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Marginal Seal of Water-Based Formulation of Light Activated Bonding Agent for Use in Combination with Adhesive Restorations

Hussien Y El Sayed[1]*, Reham M Mohamd[1] and Mona E Essa[2]

[1]*Department of Operative Dentistry, Tanta University, Egypt*
[2]*Department of Operative Dentistry, Elnahda University, Banyswef, Egypt*

Abstract

Aim: The purpose of the study was to investigate effect of water-based solvent adhesive system on micro-leakage of composite resin restorations.

Materials and methods: Class V cavities were prepared on buccal and lingual surfaces of 20 human molars, The occlusal margins were prepared in enamel while cervical ones were prepared in dentin below CEJ by one mm, and classified into four groups according to type of adhesive used five teeth each: Adper Single Bond2 was used as control, Clearfil SE Bond water-based adhesive, Clearfil S3 Bond ethanol-based adhesive, I-Bond acetone-based adhesive. All cavities were restored with Filtek Z250 composite, stored in distilled water at 37°C for 24 hours and thermal cycled. All surfaces, except for restorations and 1 mm from the margins, were coated with two layers of nail varnish. The teeth were immersed in a 3% methylene blue dye solution for 24 hours, and then rinsed in running water, blot-dried and sectioned longitudinally through center of restorations from the facial to lingual surface. The sections (10 sections for each group by n=20 readings) were blindly assessed for microleakage of dye penetration for both the occlusal and gingival margins. Data were collected and statistically analyzed.

Results: Results demonstrate a comparable significantly lesser mean dye penetration in Adper single Bond2 adhesive than in other groups at enamel margins ($p<0.05$) and a highly significant lesser mean dye penetration at dentin margins. Clearfil SE Bond water-based adhesive showed a significant higher mean value of dye penetration at dentin margins than either ethanol or acetone-based adhesives and a comparable non significantly lesser mean dye penetration score at enamel margins.

Conclusion: Clearfil SE Bond (water-based adhesive) showed a significant higher mean value of dye penetration score at dentin margins than either ethanol or acetone-based adhesives.

Keywords: Water-based self etch adhesive; Ethanol-based self etch adhesive; Acetone self etch adhesive; Marginal leakage; Composite resin; Thermocycling

Introduction

The use of direct resin-based composite as a conventional restorative procedure is still critical since class V cavities contains simultaneously enamel and cementum/dentin contours. Therefore, class V restorations should employ bonding agents that allow effective interactions with different tissues, simple handling, and quick application, due to the relative difficulty in accessing these cavities and the presence of margins adjacent to the moist gingival tissue [1]. One-step systems were developed to minimize the number of clinical steps, while incorporating the primer and adhesive into a single bottle. In addition to reducing the clinical time, these systems reduce the sensitivity of the technique and the risk of errors during application [2]. With the one step system, the infiltration of adhesive monomers occurs simultaneously with the self-etching process, decreasing the possibilities of discrepancies between the processes and of unprotected collagen fibers [3]. However, the clinical procedures are likely to be simplified at the expense of bonding performance [2-4]. Clinical microleakage is the major cause of failure for composite restorations and may lead to postoperative sensibility, marginal discoloration, secondary caries, or pulpal inflammation [5].

Adhesive systems have revolutionized the practice of restorative dentistry, these developments, which continue today, have produced results that are reflected by materials that are easy to use and have greater bond strength and lower degradation in the oral environment. These materials applied thinly serve to bond the restorative material effectively to the tooth structure, thus reducing and avoiding marginal microleakage. Adhesion to enamel has been reliable since it was introduced by Bonuocore in 1955 [6]. However, the moist, heterogeneous tubular ultra-structure of dentin poses a real challenge to adhesion. There is a tendency to use adhesives with a simplified application technique, even though it seems to reduce the bond strength to dentin [7] and increase its hydrolytic degradation, because adding increasingly hydrophilic monomers accelerates its degradation in the hybrid layer [8]. Nevertheless, the number of steps and the difficulty in standardizing drying after washing causes dentists to choose adhesives with fewer steps.

Self-etching bonding systems were introduced to resolve some of the problems which have been experienced when using etch-and rinse adhesives. Most self-etching adhesives contain specific functional monomers that, to a large extent, determine the adhesive performance. Functional monomers are used with the intent of etching tooth substrates, enhancing monomer penetration and also (potentially) establishing a chemical interaction between the adhesive and the dental substrates [9]. These systems are classified as two-step and one step self etching regardless of the actual numbers of steps involved in the bonding: 1) Two-step self-etching adhesives are based on the

***Corresponding author:** Dr. El Sayed HY, Professor of Operative Dentistry, Faculty of Dentistry, Tanta University, Tanta, Egypt, Tel: 0201002223308, 0403335631; E-mail: dr-hussein-yehia@hotmail.com

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separate application of a hydrophilic self-etching primer followed by a hydrophobic bonding resin 2) "one-step" adhesives, combine the self-etching primer and the bonding resin into a single step. However, such "one-step" adhesives may require several applications of the adhesive and thus in reality more steps are necessary for good clinical results [10].

The primer and bonding resin formulations of Two-step self-etching adhesives contain a mixture of resin monomers, and light, chemical, or dual cure initiators and other additives. Water as an ionizing medium is also contained in all the self etching primers in order to enable the etching process [11]. The primer includes hydrophilic functional monomers whose acidity allows for the demineralization process of the self etching adhesives. Additionally, they may interact with hydroxyl-apatite crystals (Hap) and collagen matrix phases with a series of atomic-level interactions, which may play a critical role in the overall adhesion process [10].

Mild self etching systems have a pH of around 2. On the enamel the pattern of etching with mild self etching systems is minimal, resulting in shallow inter-crystallite infiltration of the resin and lack of inter-prismatic resin tag formation [12]. On the dentine mild self etching systems are able to partially demineralize and penetrate the dentinal surface up to 1 μm depth, creating hybrid layers that are thinner and with less pronounced resin tag formation than strong self etching and etch-and-rinse adhesives [13]. However, with mild self etching very high dentine bond strength data have been reported. These are similar to those obtained with etch-and-rinse adhesives [14] and their stability under stress is higher compared to the strong self-etching adhesives [15].

One-Step Self Etching Adhesives are complex mixtures of hydrophilic and hydrophobic components which produce thinner hybrid layers compared to two step adhesives and etch-and-rinse adhesives. A greater simplification of the bonding procedure in one step may have clinical advantages, but some studies on their bonding performance indicated that these thin hybrid layers were prone to be less polymerised and permeable [16-18]. They are also very hydrophilic and absorb water from dentinal tubules by osmosis. Unreacted monomers or oligomers can leach out from the polymer during water sorption with subsequent polymer expansion. Generally, increases in water sorption are associated with increases in solubility, hydrolytic degradation products, nanoleakages and a resultant decline in bond strength [19].

Although phosphoric acid has been intensely used to etch the dental substrates (enamel and dentin) for bonding, self etching adhesives are consider alternative methods to prepare the tooth for restorative procedures. Self-etching adhesive systems were developed in attempt to simplify the clinical use of dental adhesives, because they do not require separated phosphoric acid etching, water-rinsing or superficial moist controlling steps. The self-etching primers and adhesives are composed of aqueous solutions of acidic functional monomers and methacrylate components, with a pH relatively higher than that of phosphoric acid etchants [20]. While the adhesion to dentin produced by self-etching adhesives has been considered effective [2], studies are in disagreement regarding the efficacy of conditioning and monomer infiltrations on enamel [21,22]. Morphological analyses of enamel surface treated with self-etching primers have showed not very demineralized surfaces and other areas that were predominantly unetched, which could impair the monomer infiltrations and hybridization process [23,24].

The solvent is a very important component of the adhesive systems. The low viscosity of primers and primer-adhesive resins is partially due to the dissolution of monomers in a solvent. This association will improve the diffusion ability in the porous conditioned substrate, especially in dentin due to its hydrophilic nature. In adhesives, water, ethanol and acetone are the most commonly used solvents. After diffusion solvents must be eliminated from adhesive, otherwise remaining solvent in the adhesive may jeopardize polymerization due to dilution of monomers and may result in voids and increase the permeability of adhesive layer. Complete evaporation is however difficult to achieve because it is limited to the short clinical time [25,26]. The evaporation of the solvent is related to its vapor pressure. Higher vapor pressure of the solvent implies faster evaporation. While the solvent evaporates, the solvent-monomer ratio decreases, as well as the vapor pressure. Thus, within the clinical time, residual solvent may remain in the adhesive and the consequences are directly related to its amount [27].

Water is an indispensable component of self-etch agents, in order to ionize the acidic monomers and trigger the demineralization process [22]. The strong self-etch agents are likely to contain higher amounts of water. A concern is the effect of residual water that remains within the adhesive interface, which hardly can be completely removed [20]. Some self-etch agents present only water as solvent, such as Adper SE Plus (3M/ESPE, St. Paul, MN, USA), AdheSE (IvoclarVivadent, Schaan, Liechtenstein), Adper Prompt (3M/ESPE, St. Paul, MN, USA). However, in many systems, the water is associated to ethanol, acetone or even to monomers, such as the N,N-diethanol p-toluidine, present in the Clearfil SE Bond (Kuraray Medical Inc., Tokyo, Japan) adhesive. On the other hand, water has been related to phase-separation, polymerization-inhibition and reduced shelf-life of self etch adhesives [28].

Another simple approach to improve bonding efficacy and stability is correlated with enhanced solvent evaporation. The air-blowing of the adhesive might help to remove interfacial water, thus improving bonding effectiveness [29]. A mild and extended air-blow should, however, be cooperative to the evaporation of solvent and residual monomers.

The aim of this in vitro study was to compare the marginal microleakage of class V resin-based composite restorations bonded with three different solvent-based, self etch adhesives (Clearfil SE Bond water-based, Clearfil S^3 Bond water-ethanol-based and I-Bond water-acetone-based) and using etch and rinse two step Adper Single Bond 2, adhesive system as control.

The hypotheses tested in this study were: 1) that the water-based self etching adhesives free of either ethanol or acetone co-solvent improves the marginal sealability of resin composite restorations at both enamel and dentin margins and 2) that there is no difference in marginal seal effectiveness between ethanol and acetone when used as solvent in self etching adhesives.

Material and Methods

Materials that have been used in this study, composition and their mode of application illustrated in Table 1.

Twenty newly extracted, sound non-carious human third molars were cleaned of calculus, soft tissue and other debris. A written consent was taken from these patients after the study was approved by the Ethics Committee of Tanta University to ensure their agreement to use their teeth in the current study. The teeth were stored in a 1% Chloramine-T solution (Fisher Chemical, Fair Lawn, NJ, USA) which consisted of 12% active chlorine diluted in distilled water prior to use. The teeth

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Material	Composition	Mode of Application
Clearfil SE Bond (CSEB) (Kuraray Co. Ltd., Osaka, Japan) Two step SE adhesive (water-based)	1- primer: 10- MDP, HEMA, hydrophilic DMA, dicamphorquinone, N,N-diethanol-p-touidine; water.	Apply primer for 20 s, Mild air stream
	2- Bond: 10-MDP, Bis-GMA, HEMA, hydrophilic MA,Dicamphorquinone, N,N-diethanol-p-touidine, silanated colloidal silica. pH 1.4- 2 dry onding lot n: 51435	Apply bond, Gentle air stream
		Light cure for 10 s.
		Apply composite.
Clearfil S³ Bond (CSB)(Kuraray Medical Inc.,Okayama, Japan) one step SE adhesive (water and ethanol-based)	10-MDP, HEMA, Bis-GMA, water, ethanol,silanated colloidal silica, camphorquinone.pH 2.7- dry bonding lot n 00143A	Apply for 20 seconds
		Dry with air pressure for 5 seconds
		Light cure for 10 seconds.
		Apply composite.
I Bond (Haraeus Kulzer,Armonk,Inc.NY USA)	UDMA, 4-META, glutaraldehyde, water, acetone, photoinitiators, stabilizers; pH 2.1; Batch: 010084 dry bonding	Shake the bottle;
		dry; apply 1 layer onto surface cavity;
		agitate slightly for 20 s;
		evaporate solvent by air flow for 5-10 s
		Apply multiple times if cavity does not appear shiny
		Light cure for 20 s
		Apply (water and acetone-based) 5-10 s; one step SE adhesive surface composite.
Adper Single Bond2 (3M-ESPE, Saint Paul, USA) cavity surface composite.	Etchant: 35% $H3PO_4$; colloidal silica pH 0.6	Etch for 15 s; rinse for 10 s
	Adhesive: DMA, HEMA, polyalkenoid acid copolymer, 5 nm silane treated colloidal silica, water, ethanol,photoinitiator; pH 4.3-4.7 - wet; Batch: 4BC bonding lot n N353034	blot excess water with a cotton pellet
		Etch and rinse two-step adhesive system as a control (glistening surface without pooling of water);
		apply 2 consecutive coats of adhesive by micro-brushing gently on the cavity surface;
		evaporate solvent by gentle air stream for 5 s;
		light-cure for 10 s. Apply composite.
Filtek Z250 Micro-hybrid resin Composite (3M ESPE, St. paul, MN, USA)	BIS-GMA and Low-viscosity resin of TEGDMA, a blend of UDMA and Bis-EMA, filled to 60% by volume with zirconia/silica particles a size range of 0.01 to 3.5 microns and an average size of 0.6 micron	Application of composite in a layers less than 2 mm in thickness and cure with light cure unit for 20s according to Manufacturer's instructions.
BIS-GMA (Bisphenol A glycidal dimethacrylate), TEGDMA (Tri ethylene glycol dimethacrylate), UDMA (urethane dimethacrylate), Bis-EMA: (Bisphenol A polyethylene glycol diether dimethacrylate), DMA: dimethacrylate, MMA: methyl methacrylate, 4-META: 4-methacryloxy ethyl trimellitate anhydryed, HEMA: 2-hydroxyethyl methacrylate, MDP: 10-methacryloyloxydecyl dihydrogen phosphate, 4-MET: 4-methacryloxy ethyl trimellitic acid; GPDM: glycerol phosphate dimethacrylate; PENTA: dipentaerythritol pentaacrylate phosphate.		

Table 1: Different adhesive systems used and composite resin restoration.

were then divided into four equal groups each of five teeth according to the type of adhesive being used.

Cavity design

In all groups, Class V cavity preparations were cut on the facial and lingual surfaces of teeth with the coronal margins located in enamel and the cervical margins located in cementum (dentin) with 90° butt joint. The preparations were cut with using a water-cooled high-speed diamond bur #330 (KG Sorensen, Barueri, SP, Brazil). Each bur was discarded following preparation of each group of teeth facially or lingually. Preparation dimensions of 4.0 mm mesio-distally×3.0 mm occluso-cervically×2 mm (depth) were measured with a periodontal probe to maintain uniformity. One operator cut all preparations to ensure a consistent calibrated size and depth.

Restoration groups

In Group 1, control group applying composite resin bonded with Adper Single Bond 2 two step, etch and rinse adhesive system (n=5)

In Group 2, applying composite resin bonded with Clearfil SE Bond (two step, self etch, water-based adhesive system) (n=5)

In Group 3, applying composite resin bonded with Clearfil S³ Bond one step, self-etch, water,ethanol-based adhesive system (n=5)

In Group 4, applying composite resin bonded with I-Bond one step, self-etch, water, acetone-based adhesive system (n=5)

After application of adhesive system into the cavities following manufacturer' instructions, all cavities were restored with a micro-hybrid resin composite, Filtek Z250 (3M ESPE, St. paul, MN USA) in two increments and each increment was polymerized for 40 seconds using a conventional halogen light-curing unit (Coltolux 75, Coltene/Whaldent AG, Alstätten, Switzerland, 500 mW/cm^2). The restorations were finished and polished with Opti-Disk (Kerr Corporation, Via Strecce 4, PO Box 268, 6934Bioggio, Switzerland).

The teeth were stored in distilled water at room temperature for 24 hours before being subjected to 500 thermal cycles between 5°C and 55°C water baths with a 30-second dwell time and a 15-second transfer time. The root apices were sealed with utility wax, and all the surfaces, except for the restorations and 1 mm from the margins, were coated with two layers of nail varnish. The teeth were immersed in a 3% methylene blue dye solution for 24 hours. They were then rinsed in running water, blot-dried and sectioned longitudinally through the center of the restorations from the facial to lingual surface with a water-cooled diamond wheel saw (Leitz 1600, Wetzlar, German) ##. The sections (10 sections for each group by n=20 readings) were blindly assessed for dye penetration by two independent evaluators using a stereo-microscope (Olympus Co., Tokyo, Japan) at 30x magnification. Dye penetration at the composite/tooth interface was scored for both the occlusal and gingival margins from 0 to 3:

0=no leakage visible,

1=penetration of dye along the cavity wall, but less than ½ of the cavity depth.

2=penetration of dye along the cavity wall, more than ½ of the cavity depth.

3=penetration of dye spreading along the axial wall.

Data were collected and statistically analyzed using the Kruskal-Wallis and Mann-Whitney U-tests. A comparison of the occlusal and gingival margins of the groups was performed.

Results

Statistical analysis was performed utilizing the Kruskal- Wallis one-way ANOVA followed by a Mann- Whitney test. The difference between the occlusal and gingival dye penetration scores for each group was analyzed by the Wilcoxon Signed Rank test. Statistical analysis was performed using the following computer program: Statistical Package for the Social Sciences (SPSS Inc, Chicago, IL, USA).

The mean microleakage score values of three different self etching adhesive systems (Clearfil SE Bond, Clearfil S³ Bond and I-Bond and using Adper Single Bond 2 etch and rinse adhesive as a control group) at occlusal enamel and gingival dentin margins are presented in Tables 2-5 and Figure 1.

Kruskal-Wallis one-way ANOVA test indicated a significant differences between occlusal and gingival mean dye penetration scores, where the enamel margins subjecting to a lower dye penetration than dentin margins (mean scores at enamel walls were 0.3 of the control group Adper Single Bond2 and 0.55, 0.5, 0.7 of Clearfil SE Bond, Clearfil S³ and I-Bond adhesive systems respectively, while at dentin walls were 0.8 of the control group Adper Single Bond2 and , 1.9, 1.55, 1.45 of Clearfil SE Bond, Clearfil S³ and I-Bond adhesive systems respectively).

At enamel margins, Adper single Bond 2 etch and rinse adhesive as a control group was record a comparable non significant lowest mean dye penetration score than the other three self etching adhesive systems (0.3, 0.55, 0.5, 0.7 respectively) at p=0.338). While, at dentin margins, Adper single Bond 2 record a highly significant lowest mean score than Clearfil SE bond, Clearfil S³ and I-Bond adhesive systems (0.8, 1.9, 1.55, 1.45 respectively) at p=0.004.

When comparing the occlusal and gingival mean score values for each group, the Wilcoxon Rank test showed significant difference in mean dye penetration for all groups where the dentin margins subjecting to a significant higher mean dye penetration scores than enamel margins at p<0.05.

Groups	Occlusal			Gingival			Paired Differences		Paired Samples Test	
	Mean	±	SD	Mean	±	SD	Mean	SD	t	P-value
Group I	0.300	±	0.571	0.800	±	0.894	-0.500	1.051	-2.127	0.047*
Group II	0.550	±	0.826	1.900	±	0.968	-1.350	1.040	-5.805	<0.001*
Group III	0.500	±	0.607	1.550	±	0.999	-1.050	1.234	-3.804	0.001*
Group IV	0.700	±	0.733	1.450	±	0.887	-0.750	1.251	-2.680	0.015*

(Gr I): Adper single Bond2 etch and rinse adhesive (Control group)
(Gr II): Clearfil SE Bond Self etch, Water-based adhesive
(Gr III): Clearfil S³Self etch, Ethanol-based adhesive
(Gr IV): I-Bond Self etch, Acetone-based adhesive
*Significant p values of paired samples test

Table 2: Mean of dye Penetration Scores and standard deviations for the tested materials at the occlusal and gingival walls.

	Occlusal						ANOVA	
	Range			Mean	±	SD	F	P-value
Group I	0.000	-	2.000	0.300	±	0.571	1.141	0.338
Group II	0.000	-	3.000	0.550	±	0.826		
Group III	0.000	-	2.000	0.500	±	0.607		
Group IV	0.000	-	2.000	0.700	±	0.733		

Table 3: Mean of dye Penetration Scores and standard deviations for the tested materials at the occlusal (enamel) walls.

	Gingival						ANOVA	
	Range			Mean	±	SD	F	P-value
Group I	0.000	-	3.000	0.800	±	0.894	4.790	0.004*
Group II	0.000	-	3.000	1.900	±	0.968		
Group III	0.000	-	3.000	1.550	±	0.999		
Group IV	0.000	-	3.000	1.450	±	0.887		

TOUKEY'S Test					
I and II	I&III	I&IV	II&III	II&IV	III&IV
0.002*	0.064	0.135	0.641	0.433	0.987

*Significant p values of Toukey's test

Table 4: Mean of dye Penetration Scores and standard deviations for the tested materials at gingival (dentin) walls.

Occlusal Gingival		
Group / score	**0 1 2 3**	**0 1 2 3**
Composite bonded with Adper single Bond2. n=20 (**ER two step adhesive)	15 4 1 0	9 7 3 1
Composite bonded with Clearfil SE Bond (water-based) n=20 (*SE two step adhesive)	12 6 1 1	3 1 11 5
Composite bonded with Clearfil S³ Bond (ethanol-based) n=20 (*SE one step adhesive)	11 8 1 0	3 7 6 4
Composite bonded with I- Bond (aceton-based) n=20 (*SE one step adhesive)	9 8 3 0	3 7 8 2

Table 5: Dye Penetration Scores for the tested materials at occlusal and gingival walls.

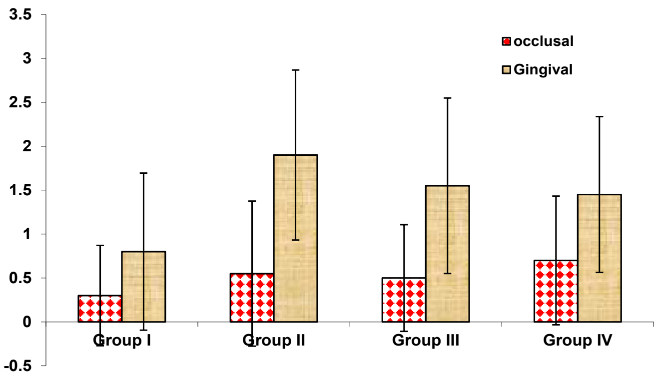

Figure 1: Mean dye penetration leakage values of all adhesives used at enamel and dentin margins.

Gr (2), Clearfil SE Bond water-based adhesive showed a significant higher mean value of dye penetration at the dentin margins than either ethanol or acetone-based adhesives, (1.9, 1.55, 1.45 respectively) at p=0.004, however at enamel margins, showed a comparable non significantly slightly higher mean dye penetration score than ethanol-based clearfil S^3 adhesive(0.55, 0.5 respectively) and non significantly lesser score than acetone-based I-Bond adhesive(0.55, 0.7 respectively) at p=0.338.

On the other hand, acetone-based I-Bond self etch adhesive system record a non significantly slightly lower mean leakage value at gingival dentin margins than ethanol-based clearfil S^3 self etch adhesive (1.45, 1.55 respectively) at p>0.05.

Discussion

The concept of self-etching adhesives is based on the use of polymerizable acidic monomers that simultaneously condition and prime dentin and enamel. Therefore, the self-etching primers eliminate the technique-sensitive rinsing step to remove the phosphoric acid from enamel and dentin. The clinical requirements for self-etching enamel–dentin adhesives are the same as for adhesives used in combination with the acid etch technique. Removal of the weak smear layer on top of the dentin and creation of an adequate etch pattern on the enamel in a clinically relevant period of time (i.e. 15-30 s). Inward diffusion of comonomers into etched enamel and dentin by resin tag formation in the etch pattern and dentinal tubules and intertubular dentin penetration by formation of the so called hybrid layer [30].

The addition of solvents to resins is indispensable to the composition of adhesives [31]. The low viscosity of primers and/or adhesive resins is partly due to the dissolution of the monomers in a solvent and will improve its diffusion ability in the micro-retentive tooth surface. In E and Rs (etch and rinse adhesives), the main function of the solvent, is to promote good penetration of the monomers in the collagen network of the demineralized dentin [32], also be capable of re-expanding the collapsed network [26,33]. In SEAs (self etch adhesives), the use of water as a solvent is indispensable to ensure ionization of the acidic monomers [34,35].

In adhesives, water, ethanol and acetone are the most commonly used solvents, MMA and HEMA, both small monomer compounds have also been described as diluents for other monomers and can therefore also be called solvents. The vapor pressure of a solvent is important to ensure good evaporation of the solvent after application of the adhesive onto tooth tissue. Complete evaporation is however difficult to achieve and is hampered by the short clinical air-blowing time [26,36,37]. Remaining solvent in the adhesive may jeopardize polymerization due to dilution of the monomers and may result in voids and hence permeability of the adhesive layer [26,38,39].

Water is a poor solvent for organic compounds (such as monomers), which are usually rather hydrophobic. This difficulty can be overcome by addition of a secondary solvent, such as ethanol or acetone. Low vapor pressure of water implies that this solvent is difficult to remove from adhesive solutions after application on the tooth. Moreover, Pashley et al. [27], showed that monomers, such as HEMA, decrease the vapor pressure of water even more, which may interfere with the removal of the last amounts of water. Tay et al. [40], showed that excess water in the adhesive resin compromises the bond strength of adhesives due to entrapment of water blisters ('overwet phenomenon').

Ethanol, Its higher vapor pressure as compared to water allows better evaporation by air-drying. Usually ethanol is used in conjunction with water as co-solvent. Moreover, water–alcohol mixtures are known to be 'azeotropic' [41], resulting in a better evaporation of these water–ethanol. Maciel et al. [42], showed that ethanol has a stiffening effect on demineralized collagen, his feature may also explain why ethanol can maintain wide inter-fibrillar spaces after evaporation of the solvent.

In relation to acetone, considered a good choice of solvent in adhesives that combine hydrophobic and hydrophilic components. Its high vapor pressure, which is about four times as high as that of ethanol, is a main advantage. However, its high volatility may also lead to reduce shelf life of acetone-containing adhesives, by rapid evaporation of the solvent. Acetone is frequently used as a solvent alone, but in SEAs it comes as co-solvent with water. Similar to ethanol, acetone and water make an azeotrope and has a very good water removing capacity, because of its high excellent evaporation capacities [36].

Microleakage has been defined as the "marginal permeability to bacterial, chemical and molecular invasion at the tooth/material interface" and is the result of a breakdown of the tooth-restoration interface, causing discoloration, recurrent caries, pulpal inflammation and possible restoration replacement [43,44].

This study evaluated the microleakage of four different adhesives comparing self-etch (Clearfil SE Bond, Clearfil S^3, I-Bond) and total-etch (Adper Single Bond 2) systems, all of which demonstrated dye penetration (leakage) at both the enamel and dentin margins. According to the Wilcoxon signed rank test, significantly less leakage was exhibited at the enamel margin compared to the dentin margin of the adhesive groups and these results in agreement with several studies that evaluated self-etch and total-etch adhesive systems, less microleakage was also reported at the enamel margin compared to the dentin margins [45,46].

Mild self etching systems have a pH of around 2. On the enamel the pattern of etching with mild self etching systems is minimal, resulting in shallow inter-crystallite infiltration of the resin and lack of inter-prismatic resin tag formation. On the dentine mild self etching systems are able to partially demineralize and penetrate the dentinal surface up to 1 μm depth, creating hybrid layers that are thinner and with less pronounced resin tag formation than etch-and-rinse adhesives [12,47]. However, with mild self etching very high dentine bond strength data have been reported and may be similar to those obtained with etch-and-rinse adhesives [14].

Possible reasons for microleakage of contaminants at dentin restoration margins include cavity configuration (C-factor), dentinal tubule orientation to cervical wall (CEJ), organic content of dentin substrate and movement of dentinal tubular fluids, incomplete alteration/removal of smear layer by acidic primers (self-etch systems) for adequate demineralization and hybrid layer formation; inefficient infiltration/penetration of primer components into the demineralized collagen fibrillar network, dentin substrate hydration level (solvent carriers [water, alcohol, acetone] in the adhesive agent reacting differently with varying degrees of surface "moisture" [water]), incomplete evaporation of the solvent from dentin surface prior to attachment of adhesive monomers, acid component composition (pH, osmolality, thickening agent), polymerization contraction of the resin composite [20,46,48]. Since the hybrid layer morphology was not evaluated microscopically in this study, the specific nature of restoration failure (microleakage) for each adhesive system is unknown, although anecdotally four factors are strongly suspected: inefficiency of acidic monomers in alteration of the smear layer for classic hybrid layer formation, cavity C-factor, orientation of dentinal tubules to the CEJ and post-treatment stresses caused by polymerization contraction.

Analysis of the data from this study revealed significantly lower microleakage values with a total-etch adhesive (Adper Single Bond 2) compared to the other adhesives at the enamel and dentin margins. This finding was in agreement with studies reporting decreased leakage associated with total-etch (especially at the enamel margin) compared to self-etch systems [46,49,50]. However, some contradictory results have been reported by other researchers [51,52] reported that Clearfil SE bond showed a bonding effectiveness equally well as the commercial etch and rinse adhesives on dentin due to the use of the functional monomer 10-MPDwhich has been exhibit high chemical interaction capacity to hydroxyapatite crystals [22].

Microleakage of restorations using self-etch adhesives could have resulted from incomplete etching of the enamel surface by acidic monomers, allowing for higher values of microleakage than the total-etch systems using a separate phosphoric acid etchant. Scanning electron microscopy (SEM) studies have shown that the use of phosphoric acid as an enamel etchant improves enamel penetration and the subsequent attachment of adhesive monomers [53].

This study revealed that, Self-etch and total-etch adhesive systems both showed a significantly higher leakage at the dentin margins than enamel margins; and moreover, the results were in agreement with other studies showed a significant difference between self-etch and total-etch adhesives at the dentin margins where self etch systems showed a higher degree of dye peneteration values [45,49,54].

Reasons for the increased leakage scores associated with total-etch systems (phosphoric acid etchants) of dentin compared to enamel surface substrate include hypomineralization of the dentin surface and subsequent collapse of the collagen fibrillar network, inadequate resin monomer infilteration and poorly enveloped of demineralized collagen fibrils and network which became not optimally impregnated, the exposed collagen fibrils not protected by resin are susceptible to hydrolytic breakdown and degradation and may jeopardize the bonding effectiveness [55].

Microleakage of restorations using Clearfil SE Bond water-based adhesive at dentin margins showed a higher significant results comparing to the other self etch adhesives Clearfil S^3 ethanol-based and I-Bond acetone-based (1.9, 1.55, 1.45 respectively) P=0.004. This low bonding effectiveness of clearfil SE bond may be due to the absence of co-solvent ethanol or acetone in this brand of adhesive which adversely affects the adhesive performance and makes it difficult to eliminate excess water in moist dentin and water ingredient in adhesive itself and sites of remaining water and solvents are thought to weaken the adhesive layer [41,56,57].

At the enamel margins, Clearfil SE Bond showed a comparable lower non significant microleakage readings than other self etching adhesive systems Clearfil S^3 ethanol-based and I-Bond acetone-based (055, 0.5, 0.7 respectively) P=0.338, and this may be due to the presence of 10-MDP monomer ingredient which have more chemical bond capacity to hydroxyapatite crystals [58].

I-Bond was self-etch system marketed as a one-bottle, no mixing and system containing a gluteraldehyde component specifically for dentin sensitivity. It showed the lowest leakage scores (not necessarily always significant) at dentin margins than other self-etch adhesive systems (Clearfil SE Bond and Clearfil S^3) (1.45, 1.9, and 1.55 respectively). Possible explanations for this occurrence include multiple applications (manufacturer's instructions) of I-Bond adhesive onto the preparation surfaces and increased waiting periods prior to light polymerization and may be due to presence of acetone co-solvent with higher vapor pressure, which aids in the elimination of excessive water in moist dentin and water ingredient in adhesive itself without compromising polymerization of adhesive [41,59,60].

Also, I-Bond adhesive at enamel margins in this study, recorded a slightly non significant lower performance in comparing with other self etch adhesives (Clearfil SE Bond and Clearfil S^3) and with etch and rinse one (Adper Single Bond 2) (0.7, 0.55, 0.5, 0.3, respectively) P=0.338, and these results may be explained by the lack of an apparent etching pattern of I-Bond and that adhesives containing 4-META also may show lower enamel-bonding capacities than 10-MDP-containing adhesives [58,61].

The viscosity, surface tension, functional monomers, pH, water concentration, and cohesive strength of adhesives may affect bonding and explain microleakage of the adhesives studied. Other features, such as the type of composite, cavity, and dye tracer, analyzed tooth section, and number of sample, may also have influenced the results [4,62].

Although, one and two step self etching adhesives generally contain the same components but the amounts of ingredients applied on the tooth surface differ considerably, whereas two step consist of a pure acidic priming hydrophilic monomer solution dissolved in organic solvent and water and a solvent free bonding agent containing hydrophobic cross-linking monomers such as UDMA and TEGDMA. Usually, one step self etching adhesives contain acidic functional monomers dissolved in high concentration of organic solvent and/or water which usually make up almost 50% of the adhesive blended with hydrophobic cross-linking monomers, so the concentration of hydrophobic cross-linking monomers is drastically reduced which responsible to the mechanical polymerization bonding of the adhesive. Relatively, less hydrophobic cross-linking monomers are available on the tooth surface after application of the one step self etching adhesives lead to impairing the performance of bonding and reduce adhesion phenomena [52].

Furthermore, Modern adhesives are very technique sensitive, and any minor mistakes in their use might give rise to disturbances in the bonding process. Therefore, an adhesive with a durable bond, easy application, and low technique sensitivity is still a clinical necessity. Consequently, research is still underway by manufacturers and they sometimes introduce newer products with claims of better bonding properties [63,64].

Conclusions

Within the limitations of this in vitro study, the following conclusions were reached:

1. All adhesive system groups exhibited dye penetration (leakage) at both the occlusal (enamel) and gingival (dentin) margins.
2. At the enamel margin, statistically significant lower leakage was exhibited among the adhesive groups than dentin margin.
3. At the enamel and dentin margin, Adper Single bond 2 etch and rinse adhesive revealed significantly lesser leakage compared to the other adhesive groups.
4. A comparison of the self-etch adhesives at the enamel margin revealed I-Bond had non significantly slightly higher leakage value.
5. Water-based self etching adhesives free of either ethanol or acetone co-solvent may improve the marginal sealability of resin composite restorations at enamel margins but does not at dentin one.
6. There is no significant difference in marginal seal effectiveness between ethanol and acetone when used as co-solvent in self etching adhesives.

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