

Bond strengths of three-step etch-and-rinse adhesives to silane contaminated dentin

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This study aimed to evaluate the effect of silane coupling agent contamination on the microtensile bond strength (μ TBS) of 3-step etch-and-rinse adhesives on dentin. Flat occlusal dentin surfaces were prepared and randomly divided into 8 groups ($n=20$) based on the tested adhesives; Scotchbond Multi-purpose or Optibond FL, with contamination of an experimental silane (2 vol% of 3-m ethacryloxypropyltrimethoxysilane at pH 4.5) before acid-etching, after-etching or after-priming; while the groups without silane contamination served as controls. μ TBS data were analyzed by two-way ANOVA and Tukey's HSD tests at a significance level of 0.05. Additional specimens of contaminated dentin were used to analyze changes in the organic molecules by Fourier transform infrared spectroscopy (FTIR). Silane contamination before acid-etching did not significantly change μ TBS ($p>0.05$), but contamination after-etching and after-priming significantly decreased μ TBS of both adhesives ($p<0.05$). Silane contamination had an adverse effect on the dentin bond strength of 3-step etch-and-rinse adhesives especially after-priming.

Keywords: 3-step etch-and-rinse adhesive, Silane coupling agent, Microtensile bond strength, Contamination, Ceramic repairing

INTRODUCTION

Esthetic metal-free restorations have become increasingly popular in clinical practice, because of not only patients' desires for esthetic restorations, but also the progressive developments in the mechanical properties and fabrication technologies of these materials. Glass ceramic or silica-based ceramic materials can be highly esthetic and possess sufficient strength for use as inlay, onlay, crown, or veneer restorations^{1,2}.

However, in clinical practice, complications; such as secondary caries, cervical abrasion and restoration fracture can occur^{3,4}, in which either replacement or repair is chosen to resolve such problems. Many advantages of repairing rather than replacing a restoration, can be summarized as follows: preservation of tooth structure, a reduction in harmful effects on the dental pulp, reduced damage to adjacent teeth, reduced treatment times and costs, and good patient acceptance⁵. One strategy is to repair with resin composite or rebond the fractured part, which can be done intraorally using the appropriate bonding procedure^{6,7}.

When repair-bonding to silica-based ceramic materials, the application of a silane coupling agent is recommended to promote chemical adhesion between silica particles and resin^{8,9}. Silanes are composed of two reactive functional groups; alkoxy functional groups, which are activated by hydrolysis before reaction with

the surface hydroxyl groups of the inorganic substrate and organo-functional groups, which can react with organic resin monomers^{10,11}. Furthermore, silane can increase the surface energy of the material to enhance resin wettability^{12,13}. Therefore, silane application is an essential step to improve the bonding performance by chemical bonding between the silica-based ceramic and resin adhesive.

In case of intraoral repairs at the margins of ceramic restorations, two or more adhesive substrates including dentin or enamel, would be involved. These adhesive substrates require different adhesive treatments, in which there are risks of cross-contamination by silane from the adjacent adhesive substrate. A previous study has demonstrated that silane contamination on dentin prior to universal adhesive application either in etch-and-rinse or self-etch mode did not affect the dentin bond strength⁷. However, long-term bonding performance of universal adhesive is still insufficiently proven. Based on evidence of successful laboratory and long-term clinical performance, 3-step etch-and-rinse adhesive has been recommended for bonding to tooth substrates¹⁴. According to the 3-step etch-and-rinse adhesive protocol, phosphoric acid etching, priming and bonding procedures should be performed on tooth substrates without any contamination. Etching the dentin surface completely removes the smear layer and exposes the demineralized collagen fibril network, where resin monomers in

the primer and bonding agent can penetrate into demineralized dentin and establish micromechanical interlocking between the resin adhesive and dentin¹⁵). It should be noted that treatment of the exposed collagen fibril network is the most critical factor for etch-and-rinse systems to exhibit higher bonding performance. Generally, a silane coupling agent contains ethanol as a solvent as well as silane. Therefore, contamination of the dentin surface during bonding procedures could affect the expansion/shrinkage aspect of the exposed-collagen fibril network¹⁶), and might disrupt the monomer penetrating process and influence resin-dentin bond strengths. Also, the sequence of contamination; before etching, after etching, or after priming might have different effects on resin-dentin bond strengths. Unfortunately, information on the effect of contamination of silane coupling agent on the dentin bond strength of 3-step etch-and-rinse adhesives is scarce.

Therefore, the purpose of this study was to evaluate the effect of silane coupling agent contamination during 3-step etch-and-rinse adhesives procedure on microtensile bond strength to dentin. The null hypothesis of this study was silane coupling agent contamination has no adverse effect on the bond strength of 3-step etch-and-rinse adhesives.

MATERIALS AND METHODS

Silane coupling agent preparation

A silane coupling agent was prepared by mixing methacryloxypropyl trimethoxy silane (MPS; Merck, Darmstadt, Hesse, Germany) with 70 vol% ethanol solution (mixture of 99.9 vol% ethanol and deionized water) to obtain 2 vol% silane solution in accordance with that published in study¹⁷). 99.8% acetic acid was used to adjust the pH to 4.5 to control the rate of hydrolysis of MPS.

Specimen preparation

One hundred and sixty human caries-free permanent third molars were used in this study after approval by the Faculty of Dentistry Human Experimentation

Committee, Chiang Mai University No. 2/2018. Teeth were stored at a temperature of -4°C and used within 1 month of extraction. The occlusal enamel was cut to expose dentin and the teeth were ground with 600-grit silicon carbide paper to create standardized smear layers. The teeth were randomly divided into 8 groups based on two commercial 3-step etch-and-rinse adhesives; Scotchbond Multi-purpose (3M, St. Paul, MN, USA) or Optibond FL (Kerr, Orange, CA, USA), with and without contamination procedures of an experimental silane before acid-etching, after acid etching or after priming (Fig. 1). The experimental silane coupling agent and the adhesives used are shown in Table 1. The adhesives were applied to the dentin surface according to manufacturers' instructions using a dry bonding technique.

Resin composite (Filtek Z350XT, 3M) was then placed in 3 increments of 1.5 mm, each of which was light-cured at the light intensity of $1,100\text{ mW/cm}^2 \pm 10\%$ for 20 s (Bluephase Style, Ivoclar Vivadent, Schaan, Liechtenstein). The teeth were stored in distilled water at 37°C for 24 h and then sectioned perpendicular to the adhesive interface using a low-speed diamond saw (IsoMet low speed cutter, Buehler, Lake Bluff, IL, USA) under water cooling to obtain beam-shaped specimens from each tooth at the resin-dentin interface with a cross-sectional area of 1.0 mm^2 ($n=20$).

Microtensile bond strength (μTBS) test

The beams were attached to a universal testing machine (Instron 5566, Instron, Bangkok, Thailand) with cyanoacrylate glue (Model repair II blue, Dentsply Sirona, York, PA, USA) for the μTBS test at a crosshead speed of 1 mm/min. The μTBS data were analyzed with SPSS 22.0 (SPSS, Chicago, IL, USA), using two-way ANOVA and Tukey's HSD tests at significance level of 0.05.

Failure mode analysis

After testing to failure, all the specimens were sputter-coated with gold (SPI-Module Sputter Coater, SPI supplies, West Chester, PA, USA) and examined using a scanning electron microscope (SEM; JSM-5910LV, JEOL,

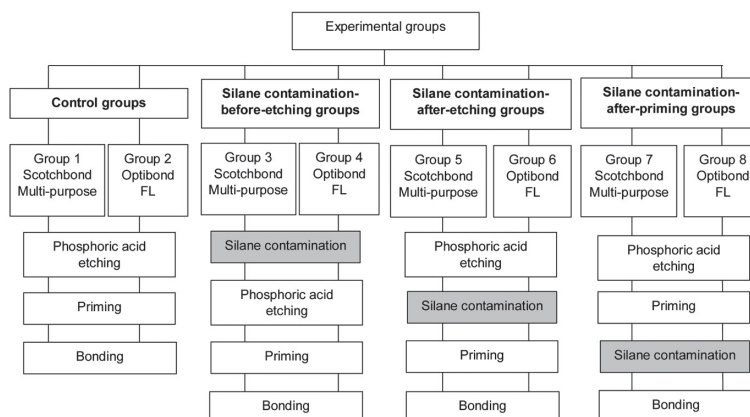


Fig. 1 Schematic representation of the experimental design.

Table 1 Materials used in this study

Name [Batch number]	Composition	Application
Scotchbond Multi-purpose (3M, St. Paul, MN, USA) [N731151]	Etchant: 35% H ₃ PO ₄ , silica thickener	Apply 15 s, rinse 20 s and air dry 5 s
	Primer: HEMA, polyalkenoic acid polymer, water, ethanol	Apply 15 s and air dry 5 s
	Adhesive: Bis-GMA, HEMA, tertiary amines, CQ, hexafluorophosphate, EDMAB, DHEPT	Apply, air dry and light cure 10 s
Optibond FL (Kerr, Orange, CA, USA) [6502640]	Etchant: 37.5% H ₃ PO ₄ , water, silica thickener	Apply 15 s, rinse 20 s and air dry 5 s
	Primer: HEMA, GPDM, MMEP, water, ethanol, CQ, BHT	Actively rub 15 s and air dry 5 s
	Adhesive: Bis-GMA, HEMA, GDMA, CQ, ODMAB, filler (fumed SiO ₂ , barium aluminoborosilicate, Na ₂ SiF ₆), coupling factor A174	Actively rub, air dry and light cure 10 s
Experimental silane coupling agent (Merck, Darmstadt, Hesse, Germany) [K48642573 704]	Methacryloxypropyl trimethoxy silane (Silane A174), ethanol, acetic acid	Apply 60 s and air dry 5 s

Bis-GMA: 2,2-bis[4-(2-hydroxy-3-methacryloyloxy-propoxy)-phenylene] propane; BHT: butylhydroxytoluene; CQ: camphorquinone; DHEPT: N,N-di-(2-hydroxyethyl)-4-toluidine (co-initiator); EDMAB: ethyl-4-(dimethylamino)benzoate; GDMA: glycerol dimethacrylate; GPDM: glycerol phosphate dimethacrylate; HEMA: 2-hydroxyethyl methacrylate; MMEP: mono-2-methacryloyloxyethyl phthalate; Na₂SiF₆: disodium hexafluorosilicate; ODMAB: 2-(ethylhexyl)-4-(dimethylamino) benzoate (co-initiator)

Tokyo, Japan). The fractured surfaces were classified into 4 categories¹⁸: 1. mixed failure (adhesive failure between dentin and resin, as well as cohesive failure within dentin and/or resin); 2. adhesive failure (failure between dentin and resin over 80% of the surface); 3. cohesive failure within dentin (failure in dentin over 80% of the surface); 4. cohesive failure within resin (failure in resin over 80% of the surface). The failure mode data were analyzed using the nonparametric Pearson Chi-square test.

Fourier transform infrared spectroscopy (FTIR)

Additional specimens for FTIR analysis were prepared in 5 conditions: acid-etched dentin with and without priming, silane contaminated dentin before and after etching and silane contaminated dentin after priming. A FTIR (Nicolet 6700 FT-IR Spectrometer, Thermo Fisher Scientific, Waltham, MA, USA) was used to take 64 scans at a resolution of 4 cm⁻¹ in the spectral range 4,000–400 cm⁻¹ to observe the composition change of collagen, resin monomer and silane on the dentin surfaces.

SEM observation

In order to observe the thickness of hybrid layer at resin-dentin interface, additional bonded specimens in each group were prepared as described above, where resin composite was built up in one 1.5-mm layer. After 24-h stored in distilled water, the specimens were sectioned perpendicularly to the interface using a low-speed diamond saw. The resin-dentin interface of each specimen was then polished using a sequence of 600-,

800-, 1000-, 1200-, 1500-, 2000-SiC papers. After being dehydrated and fixed in ascending grades of ethanol and hexamethyldisilazane (HMDS), respectively, the specimens were gold-sputter coated (SPI-Module Sputter Coater) and observed under a SEM (JEOL-5910LV) at 2,500× magnification.

RESULTS

μTBS test

The mean μTBSs of each group are shown in Table 2. The results of the two-way ANOVA revealed that the μTBS were significantly influenced by both factors: 3-step etch-and-rinse adhesives used ($p < 0.001$) and silane contamination conditions ($p < 0.001$). In addition, there was a significant interaction between the two factors ($p < 0.001$).

The silane contamination before acid-etching did not significantly change μTBS ($p > 0.05$). On the other hand, the contamination after etching significantly decreased the μTBS of both adhesives, and the contamination after priming caused a further significant reduction ($p < 0.001$).

In each of the contamination conditions, Optibond FL adhesive showed significantly higher μTBS than Scotchbond Multi-purpose adhesive ($p < 0.001$), except the silane contamination-after-priming groups ($p = 0.889$).

Modes of failure

The results of the failure mode analysis from each group are summarized in Fig. 2. Most of failure modes

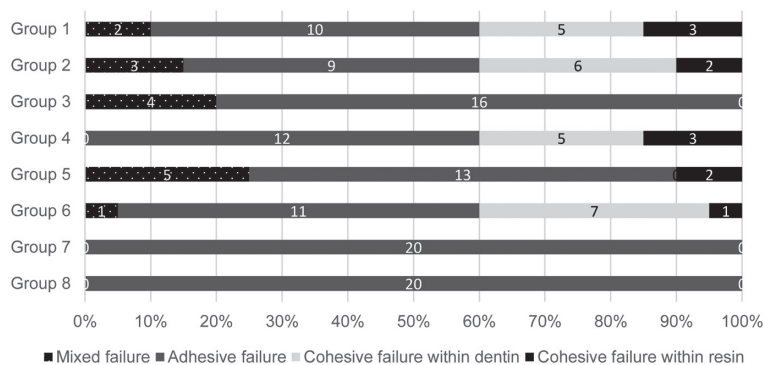


Fig. 2 Percentage of failure modes of the resin-dentin bond in each group ($n=20$). The numbers in each bar are the number of fractional failure modes in each group.

Table 2 Mean and standard deviations of microtensile bond strength (MPa, $n=20$)

Adhesive	Control group	Silane coupling agent contaminated groups		
	No contamination	Before etching	After etching	After priming
Scotchbond Multi-purpose	51.59 (4.68) ^b	49.10 (5.29) ^{bc}	32.30 (4.92) ^d	17.93 (4.68) ^e
Optibond FL	68.15 (6.85) ^a	67.29 (6.05) ^a	44.46 (4.79) ^c	20.09 (3.33) ^e

Groups identified by different superscript letter means a significant difference ($p<0.05$).

were adhesive failure, especially the groups, which were contaminated by the silane coupling agent after priming. There were significant differences in failure mode distribution among the groups ($p<0.001$).

FTIR analysis

The molecular changes, which occurred in each dentin condition are shown in Fig. 3. The FTIR spectra showed similar absorption bands around $1,633\text{ cm}^{-1}$ (stretching vibrations of peptide C=O groups) and $1,544\text{ cm}^{-1}$ (N–H bending vibrations coupled with C–N stretching vibrations) in all the dentin conditions, indicating amide I and amide II from collagen¹⁹, respectively. In the primed acid-etched dentin and silane contamination-after-etching dentin, the absorption bands at $1,715\text{ cm}^{-1}$ (carbonyl C=O stretching) and 810 cm^{-1} (C=C twist), corresponding to monomers²⁰ present in primer were observed.

An absorption band at around 476 cm^{-1} (Si–O–Si)²¹ was observed in silane contamination-after-etching and after-priming dentin, which indicated residual silane on the dentin surfaces, but not in silane contamination-before-etching dentin.

SEM observation

Resin-dentin interfaces presenting the thickness of hybrid layer are shown in Fig. 4. In both of dental adhesives, silane contamination-after-priming groups (Figs. 4D and H) exhibited the thinner hybrid layers than the other groups.

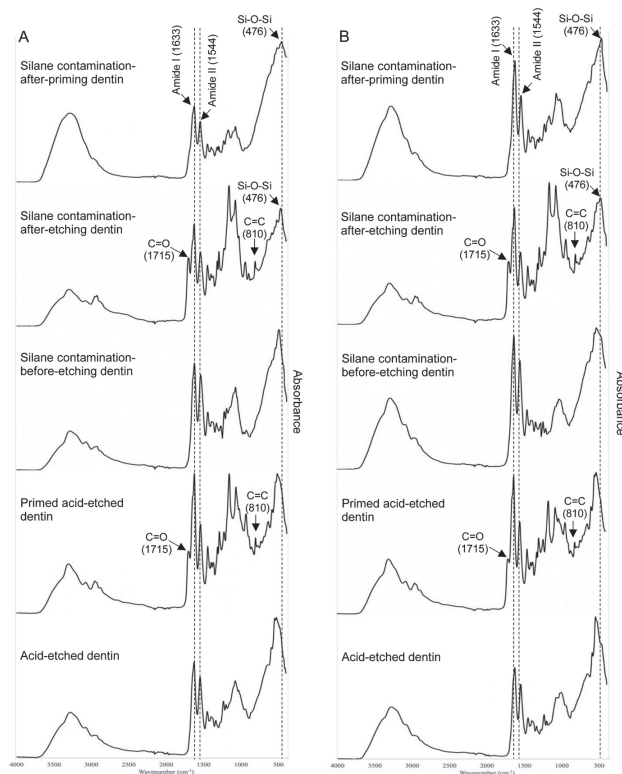


Fig. 3 FTIR spectra of each dentin condition using (A) Scotchbond Multi-purpose, (B) Optibond FL.

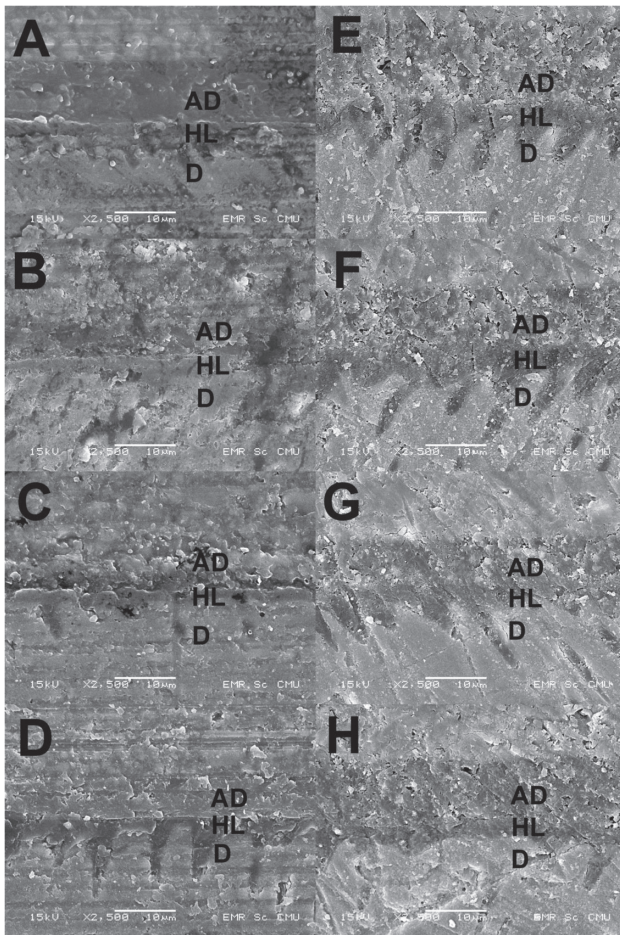


Fig. 4 SEM images of resin-dentin interfaces using (A–D) Scotchbond Multi-purpose, (E–H) Optibond FL. Morphology of resin-dentin interfaces in control (A, E), silane contamination-before-etching (B, F) and silane contamination-after-etching (C, G) groups are similar regarding hybrid layer thickness. In silane contamination-after-priming groups (D, H), the hybrid layers were thinner than that of control and the other contaminated groups in both adhesives. D: dentin; HL: hybrid layer; AD: adhesive layer.

DISCUSSION

The results of this study revealed that silane-contamination on dentin before the etching step did not change the dentin bond strengths of 3-step etch-and-rinse adhesives, but contamination after-etching and after-priming had a negative effect on dentin bond strengths. Therefore, the null hypothesis was partially rejected.

Generally, current commercial silane coupling agents contain 1–5 vol% MPS and ethanol in acidic condition. In this condition, the Si-O-CH₃ groups of MPS are hydrolyzed to silanol groups, which are responsible for chemically bonding to silica-based materials²². However, the pre-hydrolyzed silane could have short

shelf life, since the formation of Si-O-Si oligomers by self-condensation between silanol groups in the acidic condition decreases the activity of silane overtime^{23,24}. This study focused on the effect of residual products of MPS and ethanol on the dentin surface on dentin bonding. Thus, an experimental silane coupling agent was prepared by mixing MPS in the ethanol solution with acetic acid just before use in order to ensure hydrolysis to the silanol groups on the adhesive dentin surface.

In this study, the silane contamination-before-etching groups did not exhibit a significant alteration in μ TBS from the non-contaminated groups. As with previous dentin bonding studies on the effects of contamination with blood^{25,26} and saliva²⁷, and pre-application with metal primer²⁸, the contaminants would be rinsed off with water from the dentin surface etched with phosphoric acid. It was confirmed by FTIR result, where the silane contamination-before-etching dentin showed the absorption bands without Si-O-Si peak, that was referred as acid-etched dentin. The absence of a Si-O-Si peak in the absorption band of 476 cm⁻¹ indicated that silane was washed away by phosphoric acid etching and water rinsing steps. Therefore, the bond strengths of silane contamination-before-etching groups were comparable to the control groups. However, this application procedure would not affect the silane-treated ceramic, because etching and water washing did not significantly improve or reduce the bond strengths of silane-treated ceramic^{29,30}.

Normally, air-blowing after phosphoric acid etching can shrink the exposed collagen fibril network on the dentin surface as a result of the rapid evaporation of water, and the surface tension forces operating at the air-water interface³¹. These allowed interpeptide hydrogen bonds between adjacent collagen fibrils³², resulting in the reduction of interfibrillar spaces for resin monomer penetration³³. Additionally, it was found that much of the bound water still remained in the intrafibrillar compartments of the collagen fibrils even if unbound or free water in interfibrillar compartments was evaporated by air-drying³⁴. This bound water could interfere with the infiltration of the dental adhesive into the intrafibrillar compartment of collagen fibrils³⁵, resulting in a reduction in bond strength^{36,37}. On the other hand, silane-contamination after acid-etching would allow the penetrating ethanol on the dentin surface to slightly re-expand the collagen fibril network^{32,38}. Additionally, ethanol soaking could further remove some amounts of remaining water from the intrafibrillar compartment of the collagen molecules as well as the interfibrillar compartment^{34,39}. The penetrating ethanol might improve the permeation of primer components into the intrafibrillar compartment of the collagen molecules⁴⁰. Furthermore, it has been speculated that the MPS molecule (molecular weight=234 Da) dissolved in ethanol can also penetrate into the collagen fibrils, where the silanol group would then react with the hydroxyl groups of the fibrils by forming hydrogen bonds⁴¹.

The subsequent application of a primer on silane-contaminated acid-etched dentin was expected to re-

expand the collapsed collagen fibril network. Previous research has demonstrated that a primer containing HEMA-water or HEMA-ethanol could re-expand the demineralized collagen matrix on dried dentin⁴². Furthermore, the primers used in this study contain a quantity of the acidic monomers; polyalkenoic acid copolymer in Scotchbond Multi-purpose and GPDM in Optibond FL, rendering these primers to be acidic. However, the results of this study showed that the μ TBSs of the silane contamination-after-etching group were significantly lower than those of the non-contaminated control and silane contamination-before-etching groups. The unstable H-bond between MPS molecules and fibrils might be broken by following primer application, then the released MPS might undergo self-condensation, forming Si-O-Si oligomers²⁴, or interacting with acidic monomers in the primer⁴³. As evidenced in the FTIR spectra of the silane contamination-after-etching groups, Si-O-Si oligomers on the dentin surface at the absorption bands of 476 cm^{-1} ²¹ were present. Although self-condensation requires a long time in acidic solution, reported to be 2 h in a pH of 2.7 in a previous study²⁴, the relatively higher pH of the primer solution, or the higher concentration of MPS dissolved into the primer might enhance the condensation rate of MPS^{21,41}. Otherwise, the free silanol groups may undergo self-condensation during solvent evaporation as evidenced by NMR research⁴⁴.

It was speculated that the presence of Si-O-Si oligomers absorbed on dentin surface in the silane contamination-after-etching group hindered the hydrophilic character of the dentin surface, which is determined by surface -OH concentration, and instead showed hydrophobic characters on contaminated dentin²¹. Thus, the presence of Si-O-Si oligomers might interrupt the infiltration of hydrophilic monomers in the primer, resulting in a reduction of the bond strength to silane-contaminated etched dentin. The results of this study were contrary to the previous study, which demonstrated that silane contamination on dentin prior to universal adhesive application had no effect on dentin bond strength⁷. Although the silane contamination on acid-etched dentin in this study was comparable to the contamination in etch-and-rinse mode of universal adhesive, the successive applied agents were different. Universal adhesive is relatively more hydrophobic than primer of 3-step etch-and-rinse adhesive⁴⁵. Therefore, the monomers might infiltrate contaminated dentin in different manner.

The contamination of silane coupling agent after priming revealed the lowest μ TBS, and all the specimens debonded at the adhesive interface. These results would indicate inferior penetration of the adhesive monomer into etched dentin. It has been established that the priming step helps maintain the interfibrillar spaces within the collagen fibril network, as well as increasing surface energy^{46,47}. Thus, the primed acid-etched dentin would be ready for the infiltration of relatively hydrophobic monomers^{48,49}. Subsequent silane-contamination on the primed acid-etched dentin would disrupt those surface characteristics, since ethanol in

the silane coupling agent would dissolve the primer components bound to the collagen fibril network into the solution again. Attempting to evaporate concentrated ethanol by air-drying, could further remove free monomers such as HEMA⁵⁰, leaving a lesser amount of bound primer monomers on the dentin surface to maintain the collagen fibril network. As a result, air-drying after contamination of the silane coupling agent could cause re-collapse of the collagen fibril network^{31,32}, leading to a lesser degree of interfibrillar spaces in the collagen fibril network^{32,42}. This speculation was evident in SEM images of silane contamination-after-priming groups in both dental adhesives, whose hybrid layers were thinner than that of controls and the other contaminated groups. Furthermore, the application of silane coupling agent on the primed-dentin surface might create a new high concentration mixture of ethanol and monomers, which might reduce the vapor pressure of ethanol upon ethanol evaporation⁵¹. Thus, there is an increased possibility of residual ethanol being retained on the dentin surface, which could impair polymerization and weaken the strength of the adhesive layer⁵². Additionally, the MPS molecules undergo self-condensation when left on the primed acid-etched dentin surface, which could impair the penetration of adhesive monomers in the bonding agent. It is also speculated that the MPS molecules might copolymerize with resin monomers and this may worsen, as the incorporation of large molecules could increase resin viscosity and lower the degree of conversion⁵³. These would be a reason why the μ TBSs in the silane contamination-after-priming groups reduced and all the specimens showed adhesive failure.

Regarding the type of adhesive, Optibond FL showed significantly higher μ TBS than Scotchbond Multi-purpose. In accordance with a previous study⁵⁴, Scotchbond Multi-purpose contains polyalkenoic acid copolymer, which has high molecular weight and doesn't dissolve in adhesive solution, causing phase separation and resin globules formation in a polymer that cannot penetrate into collagen fibril network. This polyalkenoic acid copolymer can be deposited as gel on the collagen surface^{55,56}, and impede monomer infiltration⁵⁴.

In this study, an experimental silane coupling agent was prepared in acidic form by mixing with acetic acid before use. One might argue that the acidic pH of the silane solution resulted in further demineralization of the dentin and lead to too deep a demineralized collagen matrix for complete monomer infiltration. However, a previous study revealed a protective effect for the demineralized organic matrix presenting on the lesion surface against the acid attack by interfering with the diffusion of ions outside and inside the demineralized area⁵⁷. Thus, the effect of pH of the silane solution might be negligible. Within the limitations of this study, it was concluded that contamination of the silane coupling agent on dentin affected the dentin bond strength of the tested 3-step etch-and-rinse adhesives, which could be mainly attributed to the presence of residual MPS on the dentin surface after etching and priming as well as

the effect of ethanol solvent in the exposed collagen fibril network.

In this study, the silane contamination after priming dramatically decreased dentin bonding of 3-step etch-and-rinse system. Therefore, when bonding of 3-step etch-and-rinse system to the surface combined with ceramic restorative materials and dentin surface, the silane contamination after priming should be strictly avoided. In addition, the results of this study indicated that silane coupling agent should be specifically applied on ceramic surface before primer application to dentin surface.

CONCLUSION

The silane contamination on dentin surface especially after priming step decreased the dentin bond strength of 3-step etch-and-rinse dental adhesive. Therefore, in intraoral repair of ceramic, silane coupling agent should be applied on ceramic before primer application on dentin.

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