Original article

The Effect of Various Types of Silane Coupling Agents on The Wettability of Hydrofluoric Acid-Etched/Unetched Lithium Disilicate Surfaces

Tanapon Tarateeraseth[1], Tool Sriamporn[2], Niyom Thamrongananskul[1]

[1]Department of Prosthodontics, Faculty of Dentistry, Chulalongkorn University, Bangkok, Thailand

[2]Department of Prosthodontics, College of Dental Medicine, Rangsit University, Pathumthani, Thailand

Abstract

The aim of this study was to investigate the effect of treating the etched/unetched lithium disilicate surface with various types of silane coupling agents on the contact angle measurement. One hundred fifty lithium disilicate disks were prepared to dimensions of 10 millimeters in diameter and 3 millimeters in height. The samples were randomly divided into two groups: hydrofluoric etched and unetched lithium disilicate surfaces before silane application. Each group was further divided into five subgroups, according to type of silane coupling agent used to treat the prepared surfaces, no treatment (control), Kerr silane primer, Monobond N, Rely X ceramic primer and an experimental silane, respectively. The contact angles between deionized water and the prepared surface were measured using a contact angle tester via the sessile drop method. Data were statistically analyzed using Two-way analysis of variance and Tukey's multiple comparison tests (α=0.05). The results showed that in the unetched lithium disilicate disk group, the lowest contact angle values were observed in the control group, whereas the Monobond N group showed the highest contact angle values. Within the etched group, the control group also demonstrated lowest contact angle and the Kerr silane primer group exhibited highest contact angle value. In conclusion, application of silane coupling agents significantly reduced the wettability of deionized water on the silane-coated surface. The type of silane coupling agent selected significantly influenced the wettability of deionized water. Etching the surface with hydrofluoric acid prior to silane application significantly increased surface wettability in all treatment groups except for groups that were treated with resin-containing silane primer.

Keywords: Contact angle, Hydrofluoric acid, Lithium disilicate, Silane coupling agent, Surface treatment

Received date: Dec 23,2019 **Revised date:** Jan 23,2020 **Accepted date:** Feb 3,2020

Doi: 10.14456/jdat.2020.15

Correspondence to:

Tool Sriamporn, Department of Prosthodontics, College of Dental Medicine, Rangsit University. 52/347 Muang-Ake, Phaholyothin Road, Lak-Hok, 12000, Pathum Thani, Pathum Thani, Thailand. Tel: 0-2997-2200-30 Ext. 4312 E-mail: Tool.s@rsu.ac.th

Introduction

All-ceramic restorations have gained increasing popularity due to their biocompatibility and ability to mimic natural tooth structure that offers good esthetics. Newer types of ceramics such as lithium disilicate glass and zirconia ceramic have demonstrated more than sufficient strength to withstand intraoral forces. They can be used to fabricate inlays, onlays, and crowns in both the anterior and posterior regions.[1] However, the longevity of the ceramic restoration does not depends solely on the mechanical properties of the material alone. Several other factors affect restoration longevity; for example, caries index, type of dentition, site of restoration, size of restoration, reasons for placement, oral cleanliness, etc.[2] The quality of the tooth-restoration bond is a dominant factor that greatly influences the clinical outcome.[3,4] A reliable resin bond promotes retention of the restoration,[5] improves its marginal adaptability,[6,7] reduces microleakage[6,7] and enhances fracture resistance.[8]

Several methods had been proposed to achieve the optimal resin-ceramic bond including mechanical with chemical modifications, as mechanical treatment alone may be inadequate in providing a reliable bond between the ceramic surface and resin cement. For silica-based ceramics, a reliable bond between the resin-ceramic surfaces can be achieved by hydrofluoric acid etching (mechanical bonding) along with silane priming (chemical bonding).

Hydrofluoric acid promotes the bondability of lithium disilicate by roughening its surface which increases the total surface area available for bonding.[9] The mechanism can be explained by the bond affinity between fluoride and silicon being higher than that of silicon-oxygen.[10] The acid selectively removes the glassy matrix of the lithium disilicate surface, leaving an exposed crystalline structure which is responsible for the micro-mechanical retention with resin cements.[11,12]

Silane coupling agents are used in various applications in the field of dentistry. Trialkoxysilanes, such as 3-methacryloyloxypropyltrimethoxysilane (MPS), is one of the most commonly used silane coupling agents.[13] Silane molecules can react to water molecules via hydrolysis to produce three silanol groups (-Si-OH) from the corresponding methoxy groups (-Si-O-CH3). The silanol groups are capable of forming stable siloxane networks (-Si-O-Si-O-) on the glass ceramic surface. Application of silane may be one of the most crucial steps in obtaining an optimal bond between silica-based ceramic and resin cement.

Various types of silane coupling agents are commercially available in the market. Two-bottle systems are known to provide a longer shelf life and increase initial reactivity compared to the one-bottle system.[14] However, to simplify the bonding procedure, manufacturers tend to produce prehydrolyzed single-bottle silanes that contains other universal adhesive primers which may include many other components such as bisphenol A glycidyl methacrylate (Bis-GMA), phosphate-containing monomers, etc.[15]

Organofunctional silanes are bifunctional molecules that promote the chemical bond of silica-based ceramic and composite resins.[16] Its hydrophobic property may influence the reduction of the hydrolytic degradation of the bond.[17,18] Many studies have proven silane coupling agents to improve the bond strength of silica-based ceramic and resin cement. However, studies regarding the contact angle and wettability of substrate after treatment with silane coupling agents of different compositions is still limited. Silane coupling agents are believed to help increase the surface energy of the substrate and wettability of the luting agent to the coated ceramic surface.[16,19,20]

The purpose of this study was to investigate the effect of treating the etched/unetched lithium disilicate ceramic surface with various types of silane coupling agents on the contact angle measurement of deionized water. The null hypothesis was that the different types of silane coupling agents used would not affect the contact angle formed between the deionized water droplets and the etched/unetched lithium disilicate glass ceramic surfaces.

Materials and Methods

Lithium disilicate disks preparation

One hundred fifty lithium disilicate disks (IPS e.max Press, Ivoclar-Vivadent, Schaan, Liechtenstein) were prepared by waxing up disk-shaped samples with diameters of 10 millimeters and heights of 3 millimeters. The lost wax technique and heat-pressed processes were performed according to the manufacturer's instructions.

The samples were polished with 1000-grit silicon carbide abrasive paper[21] using a polishing machine (Minitech 233, Presi, Le Locle, Switzerland), spinning in clockwise motion at 200 rounds/minute for 5 minutes with a pressure of 2 kg/cm^2 under running water. The abrasive papers were replaced between samples. After polishing, the samples were ultrasonically cleaned in distilled water for 10 minutes using an ultrasonic cleaner (Ultrasonic cleaner VI, Yoshida dental trade distribution Co., Tokyo, Japan).

Setting up the experimental groups

The samples were randomly assigned into ten groups (n=15) based on the type of silane coupling agent used to treat the surface of lithium disilicate and whether the sample was acid-etched. (Table 1);

Table 1 *List of materials used*

Product name	Types	Compositions	Manufacturer/Supplier
IPS e.max Press (Lot Y10318)	Lithium disilicate glass ceramic	SiO_2, Li_2O, K_2O, P_2O_5, ZrO_2, ZnO, other oxides and ceramic pigments	Ivoclar-Vivadent, Schaan/Liechtenstein
Kerr silane primer (Lot 7072259)	Silane	Ethanol, (1-methylethylidene) bis[4,1-phenyleneoxy(2- hydroxy-3, 1-propanediyl)] bismethacrylate Poly(oxy-1,2-ethanediyl), α,α'-[(1-methylethylidene)di-4,1-phenylene] bis[ω-[(2- methyl-1-oxo-2-propen-1-yl)oxy]-2,2'-ethylenedioxydiethyl dimethacrylate 3-trimethoxysilylpropyl methacrylate	Kerr corporation, West Collins Avenue Orange, California, USA
Monobond N (Lot X41367)	Silane	Ethanol, methacrylated phosphoric acid ester, sulphide methacrylate, 3-trimethoxysilylpropyl methacrylate	Ivoclar-Vivadent, Schaan/Liechtenstein
RelyX ceramic primer (Lot N988623)	Silane	Ethanol, water, methacryloxypropyltrimethoxysilane	3M ESPE Dental products, Conway Avenue St. Paul, Minnesota, USA
Experimental silane: 3(Trimethoxysilyl) propyl methacrylate (Lot SHBJ3136)	Silane	Water, ethanol, acetic acid, methacryloxypropyltrimethoxysilane	Sigma-Aldrich, Missouri, USA
IPS ceramic etching gel (Lot Y06707)	Hydrofluoric acid	4.5% Hydrofluoric acid	Ivoclar-Vivadent, Schaan/Liechtenstein

Group 1. No silane (NS), serves as a control group

Group 2. Kerr silane primer (KP)

Group 3. Monobond N (MN)

Group 4. RelyX ceramic primer (RX)

Group 5. Experimental silane (ES)

Group 6-10 were identical to Groups 1-5 in terms of the silane coupling agent used; however, these groups were etched with 4.5% hydrofluoric acid gel (IPS ceramic etching gel, Ivoclar-Vivadent, Schaan/Liechtenstein) for 20 seconds, rinsed with deionized water spray for 60 seconds and gently air dried[12] prior to application of respective silane coupling agents.

The experimental silane was prepared by mixing a solution of 95% ethanol / 5% distilled water in a beaker. The pH of the solution was adjusted to 4.5-5.5 with acetic acid using a digital pH meter (Orion 420A pH meter, Thermo Electron Corporation, Massachusetts, USA). The solution was transferred to a plastic bottle and silane coupling agent (3-(Trimethoxysilyl)propyl methacrylate, Sigma-Aldrich, Missouri, USA) was added with stirring to yield a 2% final concentration. The solution was left untouched for five minutes allowing the hydrolysis and silanol formation. A magnetic stirrer and bar (Hotplate stirrer UC152, Stuart Scientific, Staffordshire, UK) was used to gently mix the solution for 1-2 minutes.

A drop of the respective silane coupling agent was applied to each sample using a micropipette (10 microliters) and smeared into thin coat using a microbrush (Citisen Micro Applicator, Huanghua Promisee Dental, Hebei, China). After silane application, the treated samples were left untouched, allowing the silane to react with the disk surfaces according to the manufacturers' instruction. New microbrushes were used to remove any remaining excess around the borders of the samples. Then, the sample were air-dried for ten seconds using a triple syringe from a mobile dental unit (10 millimeters from the sample, pressure 40-50 pound per square inch). Before proceeding to the next step, the samples were checked to make sure that the surface was completely dried (no movement of solution).

Sessile drop test, contact angle measurement

The degree of wettability was determined by contact angle measurement. Using a needle, ten microliters of deionized water was placed on the center of the treated/ untreated substrate surface to examine the contact angle formed between the deionized water droplet and the prepared substrate. The contact angles were measured digitally with a goniometer (DSA10 MK2, Krüss, Hamburg, Germany) after five seconds. For each drop, the angles obtained from both ends of the captured image were averaged and the mean values of each tested group recorded.

Data analysis

The contact angle between deionized water and the substrate surface are presented as mean ± SD. The statistical analysis of the contact angles of all groups were performed using SPSS 20.0 software for Windows (SPSS Inc, Chicago, Illinois, USA). The data were normally distributed and Two-way analysis of variance was applied. Tukey's multiple comparison tests were conducted to determine the significant differences between all treatment groups (α=0.05).

Results

Figure 1 shows contact angle images for all experimental groups. The average contact angles obtained from each respective silane groups are shown in Table 2.

The non-etched samples (G1-5) generally yielded larger contact angles than the etched samples (G6-10). In the unetched groups, the control group exhibited the smallest contact angle (G1, 17.02°), while the Monobond N group showed the largest contact angle (G3, 46.37°). Kerr silane primer exhibited a larger contact angle (G2, 38.99°) than both RelyX ceramic primer (G4) and the experimental silane group (G5), which were relatively similar (32.09° and 32.53°, respectively). There were significant differences between all tested groups except RelyX ceramic primer and the experimental silane group.

In hydrofluoric acid-etched groups, the control group also exhibited the smallest contact angle (G6, 8.23°),

while Kerr silane primer demonstrated the largest contact angle (G7, 38.82°). The Monobond N group showed significantly larger contact angles (G8, 20.63°) than both RelyX ceramic primer (G9) and the experimental silane group (G10), which were relatively similar (13.53° and 13.29°, respectively).

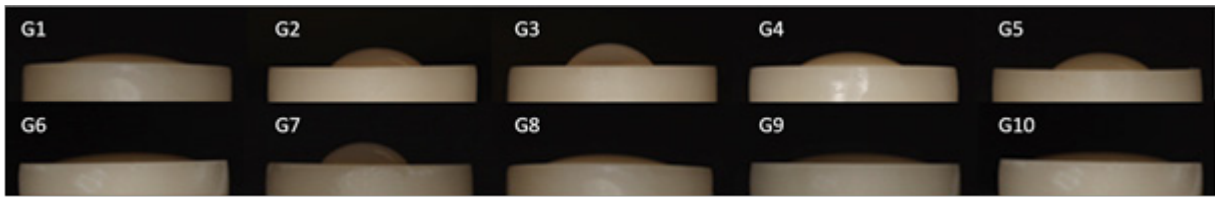

Figure 1 *Contact angle of deionized water on etched and unetched lithium disilicate disks treated with different types of silane coupling agent. Group without hydrofluoric etching: (G1) Control, (G2) Kerr silane primer, (G3) Monobond N, (G4) RelyX ceramic primer, (G5) Experimental silane primer. Group with hydrofluoric etching prior to silane application: (G6) HF-no silane, (G7) HF-Kerr silane primer, (G8) HF-Monobond N, (G9) HF-RelyX ceramic primer, (G10) HF-Experimental silane*

Table 2 *Average contact angle of respective silane group in degree (°) on etched and unetched lithium disilicate glass*

Silane coupling agents (n=15)	Mean contact angle Mean (SD)	
	Unetched disks	Etched disks
Control	17.02 (2.42)[a]	8.23 (1.11)[e]
Kerr silane primer	38.99 (3.76)[b]	38.82 (3.54)[b]
Monobond N	46.37 (4.24)[c]	20.63 (1.87)[f]
RelyX ceramic primer	32.09 (3.80)[d]	13.53 (1.42)[g]
Experimental silane	32.53 (3.29)[d]	13.29 (1.51)[g]

Values with different letters superscripted vary significantly.

Discussion

Based on the results of this study, there were significant differences in the contact angle formed between the deionized water and the etched/unetched lithium disilicate glass ceramic surface treated with various types of silane coupling agent. Therefore, the null hypothesis was rejected. Many silane coupling agent brands are not composed of pure MPS but of a mixture of MPS and other adhesive substances. Recent studies have suggested that the simplified systems, using combinations of different functional groups along with other components, may reduce the bond strength of glass ceramic and resin cement when compared to the conventional two-bottle system.[15,22]

Contact angle values indicate the wettability of a surface and can be used to calculate the surface energy.[18] Contact angle is defined as the angle where the liquid/vapor interface meets a liquid interface/solid surface. The degree of wettability depends upon the surface energy (surface tension) of the interfaces involved such that the total energy is minimized. Several methods can be used to measure contact angle of surfaces (static sessile drop, dynamic sessile drop, dynamic Wilhelmy, or single-fiber Wilhelmy method). In this study, static sessile drop was used due to its convenient operation and popularity.[23]

The lithium disilicate disks were polished with 1000-grit silicon carbide abrasive paper to standardize the roughness of the disks' surfaces. Alteration of surface topography (e.g. grinding, acid-etching, air-borne particle abrasion) can physically contribute to the adhesion pro-

cess by altering the surface area and wetting behavior of ceramic which in turn affects the surface energy and adhesive potential to resin.[17,24]

Selecting a probing medium is an important aspect when measuring the contact angle. Liquid of known surface tension such as 1-bromo-naphthalene (44.4 mJ/m^2), diiodomethane (50.8 mJ/m^2) and water (72.8 mJ/m^2) are commonly used.[25] However, selecting an appropriate probing liquid depends on the objective of the study.[18] If the purpose was to examine the wettability of silane coupling agent on a ceramic surface, the respective silane coupling agent should be used as the probing liquid. In this study, deionized water was used as a probing liquid to investigate the differences between the silane-treated substrate surfaces.

Based on the results obtained in this study, in the non-etched lithium disilicate group, all silanated surfaces (G2-5) exhibited significantly larger contact angles than the control group (G1). This result suggests that application of silane coupling agents may lower the surface energy of the substrate. Similar findings were observed in a study by Della Bona *et al.*[17] which conducted an experiment using lithium disilicate-based ceramic treated with different protocols including application of silane coupling agent before measuring the contact angle. The author explained that treating the ceramic surface with silane makes the surface hydrophobic. The hydrophobic property may reduce hydrolytic degradation of the bond and would also promote the wetting of adhesive. In his study, 8 % methacryloxy propyl trimethoxy silane (MPTMS) was used to treat the ceramic surface. High-performance liquid chromatography grade water (HPLC water) and liquid resin of known surface tension were used as the probing liquid (72.6 mN/m and 39.7 mN/m, respectively). However, when the silane coupling agent was applied and the liquid resin was dropped, 'beads up' of the liquid resin was observed on the silanated ceramic surface. The author explained that for an adhesive to completely wet the substrate surface, it must be of low viscosity and the surface tension of the adhesive must be lower than the critical surface energy of the substrate. Another study by Farge *et al.*[26] also demonstrated the relationship of surface energy/tension to its wetting property. The author used different adhesive systems that differed in the com-position of the solvent. It was reported that the liquid (solvent) with lower surface tension had better wettability than ones with higher surface tension; for example, ethanol (22.4 mN/m) shown superior wetting property than ethanol-water.

The contact angle shown in the pure silane groups, RelyX ceramic primer (G4) and experimental silane (G5), were not significantly different from one another but were significantly smaller than groups with additives, Kerr silane primer and Monobond N. Additional components other than MPS may result in an increase or decrease in the contact angle. In the current study, silane with additives produced larger contact angles than ones without additives. The additives such as extra resins in Kerr silane primer, which was meant to eliminate the bonding steps following the priming procedure or the phosphate-containing and sulfide methacrylate monomers in Monobond N and those were believed to promote chemical adhesion with various substances, may alter the polarity of substrate surfaces and/or the surface energy leading to an increase in contact angle. In a study by Chen *et al.,*[27] the application of Monobond Plus phosphate-containing monomer and sulfide monomer on zirconia significantly increased the contact angle of deionized water when compared to the unconditioned surface (15.1° to 74.1°). However, the author believed that incorporating BisGMA resins to silane coupling agents may reduce the contact angle formed between deionized water and the silanated lithium disilicate surface. The author explained that the extra resins might inhibit the condensation reaction of silane coupling agent, thus, lowering both the contact angle and bond strength.

In the hydrofluoric acid-etched lithium disilicate groups (G6-10), all treatment groups exhibited lower contact angles than the unetched group, except for Kerr silane primer group (G7). Generally, the results can be explained by the effect of acid-etching. Acid-etching

altered the surface topography of the samples.[17] The total surface area and surface energy were increased in the roughened surfaces allowing them to draw more medium onto their surface, increasing the wettability of the substrate. Ramakrishnaiah *et al.*[21] studied the effect of hydrofluoric acid etching duration on silica-based ceramic. The study showed that increasing the etching duration significantly altered the surface topography. The longer the duration, the rougher the surface. Increasing etching duration increases surface roughness and wettability which in turn lowers the contact angle. However, when Kerr silane primer was applied on the etched-surfaces, a thin layer of resin was formed on the substrate's surface. This layer of resin may have filled the pits created from the etching process which may have masked the roughening effect of hydrofluoric acid.

The degree of surface wettability and surface energy may contribute to the improvement of bond quality; however, its physical contribution is not the only factor. Adhesion of dental ceramics to resin based material is the result of physico-chemical interactions between the substrate and adhesive.[18] A clean and dry surface of the restoration is a prerequisite to create a proper bond with the adherend,[28, 29] as surface contamination or surface impurity can reduce the surface energy of the substrate,[24] and thus have a negative effect on the quality of bond. In a study by Tani *et al.*,[29] it was proven that a surface with superior wettability and surface energy might not be able to provide optimal bond. Therefore, chemical adhesion also plays an important role in obtaining good bond quality. As long as the bonding site is clean and has a sufficient amount of Si-OH site on the ceramic surface, a reliable bond is achievable.[24] Further studies should investigate the relationship between contact angles and the shear bond strength of silanated glass ceramic surfaces.

Although treating the lithium disilicate surfaces with silane coupling agent may seem to reduce the wettability of the substrate surface to deionized water, hydrofluoric acid etching along with silane application are proven to be the gold standard treatment protocols that are crucial to obtain optimal bond.[30,31]

Conclusions

Within the limitations of this in vitro study, the following can be concluded:

1. Silane coupling agents significantly reduced the wettability of deionized water on treated lithium silicate surfaces.
2. The types of silane coupling agent significantly influence the degree of wettability.
3. Hydrofluoric acid etching generally significantly increased the wettability, except for groups that were treated with resin-containing silane primer.

Acknowledgements

We would like to thank Chulalongkorn Dental materials R&D center and Oral biology research center staffs for assisting in preparation of the experimental silane. Thanks to Lily See for proofreading the article.

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