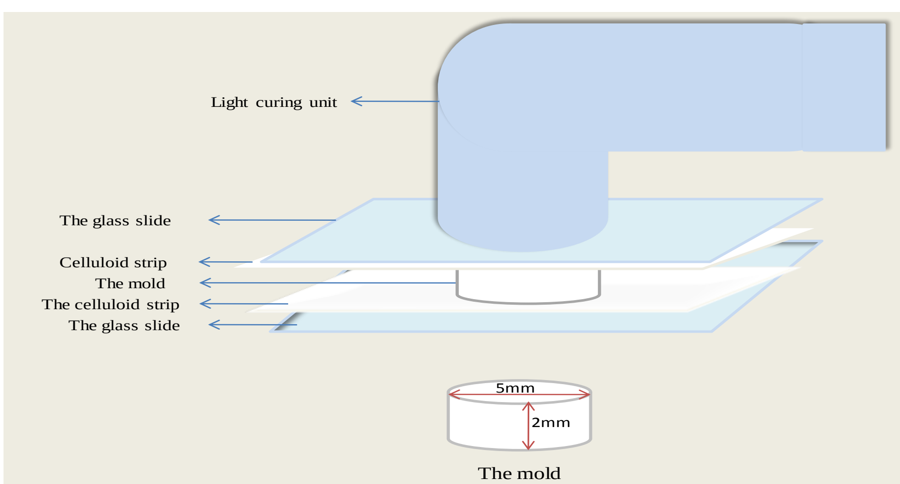

Figure 2: Sample preparation

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Figure 3: The prepared samples

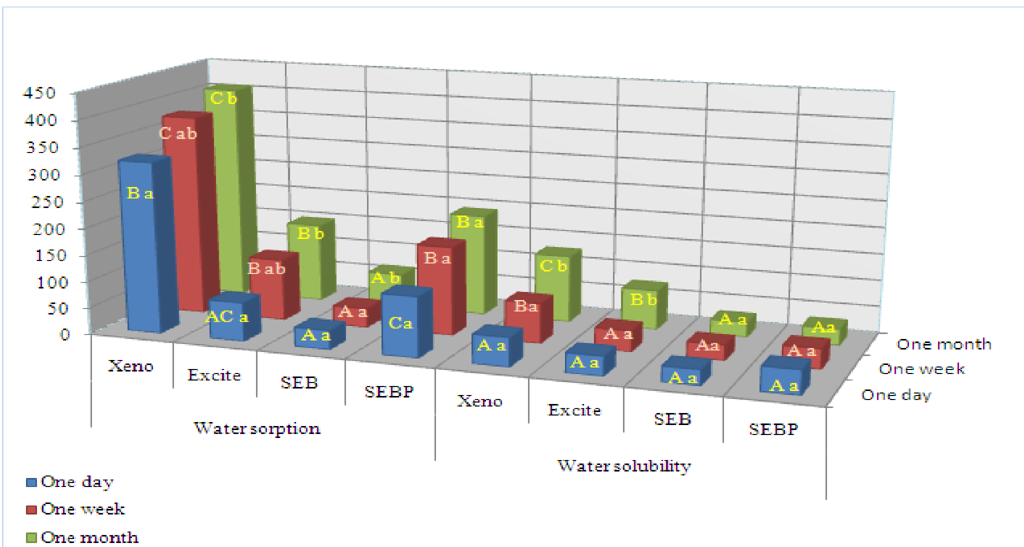

Figure (4): Tukey HSD Homogeneous Subsets for water sorptionand solubility (µg/mm^3)among the adhesive groups at each storage time andamong the storagetimes for each adhesive.

Different capital letters indicate significant difference between adhesives at each period for each test (water sorption, water solubility), Different small letters indicate significant difference between storage times for each adhesive for each test (water sorption, water solubility).

Table 2: The mean of water sorption (µg/mm^3) for each group.

Test	Adhesives	Storage periods		
		One day	One week	One month
Water sorption	Xeno(X)	321.9450 B a	377.9300 C ab	407.5300 C b
	Excite (E)	72.7600 ACa	116.6350 Bab	149.4600 Bb
	SEB	35.7850 Aa	35.8975Aa	63.9325Ab
	SEBP	113.4325Ca	166.9375Ba	192.1350Ba

Different capital letters indicate significant difference between adhesives at each period. Different small letters indicate significant difference between storage times for each adhesive.

Table 3: The mean of water solubility(μg/mm^3) for each group.

Test	Adhesives	Storage periods		
		One day	One week	One month
Water solubility	Xeno	54.0475Aa	75.2075Ba	126.7400Cb
	Excite	36.3800Aa	38.8250Aa	74.4275Bb
	SEB	28.7225Aa	28.7475Aa	28.4700Aa
	SEBP	45.2925Aa	37.8300Aa	31.3425Aa

Different capital letters indicate significant difference between adhesives at each period,Different small letters indicate significant difference between storage times for each adhesive.

DISCUSSION

Three dental adhesives of different category were tested in the present study. Excite adhesive (E) represent two-steps total etch, AdheSE represent two-steps self-etch adhesives and Xeno (X) represent one-step self etch adhesive (all in one).In order to study the effect of primer on the water sorption and solubility of adhesiveAdheSE bonding agent was mixed with the primer (AdheSE Bond and Primer) (SEBP) or bonding agent used alone without primer (AdheSE Bond) (SEB). Water sorption of resin in polymer network depends on resin hydrophilicity and chain topology[13]. Many factors affect polymer water sorption and solubility such as hydrophilic and hydrophopic monomers[14], presence and type of filler[15], presence and type of solvent[16], storage temperature [17]. Resin polarity influences the number of hydrogen bonding sites and the attraction between the polymer and water molecules, while chain topology is related to the spatial or geometric characteristics of the molecular segments and the availability of nanopores within the polymer structure [13,18].

According to the result of this study the X adhesive exhibited higher water sorption value than other adhesive groups at all storage times with significant differences. This may be due to this adhesive more hydrophilic than other adhesives. The all in one adhesive contain ionic monomers hydrophilic, hydrophopic monomers, water and solvents in a single bottle[19], so these adhesive are extremely hydrophilic nature because they contain large quantities of bothionic and hydrophilic monomers. Many studies revealed a strong correlation between the water sorption into adhesive polymers and thehydrophilicity of adhesives, the more hydrophilic the adhesives are, the more water their polymers absorb[10,20].Malacarne*et al.*, 2006[21] andYiu*etal.*, 2006[18]showed that the extent and rate of water sorption increased with the hydrophilicity of the resin blends.The presence of hydroxyl, carboxyl and phosphate groups in monomers and their resultant polymers make them more hydrophilic and more prone to water sorption[22].Dhanpal*etal.*, 2009[23] showed higher water sorption for all-in-one adhesives compared with three-steps etch-and-rinse and two-steps self-etch adhesives.In the study by Feitosa *etal.*, [24] the self-etching/one-step adhesive had the highest ability to absorb water than self-etching/two-steps adhesive and totaletching/self-priming adhesive.In addition a study concluded that the percentage of residual solvent retained in experimental adhesives was significantly influenced by the degree of hydrophilicity of resin monomers, that is, the more hydrophilic the resin, the more solvent it retained [25]. So the incomplete removal of solvent from X may reduce the crosslink density of the polymer chains, resulting in higher free volume and greater water sorption would be seen.

At one week and one month storage time, the water sorption value of the adhesives were seen in the order of X>SEBP=E>SEB. SEBP were exhibited higher water sorption than SEB. In two-bottles adhesive systems, the primers contain hydrophilic monomers and solvents in one bottle, and the adhesives in the second bottle, which are more hydrophobic. When primer monomers are mixed with the adhesive constituents or in a mixture of two separate bottles, the resulting formulation is relatively hydrophilic[26]. SEB showed lower water sorption values than SEBP, a possible explanation for the low water sorption of SEB is the low concentration of hydrophilic monomer in the bonding agent. In addition, the non-solvated SEB resins formed denser polymer networks with a more stable and cross-linked structure, so restricting water sorption.Reis *etal.*, 2007[27] showed that the one-step self-etching adhesive and self-etching primer/adhesive mixtures presented the highest water sorptionvalues. Excite contain BisGMA/HEMA/MA-154,MA-154 =

2-[4-(dihydroxyphosphoryl)- 2-oxabutyl) acrylate, the ionic resin monomers with phosphate groups present in Excite[10], so higher water sorption values were observed in this resin compared to SEB .

The high solubility of X at one week and one month storage period may be due to resin hydrophilicity. Resin solubility increased with the hydrophilicity of the resin blends[5]. However, the hydrophilic nature of bonding resins makes them vulnerable to water absorption and leaching of water-soluble hydrophilic monomers permits water movement across the bonding resin even after curing[28].The two-steps total etch adhesives combine the primer and adhesive resin into one bottle that are dissolved in volatile organic solvents such as acetone, ethanol. This mixture of the different components causes the simplified adhesives to behave more like a hydrophilic primer than a hydrophobic resin[29]. This study support previous studies that demonstrate the adhesives with the high water sorption associated with high solubility behavior, and vice versa[30].At one month storage the (E) showed higher solubility than SEB and SEBP.

The storage periods had a significant effect on the water sorption of the X and E and SEB adhesives. For X and E, one month storage time result in more water sorption than one day, this may be because one month storage time allow sufficient time for water to diffuse through these materials.

For SEB the one month storage time showed more water sorption than one week and one day, while for SEBP, the storage periods had no significant effect on its water sorption, SEBP reached saturation within one day this may be because it is a water based adhesiveaccording to the manufacture, in addition the amount of bonding agent (HEMA) used in SEBP was less than the SEB. It was reported that the amount of water sorption and solubility of adhesive polymers increased proportionally to their HEMA concentrations[31].

The X and E adhesives showed higher solubility significantly at onemonth storage.Increased water-storage time resulted in increased solubility values for X and E adhesives, the continuous solubility of X and E may represent a realistic hydrolytic breakdown of resin compounds, instead of a simple release of unreacted monomers while for SEB and SEBP increased water-storage time not increased their solubility values, This could be explained by the fact that most of the water-soluble components of these adhesives had leached out during the first day.It was reported thatthe adhesives stored in water presented a time-dependent increase in water sorption and solubility[32].

CONCLUSIONS

Under the limitation of this study the higher water sorption and solubility was for all in one adhesive system than two steps self etch and total etch adhesive systems. Increase in water storage time lead to increase the water sorption and solubility of some adhesives.

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