



## Evaluation of Degree of Conversion, Resin-Dentin Bond Strength, and Durability of Polydopamine Incorporated Total Etch Adhesive System

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### ABSTRACT

**Objectives:** This study aimed to evaluate the degree of conversion (DC%), shear bond strength (SBS), bond durability, and the resin-dentin interface of polydopamine (PDA) incorporated total-etch adhesive system.

**Materials and Methods:** Five percentage by weight (wt%) concentration of PDA incorporated adhesive was prepared and its polymerization was evaluated using Fourier-transform infrared spectroscopy. The results were compared with the DC% of conventional total-etch adhesive (Adper Single Bond 2) using independent t-test. Occlusal surfaces of 80 freshly extracted human premolars were sectioned to expose the dentin, which was acid-etched. The samples were divided into two groups (n=40) of total-etch adhesive and total-etch adhesive+PDA. Composite resin blocks were built up over the bonded surface and cured. Each group was subdivided into two subgroups (n=20) of immediate and post-aging evaluation. Samples were immersed in 10% sodium hypochlorite for five hours, and then, the SBS was evaluated using a universal testing machine at a crosshead speed of 1 mm/minute. The resin-dentin interface was evaluated using confocal laser scanning microscopy. Data were analyzed using two-way analysis of variance and post-hoc Bonferroni test.

**Results:** DC% was not affected by the addition of 5% PDA to the adhesive (group 2). The SBS of group 2A was significantly higher than that of group 1A. There was no significant reduction of SBS in group 2B. The hybrid layer was less degraded after aging in group 2B compared to group 2A.

**Conclusion:** PDA incorporated adhesive increased the immediate bond strength and durability without changing the DC%.

**Keywords:** Polydopamine; 3M Single Bond Dental Adhesive; Dental Etching; Polymerization; Dental Bonding

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### INTRODUCTION

Clinical longevity of adhesive resin restorations depends on the durability of resin-dentin bonds [1]. Though the initial

bond strength of total-etch adhesives is very high, several studies have reported that the resin-dentin interface gradually degrades [2-4]. An in-vivo study by Hashimoto et al [5] has

shown that the bond strength of a three-step total-etch adhesive can be reduced by 50-65% after 2-3 years. In total-etch systems, 37% phosphoric acid is used to demineralize the smear layer and underlying intact dentin to expose the collagen network. The diffusing monomers are not able to completely displace the loosely bound water from the internal and external compartments of collagen fibrils [6]. The discrepancy in the depth of resin infiltration creates an exposed zone of demineralized dentin beneath the hybrid layer that is vulnerable to degradation [7,8]. This could be followed by proteolytic degradation of collagen fibrils by the acidic activation of matrix metalloproteinases (MMPs) [9-11] and hydrolytic degradation by elution of resins from the suboptimally polymerized hybrid layer [1,12]. This results in the formation of a water-rich collagen substrate with partially depleted hydroxyapatite crystals, which are susceptible to proteolytic attacks. Further, it has been reported that the water content of caries-affected dentin is 2.7 times greater than that of normal dentin [13]. As most of the current adhesives are very sensitive to excess moisture, bonding to wet dentinal substrates becomes even more complicated. Hence, the development of dental adhesives that can effectively adhere and provide adequate bond strength, even in an aqueous environment, is of great importance. Strategies to reduce proteolytic activity within hybrid layers include modification of dentin adhesives with agents such as quaternary ammonium methacrylates or benzalkonium chloride, chlorhexidine, and proanthocyanidin for collagen cross-linking and anti-proteolytic properties [6]. However, an adhesive system with both potential antiproteolytic activity and water compatibility is the current area of focus in the development of novel dentin adhesive systems.

Recently, the adhesion strategies of maritime creatures, such as mussels, have gained attention in the medical field. Mussels can attach to any natural inorganic, organic, and synthetic material in an aqueous environment by secreting mussel adhesive foot proteins (MAPs) that harden in situ to form water-

resistant bioadhesion [14]. The strong adhesion capability of mussels has been ascribed to the amino acid composition of MAPs, which is rich in 3,4-dihydroxy-L-phenylalanine (L-DOPA) and lysine amino acids. L-DOPA also forms strong covalent and non-covalent interactions with the substrates [15]. DOPA readily donates one or two electrons and converts to quinone, which is highly reactive to other functional groups such as thiols, amines, and catechol-quinones [16]. The oxidative polymerization of DOPA readily forms polydopamine (PDA), which mimics DOPA in its adhesive property. Researchers have utilized the adhesive characteristic of MAPs in the synthesis of mussel mimetic synthetic DOPA containing polymers (PDA) that serve as adhesive for biomedical applications such as for bone adhesives in orthopedic surgeries and surgical wound closure [17]. Catechol functionalized PDA methacrylate with ferric ions has been proven to increase the shear bond strength (SBS) of dentin adhesive systems with no leakage pattern at the bonding interface when used as a surface pretreatment [18]. In addition, catechols are known to adsorb onto hydroxyapatite more than alcohols, amines, and carboxylic acids [19]. Xu et al [20] proved that PDA promotes enamel and dentin remineralization and can inhibit the degradation of dentinal collagen fibers by MMP inhibition activity. There is no evidence in the literature on the effect of incorporation of PDA in dentin adhesive systems. Hence, the purpose of the present study was to evaluate the effect of the addition of PDA to a total-etch adhesive system on the degree of conversion (DC%), SBS, and durability of the resin-dentin bond. The null hypothesis was that PDA incorporated dentin adhesives do not affect the DC%, SBS, or bond durability of the resin-dentin bond.

## MATERIALS AND METHODS

*Preparation of the experimental bonding agent:* First, 400mg of dopamine hydrochloride powder was added to 200ml of Tris buffer diluted hydrochloric acid (HCl; pH=8.5) solution and allowed to self-oxidize for four hours to form PDA at a 2mg/ml concentration.

A commercially available total-etch adhesive system (Adper Single Bond 2; 3M ESPE, St. Paul, MN, USA) was used in this study. The optimal percentage was obtained by a pilot study using various concentrations, including 1, 3, 5, and 8%. The optimal percentage of 5% was chosen by evaluating DC% using Fourier-transform infrared spectroscopy (FTIR). The optimal percentage of PDA was added to total-etch adhesives to obtain five percentage by weight (wt%) concentration of PDA incorporated bonding agent. The mixed vial was kept in a cyclomixer (Sigma Scientific Products, Tamil Nadu, India) for one minute to obtain a uniform mixture. The samples were divided into group 1 (total-etch adhesive) and 2 (total-etch adhesive+PDA).

#### *Evaluation of DC%:*

For FTIR analysis, five specimens were made for each group (n=5). Equal amounts of each adhesive resin were placed on a transparent polyethylene film. The solvents were evaporated by airstream for 30 seconds, covered with a second film, and pressed to form a thin adhesive layer, which was placed into the sample holder of the FTIR spectrometer (PerkinElmer Spectrum One, USA). During spectra recording, the resin was continuously in contact with the sensor. The absorbance peaks were recorded in the transmission mode at a resolution of 4 cm<sup>-1</sup> with scans in the range of 400-4000 cm<sup>-1</sup>. The adhesives were then light-cured with a light-emitting diode (LED) Bluephase unit (Vivadent, Schaan, Liechtenstein) at a light intensity of 600 mW/cm<sup>2</sup> for 20 seconds. The absorptions were recorded for the cured adhesive specimens in the same manner. DC% was calculated from the aliphatic C=C peak at 1638/cm and normalized against the aromatic C=C peak at 1608/cm according to the following formula:

$$DC\% = \left\{ \frac{1-[C-aliphatic/C-aromatic]}{[U-aliphatic/U-aromatic]} \right\} \times 100$$

where U aliphatic and U aromatic correspond to non-polymerized specimens while C aliphatic and C aromatic correspond to polymerized specimens. Data were analyzed using independent t-test and Tukey's HSD (honestly significant difference) test at the significance level of 0.001.

#### *Sample preparation:*

Eighty caries-free human premolars, extracted for orthodontic reasons, were selected for the study after obtaining written consent from the patients. The teeth were stored in deionized water and a 0.1% thymol solution until use. Teeth with cracks, caries, erosion, abrasion, attrition, restorations, and crown or root fractures were excluded. The teeth were divided into two groups of 40 samples each. The occlusal surfaces of the teeth were ground below the dentino-enamel junction with a diamond disk to expose the middle section of the dentin, which was smoothed with 600-grit silicon carbide papers to obtain a flat and uniform dentin surface devoid of enamel remnants. The surface was acid-etched with 37% orthophosphoric acid (D-Tech Etching Gel, Prime Dental, India) for 15 seconds followed by rinsing for 30 seconds. The surface was blot dried, and the respective adhesives were applied, gently air-dried, and light-cured for 20 seconds. The bonded surfaces were restored with a light-cured composite resin (Tetric N-Ceram, Ivoclar Vivadent, Schaan, Liechtenstein) using a Teflon mold of a 2mm thickness and 6mm diameter and cured according to the manufacturer's instructions. The groups were divided into two subgroups of A and B, based on the aging process. Twenty samples in each group were subjected to SBS evaluation immediately (subgroups 1A and 2A), and the remaining samples were subjected to aging (subgroups 1B and 2B) according to Toledano et al [21] by immersing in 10% sodium hypochlorite (NaOCl) solution for five hours, which was followed by SBS and resin-dentin interface evaluation. The bond strength attained by this method was proved similar to the results of observed reductions in the resin bond strength obtained by in-vivo degradation studies. This was the reason for using this aging protocol in this study.

#### *Evaluation of the resin-dentin interface:*

The remaining ten samples from each group were examined for the resin-dentin interface concerning the presence and quality of resin penetration depth using confocal laser scanning microscopy (CLSM; Carl Zeiss MicroImaging GmbH, Göttingen, Germany). Samples for CLSM analysis were prepared similarly, except that 0.1 mg/ml of rhodamine

B dye was added to the experimental bonding agents before application. The teeth were longitudinally sectioned into six parts towards the deeper surface using a slow-speed diamond disc under copious water supply to expose the resin-dentin interface. The sectioned surface was polished with a 600-grit silicon carbide paper. The dentin-adhesive interfacial region was examined under the CLSM at four points. A 543nm excitation line was used, and the fluorescence emissions were collected beyond 560nm. The optical sections were taken at 1.0µm intervals starting from 30µm with 40× oil immersion objective and 1.4 numerical aperture (NA). The penetration depth of the adhesive resin along the resin-dentin interface and the hybrid layer formed were checked for thickness and quality.

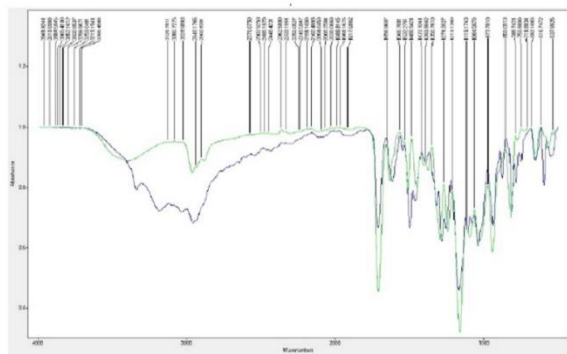
*Statistical analysis:*

The DC% data were analyzed using independent t-test and Tukey's HSD test at the significance level of 0.001. SBS results were statistically analyzed using two-way analysis of variance (ANOVA) and post hoc multiple comparison Bonferroni test.

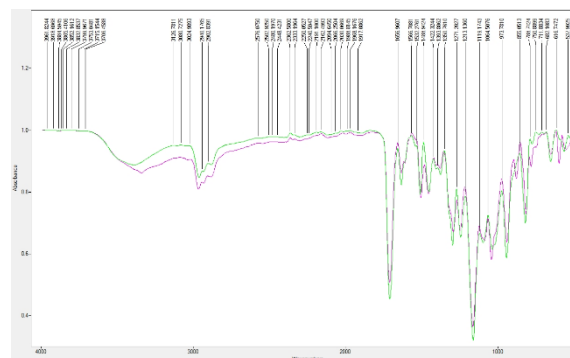
**RESULTS**

*DC% evaluation:*

On analyzing the DC%, there was no statistically significant difference ( $P>0.05$ ) between groups 1 ( $37.03\pm0.34$ ) and 2 ( $36.87\pm0.37$ ). The FTIR spectra of the study groups are shown as graphical representations of the absorbance peaks for non-cured and cured resins in Figures 1 and 2, respectively.



**Fig. 1.** The absorbance peaks of the infrared rays by C=C and N—H in the monomer of the non-cured adhesive resin (green=group 1, blue=group 2)



**Fig. 2.** The absorbance peaks of the infrared rays by C=C and N—H in the monomer of the cured adhesive resin (green=group 1, pink=group 2)

The absorbance peaks of the infrared rays for aliphatic C=C of the monomer were 1635 cm<sup>-1</sup> and 1636 cm<sup>-1</sup> for non-cured and cured resins, respectively. The absorbance peaks of the infrared rays for aromatic C—C of the monomer for non-cured and cured resins were 1534 cm<sup>-1</sup> and 1537cm<sup>-1</sup>, respectively.

*SBS evaluation:*

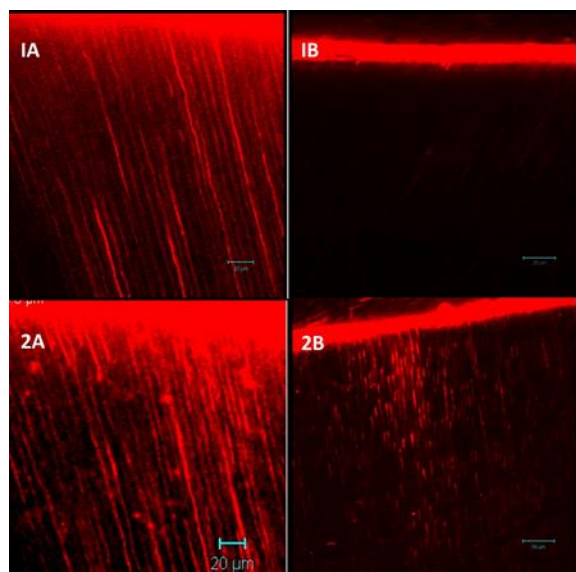
The two-way ANOVA was used to examine the effect of various groups and aging on the SBS. Normality was checked using the Shapiro-Wilk test. Homogeneity of various groups was assessed by Levene's test for equality of variances, and outer layers were assessed by inspection of the boxplot. There were no outer layers, and data were normally distributed. There was homogeneity of variables ( $P>0.05$ ). There was a statistically significant interaction between groups 1 and 2 and between subgroups A and B. The mean SBS values for the experimental groups are given in Table 1.

**Table 1.** Comparison of the mean shear bond strength (SBS) between groups 1 and 2 (N=10)

Groups	Mean	SD	95% Confidence Interval	
			Lower Bound	Upper Bound
1A	22.81 <sup>b</sup>	0.49	22.53	23.1
1B	13.68 <sup>a</sup>	0.3	13.4	13.1
2A	26.74 <sup>c</sup>	0.4	26.47	27.03
2B	24.68 <sup>b</sup>	0.53	24.4	24.1

SD=Standard Deviation; Different superscripts indicate significant differences in the mean SBS values among the groups ( $P<0.05$ )





**Fig. 3.** Representative confocal laser scanning microscopy images showing interfacial characterization of the resin-dentin interface. 1A, immediate-bonding agent (BA), shows a clear hybrid layer and long resin tag penetration; 1B, post-aging-BA, shows no depth of penetration with short resin tags; 2A, immediate-polydopamine (D)-BA, shows better resin tag penetration with a thick hybrid layer; 2B, post-aging-D-BA, shows a comparatively lesser reduction in the hybrid layer thickness and resin tag penetration

Group 2 showed significantly higher SBS than group 1 in the immediate and post-aging samples. In group 1, subgroup B showed a significant reduction of SBS after aging compared to subgroup A. However, there was no significant difference between the subgroups of group 2. There was also no significant difference between subgroups 1A and 2B ( $P < 0.001$ ).

#### *Penetration depth evaluation:*

The confocal image of group 1A shows a comparatively thicker hybrid layer and better resin tag penetration compared to group 1B. Group 1B showed less degradation of resin tags after aging, indicated by a reduction in the depth of penetration. The addition of PDA did not affect hybrid layer formation or resin tag penetration as shown in group 2A. However, in group 2B, there was a lesser reduction in the hybrid layer thickness and resin tag penetration compared to groups 1B and 2A (Fig. 3).

## DISCUSSION

The results of the present study accept the null

hypothesis that PDA incorporated adhesive does not negatively affect any of the tested parameters. It has been identified that the presence of DOPA and lysine residues in MAPs is mainly responsible for moisture-resistant adhesion through a hydrogen bond, metal com-plexation, covalent interaction with the substrate, etc. [22]. Lee et al [18] prepared a catechol functionalized polymer, called dopamine methacrylate methoxyethyl acrylate. It was found that this new polymer improved the mechanical and adhesive properties of a commercial dental adhesive when pretreated onto the artificial saliva-contaminated dentin surface. [18] Moreover, histological analysis of the bonding interface showed no leakage pattern, proving that the polymer is effective in improving the properties of the interface between the hybrid layer and adhesive resin [18]. Thus, this study intended to evaluate the effect of the addition of PDA to a total-etch adhesive on DC%, bond strength, and the hybrid layer interface.

The physical and mechanical properties of photo-cured adhesives are directly influenced by the DC% attained during polymerization [23]. DC% is determined by the proportion of the remaining concentration of the aliphatic double bonds in a cured sample relative to the total number of double bonds in the non-cured material [24]. The addition of 5w% of PDA to the adhesive did not cause any statistical difference in DC% between groups 1 ( $37.03 \pm 0.34$ ) and 2 ( $36.87 \pm 0.37$ ). Lee et al [24] evaluated photopolymerized DOPA functionalized copolymers in the presence of atmospheric oxygen and found that DOPA content had no apparent effect on the gelation process. DOPA and other catechol compounds readily convert themselves into quinone through donating two electrons to reduce a counter molecule. DOPA promotes the crosslinking reactions of MAPs through the oxidation of catechol hydroxyl groups to ortho-quinone [25], which subsequently triggers intermolecular crosslinking, providing cohesion and bulk elastic properties for the protein network [26].

In this study, aging was done according to the protocol followed by Toledano et al [21].

Garbui et al [27] proved that there was no difference in the microtensile bond strength ( $\mu$ TBS) of the etch-and-rinse adhesive system between the samples stored for the short-term in 5% NaOCl and for the long-term in water. NaOCl is a nonspecific deproteinizing agent. In aqueous solutions, superoxide radicals are formed and induce oxidations, which break long peptide chains of proteins followed by chlorination of protein terminal groups. Furthermore, hypochlorous acid-derived chloramines have been shown to increase the proteolytic susceptibility of the collagen. Thus, storage in NaOCl causes hydrolytic degradation of resin and solubilization of unprotected collagen fibrils within the decalcified dentin, which has been proven equal to in-vivo degradation of 2-3 years [21]. On SBS evaluation, group 2 showed significantly higher immediate bond strength ( $26.74 \pm 0.40$ ) compared to group 1. It has been found that PDA is highly reactive to other functional groups, including thiols, amines, and other catechol-quinones through Schiff base substitution or 1,4 Michael-type addition [22,28,29].

Direct interactions between the catechol side chains of dopamine and the contacting surfaces have been considered as the mechanism of adhesion. These could be the reasons for the improved bond strength of PDA incorporated adhesive. The SBS of group 1 in our study was  $22.81 \pm 0.48$ , which significantly reduced to  $13.68 \pm 0.29$  after aging, which is in accordance with various in-vitro and in-vivo studies on bond strength [3,4,12].

The CLSM evaluation of the resin-dentin interface showed less defined hybrid layers with short resin tags in aged samples when compared to the immediate evaluation of samples. It has been shown that simplified etch-and-rinse adhesives and self-etch adhesives can release and activate endogenous MMPs during dentin bonding, which could lead to thinning and disappearance of collagen fibrils from incompletely infiltrated hybrid layers in aged dentin [4,7].

Our study showed that there was no significant reduction in the SBS of group 2B ( $24.68 \pm 0.53$ ). This was supported by the CLSM view of the resin-dentin interface of post-

aging samples, which showed lesser degradation of the hybrid layer and resin tags. This increase in the bond durability could be attributed to the MMP inhibitory effect on dopamine. Xu et al [20] proposed that dopamine could be used as a pretreatment agent to inhibit MMP activity and to maintain the integrity of exposed collagen fibers. The polymerized dopamine binds strongly with collagen, stabilizes it via extensive hydrogen bonding augmented by hydrophobic interactions, and prevents free access of collagenases to active sites on collagen chains. On the other hand, catechol groups of dopamine are oxidized to form dopamine quinones, which create a film over the substrate, blocking the site of action of MMP [30]. Hence, future research is necessary to assess the MMP inhibitory activity of PDA incorporated total-etch adhesives on the hybrid layer. In addition, long-term clinical studies on bond strength evaluation are necessary to optimize the constituents of total-etch adhesive systems with PDA.

## CONCLUSION

Under the limitations of the present study, it can be concluded that the addition of 5% PDA does not affect the DC% of the two-step total-etch adhesive system (Adper Single Bond 2; 3M ESPE, St. Paul, MN, USA). Furthermore, SBS and bond durability were enhanced after the aging process.

## CONFLICT OF INTEREST STATEMENT

None declared.

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