Caries-preventive Activity of Fluoride-containing Resin-based Desensitizers

S Sohn • K Yi • HH Son
J Chang

Clinical Relevance

Resin-based desensitizing agents may serve as persistent mechanical barriers and prevent the development of root caries.

SUMMARY

Objective: The purpose of this study was to evaluate the effects of different desensitizing agents on the prevention of root caries when applied to root surfaces.

Materials and Methods: Thirty human roots were sectioned into quarters with a 3 × 4 mm window. A desensitizer (VX, Clinpro™ XT Varnish; SP, Seal & Protect®; or PB, Clearfil™ Protect Bond) was applied to three of the quarters in each window. Teeth were stored separately in water for one day, 30 days, or 60 days. The remaining quarter, without the application of desensitizer, served as a control. After storage in water, all specimens were subjected to pH cycling. Scanning electron microscopy was used to observe the demineralization bands created on the subsurface layer. The weight percentages of fluorine (F), silica, and calcium (Ca) were determined using electron probe microanalysis to quantify the elemental distributions in the root dentin. The concentrations of F released during a pH cycling were measured.

Results: For the control group, the average lesion depth was 18.92 ± 5.42 μm, and the average Ca loss was 15.66% ± 6.80% in the superficial layer and 30.44% ± 9.61% in the subsurface layer. No Ca loss occurred in the desensitizer-treated groups. All desensitizing agents remained intact for at least 60 days. F levels were increased in the hybrid layer but not in the subhybrid area. Outward release of F diminished with time.

Conclusion: The F-containing resin-based desensitizers protected exposed root surfaces

Suhjin Sohn, DDS, MSD, Department of Conservative Dentistry, School of Dentistry, Seoul National University, Dental Research Institute, School of Dentistry, Seoul National University, Seoul, Korea

Keewook Yi, MS, PhD, Geochronology Team, Korea Basic Science Institute, Daejeon, Korea

Ho- Hyun Son, DDS, MSD, PhD, Department of Conservative Dentistry, School of Dentistry, Seoul National University, Dental Research Institute, School of Dentistry, Seoul National University, Seoul, Korea

*Juhea Chang, DDS, MSD, PhD, Clinic for Persons with Disabilities, Seoul National University Dental Hospital, Dental Research Institute, School of Dentistry, Seoul National University, Seoul, Korea

*Corresponding author: 62-1 Changgyeonggungno, Jongro-gu, Seoul, 110–768, South Korea; e-mail: juhchang@snu.ac.kr

from demineralization. F liberated from the desensitizers was detected only at minimal levels.

INTRODUCTION

Exposed root dentin often contributes to the clinical signs of dentin hypersensitivity.[1] Dentin hypersensitivity can be treated by covering the exposed tubules, thereby preventing the transmission of pain-causing stimuli to pulpal nerve fibers. Denuded root surfaces have a large proportion of organic material, which makes these surfaces more soluble than enamel in acidic environments, resulting in increased susceptibility to caries development.[2,3] Fluoride-containing gels or varnishes are good choices for treatment of exposed roots with dentin hypersensitivity. Specifically, the creation of a calcium fluoride (CaF_2) barrier on root surfaces not only provides a fluorine (F) reservoir but also blocks the patent tubules. To maintain a formidable barrier composed of calcium (Ca) and F, topical F needs to be reapplied to the root surface on a regular basis.[4] For this reason, patient compliance is essential for successful outcomes using this treatment modality.[5]

F-containing resin-based desensitizers were introduced recently as an adjunctive option for the prevention of root caries. The rationale for the use of F-containing desensitizers for the prevention of caries development is that these types of desensitizers enhance the chemical resistance of dentin to mineral dissolution. Additionally, the polymerized resin layer of these desensitizers is expected to provide a physical shield for a prolonged period of time.[6] In this study, three commercial desensitizers were applied to the root dentin, and the effectiveness of these desensitizers as caries-protective agents was examined under conditions of mild acidic challenge. Scanning electron microscopy (SEM) was used to examine the extent of dentin demineralization. The levels of F, silica (Si), and Ca content were measured in the cross-sectioned surfaces of dentin that had been treated with desensitizers using electron probe microanalysis (EPMA). In order to compare the F-releasing abilities of the desensitizers, F concentrations were also assessed after aging at one day, 30 days, and 60 days.

The aims of this study were 1) to determine whether the root dentin treated with F-containing resin-based desensitizers consistently resists acidic challenge and 2) to quantify the amount of the F ions released from these desensitizers in two directions: outward liberation and inward penetration.

MATERIALS AND METHODS

Specimen Preparation

This study was approved by the Institutional Review Board of the Seoul National University Dental Hospital. Thirty human lower premolars extracted during orthodontic treatment were used within six months of extraction. The teeth were disinfected in 0.5% chloramine-T for a week and stored in distilled water at 4°C. The teeth were inspected to ensure that they were free of fractures or other defects. Cementum was removed using polishing discs (Sof-Lex, 3M ESPE, St Paul, MN, USA) under microscopy. The crowns of these teeth were removed at the cementoenamel junction using a low-speed diamond saw (Isomet™, Buehler Ltd, Lake Bluff, IL, USA), and the apical roots were also removed leaving 5-mm-long root segments. The segments were sectioned mesiodistally and buccolingually into four parts (Figure 1). Each of the root quarters was covered with a thin layer of acid-resistant nail varnish, with the exception of a 3 × 4 mm window on the outer root surface. Three of these window surfaces were coated using one of the following materials: Clinpro™ XT Varnish (3M ESPE; designated the VX group), Seal & Protect® (Dentsply, Konstanz, Germany; designated the SP group), or Clearfil™ Protect Bond (Kuraray, Okayama, Japan; designated the PB group). Additionally, one control window surface was left free of desensitizer (CC group: no treatment) (Table 1).

Following light curing, each of the 10 specimens in the three experimental groups was stored in distilled water for one day, 30 days, or 60 days, respectively. The 10 control specimens were stored in distilled water for one day. Specimens were then immersed in 10 mL of demineralizing solution (1.5 mM $CaCl_2$, 0.9 mM KH_2PO_4, and 50 mM acetate buffer, pH 4.8) for three hours, followed by immersion in 10 mL of remineralizing solution (1.5 mM $CaCl_2$, 0.9 mM KH_2PO_4, and 20 mM 4-(2-hydroxyethyl)-1-piperazineethanesulfonic acid, pH 7.0) for 20 hours. During a five-day cycle, each specimen in solution was placed in a shaking incubator (SI-600R, JEIO TECH, Korea) at 37°C with agitation. Solutions were changed every day and stored at 4°C.

SEM/EPMA Analysis

Specimens were embedded in epoxy resin (Epofix, Struers, Glasgow, UK) and horizontally cross-sectioned along the midline. The exposed cut surfaces were serially polished with 1200, 2400, and 4000 grit
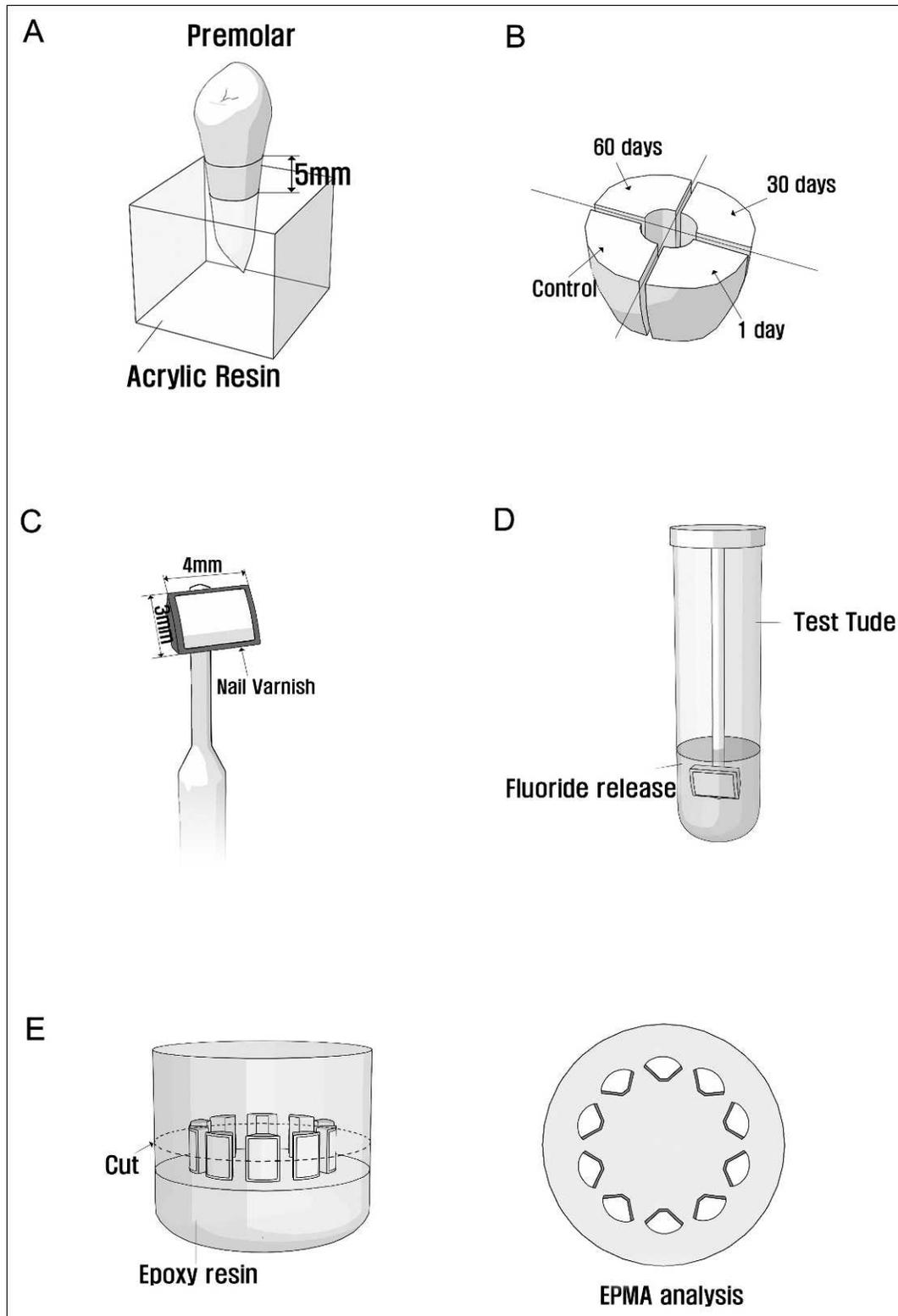
Figure 1. *Schematic diagrams of specimen preparation. (A) A human lower premolar was sectioned to produce a 5-mm-long root segment. (B) Each quarter segment was divided into one control and three treated groups. (C) Each quarter was painted with nail varnish, leaving a 3 × 4–mm window on the root surface. (D) After treatment, each specimen was immersed in pH cycling solution. (E) After cycling, specimens were embedded in epoxy resin and cross-sectioned at the midline for EPMA analysis.*

Table 1:	*Materials Used in the Study*		
Product	**Component**		**Application**
Group VX: Clinpro™ XT Varnish (3M ESPE, St Paul, MN, USA)	Copolymer of acrylic and itacoic acids, water, HEMA, silane-treated glass, silane-treated silica, Bis-GMA		Etch for 15 seconds with 35% phosphoric acid (3M™ ESPE™ Scotchbond™ Etchant). Rinse for 15 seconds. Mix paste/liquid components together rapidly (10–15 seconds). Apply a thin layer (1/2 mm or less) of the mixed material to the tooth surface. Light-cure for 20 seconds.
Group SP: Seal & Protect® (Dentsply, Konstanz, Germany)	Di- and trimethacrylate resins, PENTA, functionalized amorphous silica, photoinitiators, butylated hydroxytoluene, cetylamine hydrofluoride, triclosan, acetone		Apply Seal & Protect to the dentin surface and leave it undisturbed for 20 seconds. Remove excess solvent by gentle air-blowing. Light-cure for 10 seconds. Apply a second layer of Seal & Protect and remove excess solvent from the second layer by gentle air-blowing. Light-cure for 10 seconds.
Group PB: Clearfil™ Protect Bond (Kuraray, Okayama, Japan)	PRIMER	MDP, MDPB, HEMA, hydrophilic dimethacrylate, water	Apply PRIMER to the dentin surface and leave it in place for 20 seconds. Remove excess solvent by gentle air-blowing. Apply BOND and create a uniform bond film using a gentle air flow. Light-cure for 10 seconds.
	BOND	MDP, Bis-GMA, HEMA, hydrophobic dimethacrylate, DL-camphorquinone, N, N-diethanol-p-toluidine, silanated colloidal silica, surface-treated sodium fluoride	

Abbreviations: Bis-GMA, bis-phenol A diglycidylmethacrylate; HEMA, 2-hydroxyethyl methacrylate; MDP, 10-methacryloyloxydecyl dihydrogen phosphate; MDPB, 12-methacryloyloxydodecylpyridinium bromide; PENTA, dipentaerythritol penta acrylate monophosphate.

aluminum oxide (Al_2O_3) abrasive papers, followed by 1 μm and 0.25 μm diamond and 0.1 μm and 0.05 μm alumina polishing suspensions (Struers, Copenhagen, Denmark). The specimens were ultrasonically cleaned in deionized water for 10 minutes, dried for 72 hours in a desiccator, and then sputter-coated with carbon. Demineralization bands on the cross-sectioned surfaces were identified at a magnification of 600× using the phase contrast of backscattered electron imaging mode of SEM (JEOL JSM-6610LV, JEOL, Akishima, Japan). To identify variations in the amounts of the specified elements from the outer dentin to the inner dentin, two scans were performed perpendicular to the outer surface at 0.3 μm pixel intervals. The observation areas (superficial layer, demineralized layer, sound dentin, desensitizer layer, and hybrid layer) were determined according to changes in Ca, F, and Si content using EPMA (JEOL JXA-8100, JEOL). The operating conditions for both the image and elemental analyses were 15 kV of accelerating voltage and 50 nA of beam current. Measurements along the scan were averaged into a single value for each area. The F content was expressed as the percentage of weight relative to

the total weight of a standard material where the measurement was taken (Table 2). A fluorapatite crystal (3.38% F) was used as a standard comparison for F.

Table 2:	*The Mean Weight Percentage (Standard Deviation, SD) of Fluoride in the Desensitizer Layer[a]*			
Aging Period, d	**N**	**Group**		
		VX Desensitizer Layer	**SP Desensitizer Layer**	**PB Desensitizer Layer**
1	20	11.94 (2.39) A	1.01 (0.45) A	1.32 (0.25) A
30	20	11.88 (2.74) A	0.67 (0.13) B	1.23 (0.13) A
60	20	10.77 (2.63) A	0.56 (0.13) B	1.30 (0.13) A

Abbreviations: PB, Clearfil™ Protect Bond; SP, Seal & Protect®; VX, Clinpro™ XT Varnish.
[a] Online small-capital letters denote values that are not significantly different from one another in each column ($p < 0.05$).
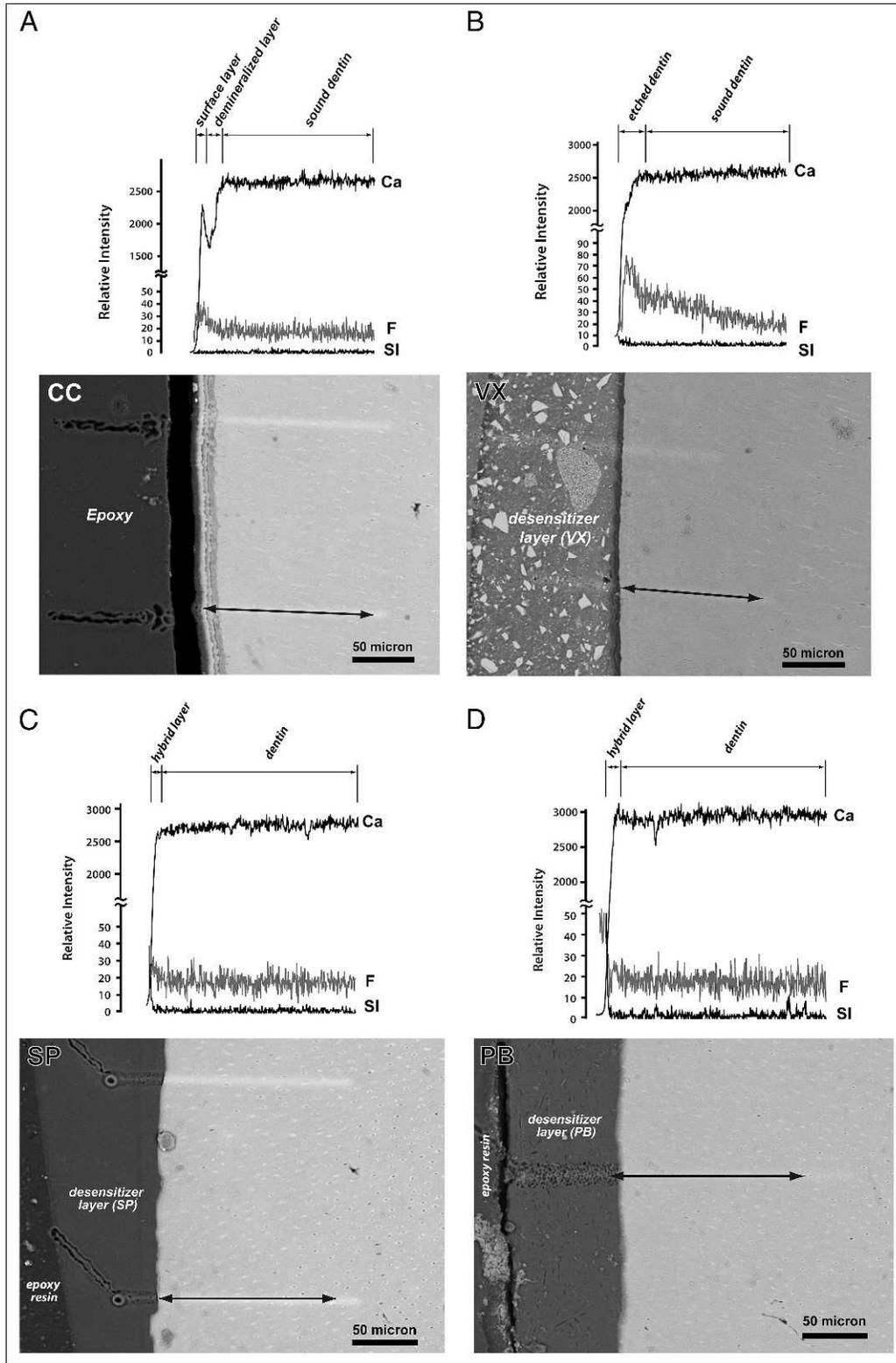
Figure 2.   *SEM images and EPMA graphs were compared for each experimental group at 60 days of aging (600× magnification). (A) CC group. Elemental analysis along the scan line (dark arrow) illustrated the differentiated elemental composition (Ca, F, Si) of root dentin. Repeated demineralization bands were produced by a pH cycling series. Note that Ca level decreased in the superficial layer, followed by a sharp decline in the subsurface demineralized layer. F was detected at minimal amounts throughout the sound dentin, while no detectable Si was observed. (B) VX group. In a thickly built-up desensitizer layer, RMGI displayed various sizes of fillers embedded in matrix. No demineralization bands were observed in*

Fluoride Concentration Measurement

A fluoride-specific ion electrode connected to a digital pH/millivolt meter (Accumet© Research Model AR 25, Thermo Fisher Scientific, Beverly, MA, USA) was calibrated using F reference solutions (1.0 and 10.0 ppm). The demineralization solution (4 mL) was mixed with total ionic strength adjustment buffer (TISAB II; 4 mL; Thermo Fisher Scientific) to obtain a constant background ionic strength. The mixed solution was placed over a nonheating magnetic stirrer, and readings were taken after a five-minute immersion period. The temperature of the solution was maintained at 22°C. Sample readings were obtained in µg F/mL (ppm F).

Statistical Analysis

The normality and homogeneity of the samples were tested using the Kolmogorov-Smirnov test. The weight percentages of F in the desensitizer layer and the hybrid layer were compared among all groups using two-way analysis of variance (ANOVA) and the Tukey *post hoc* test. The F released during demineralization cycling was compared among groups using repeated-measures ANOVA with mixed model. A *p*-value of 0.05 was selected as the threshold for statistical significance. Analyses were performed using SAS 9.1.3 (SAS Institute, Cary, NC, USA).

RESULTS

SEM revealed the formation of demineralization bands under the root surfaces in the control group (CC group) (Figure 2A). The width of the surface layer was 4.92 ± 0.92 µm, and that of the subsurface demineralized layer was 13.97 ± 4.44 µm. In the desensitizer-treated groups (VX, SP, and PB), no demineralized layers were observed at any stage of aging (one day, 30 days, or 60 days). Additionally, the desensitizer layer covering the dentin surface remained intact even at 60 days.

Line analyses revealed the individual distributions of Ca, F, and Si, and it was possible to distinguish the desensitizer layer from the dentin (Figure 2B-D). In the CC group, the average Ca loss was $15.66\% \pm 6.80\%$ in the superficial layer and $30.44\% \pm 9.61\%$ in the subsurface layer. In the experimental groups (VX, SP, and PB), Ca levels were maintained from the surface layer to a depth of 200 µm without any discernable changes. Si levels were elevated in the desensitizer layer of each of the three experimental groups. However, these levels abruptly dropped to near zero at the dentin interface. In the desensitizer layer, F levels were significantly higher in the VX group ($p<0.001$). Ca levels started to rise at the dentin interface and reached a plateau at the inner dentin layer. In the SP and PB groups, there were transitional areas or hybrid layers in which a decrease in Si levels and an increase in Ca levels overlapped (Figure 2C,D). Unlike the SP and PB groups, in the VX group no Si was detected in the etched dentin layer. However, a significantly higher level of F was detected in the etched dentin area of the VX group than in the hybrid layers of the SP and PB groups ($p<0.001$). F levels were significantly different in the desensitizer layers of the SP group (between one day and 30 days; Table 2). The hybrid layers showed a significant change in F levels in the CL group (between one day and 30 days; Table 3). The mean widths of the etched dentin layer of the VX group and the hybrid layers of the SP and PB groups are listed in Table 4.

Table 5 summarizes the concentrations of F released during each demineralizing cycle. F release was the highest on the first day and significantly decreased throughout the duration of cycling ($p<0.001$; Figure 3). There was also a significant difference in F release between the one-day and 30-day time points for each of the experimental groups (VX, $p=0.022$; SP, $p=0.036$; and PB, $p<0.001$). The VX group released significantly more F than did the SP and PB groups at one day and 30 days ($p<0.001$), and the SP and PB groups did not differ significantly at either time point. The level of released F was not obtained from the SP and PB groups at 60 days.

DISCUSSION

Fluoride has been shown[7] to be one of the materials used to decrease the permeability of dentin, possibly by precipitation of insoluble calcium fluoride within the tubules. The fluoride in the desensitizers examined in this study was expected to result in

←

dentin. Ca levels started rising in the etched dentin layer, leveling off in the sound dentin. F and Si in the desensitizer layer were detected at elevated levels but were not depicted in the above graph. In the etched dentin, only F was maintained at an increased level compared to the sound dentin. (C) SP group. The desensitizing layer was protecting the dentin surface well, without signs of demineralization. Ca started from the zero level, abruptly increased, and leveled off in the sound dentin area. In contrast, Si started decreasing at the beginning of the hybrid layer and disappeared in the sound dentin. F showed a similar pattern to Si in the hybrid layer. (D) PB group. A well-maintained desensitizer layer covered the dentin surface. No demineralization occurred. Ca, F, and Si showed the same pattern as in the SP group.

Table 3:		*The Mean Weight Percentage (Standard Deviation, SD) of Fluoride in the Hybrid Layer[a]*		
Aging Period, d	**N**	**Group**		
		VX **Etched Dentin Layer[b]**	**SP** **Hybrid Layer**	**PB** **Hybrid Layer**
1	20	1.94 (0.51) A	0.88 (0.49) A	0.90 (0.18) A
30	20	2.14 (0.88) A	0.73 (0.55) A	0.66 (0.23) B
60	20	2.00 (0.68) A	0.65 (0.38) A	0.67 (0.31) B

Abbreviations: PB, Clearfil™ Protect Bond; SP, Seal & Protect®; VX, Clinpro™ XT Varnish.
[a] Online small-capital letters denote values that are not significantly different from one another in each column (p<0.05).
[b] No hybrid layer was determined in the VX group. Instead, the etched dentin layer in the VX group was compared with the hybrid layers in the SP and PB groups.

Table 4:		*The Mean Width (Standard Deviation, SD) of the Superficial Layer and the Demineralized Layer in the Control Group (CC) and of the Hybrid Layer in the Experimental Groups (VX, SP, and PB) (μm)*			
N	**Group**				
	CC		**VX**	**SP**	**PB**
	Superficial Layer	**Demineralized Layer**	**Etched Dentin Layer**	**Hybrid Layer**	**Hybrid Layer**
60	4.92 (0.92)	13.97 (4.44)	16.63 (1.91)	4.24 (1.12)	4.41 (1.07)

Abbreviations: CC, no treatment; PB, Clearfil™ Protect Bond; SP, Seal & Protect®; VX, Clinpro™ XT Varnish.

tubule obturation and caries inhibition. In addition, a resinous coating of the desensitizers is capable of blocking the acid transportation into the root dentin.[4] The first aim of our study was to determine whether the application of the desensitizers to the root surfaces is protective against the effects of exposure to an acidic environment. We subjected freshly exposed root dentin to a mild acidic challenge and simulated a previously low- to moderate-caries risk group that became newly susceptible to the disease as a result of recently uncovered root surfaces. The use of pH cycling produced multiple demineralization bands underneath a relatively intact superficial layer, implying that a series of dynamically balanced reactions had occurred (Figure 2). The desensitizer-coated dentin showed no signs of demineralization, indicating that the coating material provided a complete seal against the effects of acidic challenge. The acid-resistant protection remained for 60 days in all three desensitizer groups. VX, a resin modified glass ionomer (RMGI)-based desensitizer, had a thick consistency and produced thicker layers than did SP, a desensitizer based on one-step self-etch resin adhesive, and PB, a two-step self-etch resin adhesive. Many *in vitro* analyses of artificial caries have been performed under conditions of severe acidic challenge (immersion in strong acid without the use of a remineralizing cycle).[8–10] When the polymerized resin barrier is thick, less soluble, and water stable, it is able to prevent acid penetration.[9] Clinically, it is rare to use only adhesive resin without consecutive build-up of

composite resin and to directly expose adhesive resin-applied dentin to an aqueous environment. We wondered whether a desensitizer, composed largely of solvent and hydrophilic monomers, would resist hydrolytic degradation. SEM images showed that all three desensitizers remained intact with a minimum thickness of 40–50 μm. Additionally, relatively mild pH cycling did not result in disintegration of the surface coating layer during the 60 days of water storage. The mechanical impact induced by toothbrush abrasion on resin-adhesive bonded root surfaces should be considered in future studies.

Another purpose of our study was to quantify the amount of F released from the solidified desensitizer layer. The manufacturers of the three desensitizers claim that their supplementation with F has the benefit of caries prevention. However, the resin-based desensitizers used in this study may not contain as much F as conventional F varnishes, which often contain 5% sodium fluoride (22,600 ppm F).[4] Furthermore, the way in which sodium F in CL and amine F in SP dissociate from the photopolymerized resin composite and dissolve in water is unclear. If the F were to dissolve, surface wash-off processes could be induced by water sorption of hydrophilic monomers. In this case, the resin matrix may provide an aqueous environment to facilitate the transport of F ions.[11] In RMGI, an acid-base reaction enhances the leaching of F ions to form a polysalt matrix, enabling the release of more F than is released from resin adhesives with F-incorporated filler particles.[12] However, even RMGI has a limited span of rapid F release, with a significant decrease within an initial period.[13] The amount of F released

Table 5:	*The Mean Daily Fluoride Release (Standard Deviation, SD) During a Demineralization Cycle at One Day and 30 Days (10^{-1} ppm)*		
Aging Period, d	**pH Cycle**	**Group**		
		VX	**SP**	**PB**
1	Day 1	3.78 (1.45)	0.64 (0.04)	0.83 (0.02)
	Day 2	3.18 (1.23)	0.53 (0.01)	0.65 (0.03)
	Day 3	2.73 (0.89)	0.50 (0.01)	0.64 (0.04)
	Day 4	2.42 (0.61)	0.50 (0.00)	0.57 (0.03)
	Day 5	2.31 (0.48)	0.49 (0.00)	0.54 (0.01)
30	Day 1	3.31 (0.44)	0.61 (0.05)	0.62 (0.01)
	Day 2	2.07 (0.59)	0.49 (0.01)	0.52 (0.01)
	Day 3	1.15 (0.11)	0.48 (0.01)	0.51 (0.01)
	Day 4	1.10 (0.10)	0.47 (0.01)	0.51 (0.01)
	Day 5	1.06 (0.09)	0.48 (0.15)	0.49 (0.01)

Abbreviations: PB, Clearfil™ Protect Bond; SP, Seal & Protect®; VX, Clinpro™ XT Varnish.
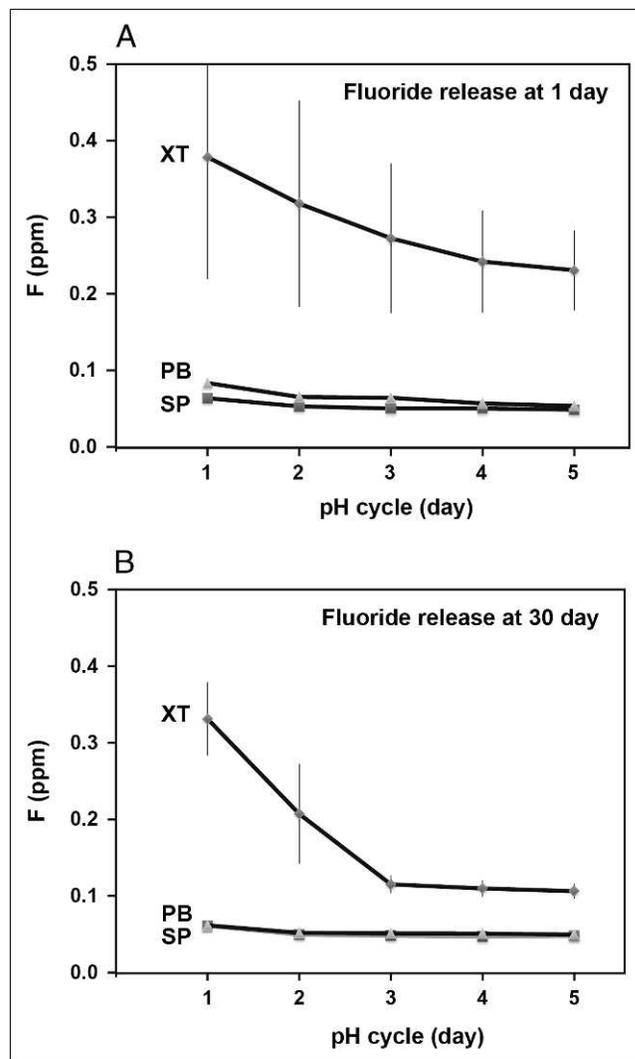

Figure 3. *Concentration of F released during a demineralizing cycle was measured at one day and 30 days. VX released significantly more fluoride than did SP and PB at each cycle at both time points (*p<*0.001).*

in *in vitro* models is largely dependent on the volume of the material used, the size of the exposed surface, the duration of immersion, and the pH of the solution.[14,15] Each quarter root of human premolar only had a 3×4–mm surface area exposed and allocated to each of four groups. Furthermore, the demineralization solutions were changed at each cycle, which is more similar to a clinical situation than to cumulative collection. The short duration (three hours) of the demineralizing cycle allowed a very small amount of F to be leached, and levels were below levels of detection for the SP and PB groups at 60 days. Previous studies have often used cumulative concentrations of F in order to obtain the viable data for statistical analysis. This is especially true when low-concentration F-containing resin adhesives or resin composites have been investigated.[12,13,16-18] *In vitro* caries models have indicated that materials containing relatively small quantities of F should be capable of releasing, absorbing, and re-releasing over a long period in order to be effective in caries inhibition.[19] Based on the result of this study, the resin-based desensitizers did not seem to

release sufficient levels of F to bring about a clinically remarkable impact.

Freshly exposed dentin surfaces after the grinding of cementum were used in this study and were made to resemble cavity walls prepared for restoration. Few studies have quantified the depth and the intensity of F penetrated into dentin in terms of the inward migration of F from resin-based materials. Using energy dispersive spectroscopy (EDS) and wavelength dispersive spectroscopy WDS, Ferracane and others[20] determined that F moved from the filler particles through the adhesive matrix and then down a concentration gradient into the hybrid layer and dentin. Additionally, the migration of F was observed only when marginal leakage was produced

and when water was available to dissolve F ions. More recently, Han and others[8] used EPMA to show that the F density in the interfacial area is increased by the diffusion of pre-reacted glass ionomer fillers. However, this was the case only with a new form of adhesive resin, but not with conventional F-containing adhesive resin. When F ions can be supplemented from the bottom of the hybrid layer, the acid-resistant zone may build up to prevent secondary caries formation and lesion progression.[21] Previous studies[3,9,13,16] have examined the association between the extent of secondary lesions produced in root dentin and the availability of F in the adhesive resin. In the case of a self-etch adhesive system, it was suggested that the spaces created by acidic monomers underneath the hybrid layer may be filled by dissolved calcium and phosphate ions and that F ions may migrate from the hybrid layer.[11] This F concentration at the base of hybrid layer may then continue to diffuse into the deeper dentin over time. We detected significant changes of F in the desensitizer layer of the SP group and in the hybrid layer of CL over time (Tables 2 and 3). The time-related observations were not performed with identical specimens, since specimens needed to be sectioned and subjected to the elemental analysis. Instead, each quarter segment produced from a single root was compared in order to decrease individual substrate variation. Newly dispensed materials were also used for every application, and the manufacturer's instructions were carefully followed in order to produce a uniform mixture of coating on each specimen. No elevated levels of F were detected in the subhybrid layers, and the diffused F ions, if any, may not have been sufficient in number to be identified by EPMA. The root surface was etched with 37% phosphoric acid prior to application of VX in order to facilitate F incorporation.[22] As dentin permeability increased, the effective surface area for F ion absorption and ionic exchange was expected to be enhanced. The phosphoric acid–etched dentin area was wider (16.63 ± 0.78 μm) than the hybrid layer in two of the groups (SP, 4.24 ± 1.12 μm; CL, 4.41 ± 1.07 μm). Si was not detected in this partially Ca-depleted area, while F was present at elevated levels throughout the area. The amount of F in the etched dentin area was higher than in the hybrid layer for the SP and CL groups ($p < 0.001$). This F-infiltrated interface may serve as a front against acidic challenge.

Occlusion of the exposed dentinal tubules *via* wear-resistant surface coating is a key mechanism for the prevention of dentin hypersensitivity.[3]

When resin-based desensitizing agents are micromechanically attached to dentin, they build hermetic barriers against acid penetration. Even non–F-containing resin adhesives were shown to successfully protect root dentin surfaces from caries development.[9,10,23] Furthermore, the thinly applied desensitizer layer would likely be minimally influenced by its own polymerization shrinkage, thereby retaining bonding integrity better than composite-filled cavities. Another mechanical impact related to *in vivo* simulation may be mechanical abrasion mimicking daily brushing practices, which might result in wear-off of the resin coatings. However, when root surfaces are exposed as a result of gingival recession, some areas are not easily accessible by a toothbrush. Elderly patients who have difficulties maintaining adequate oral hygiene are particularly prone to rapid root caries initiation.[24] Thus, the susceptible area with plaque retention may benefit from root surface sealants such as resin-based desensitizers as a form of intensive preventive care.

CONCLUSIONS

Within the limitations of this *in vitro* study, F-containing resin-based desensitizers were shown to be effective in sealing dentinal root surfaces for at least 60 days. The release of F from the desensitizers dropped over time, and the inward diffusion of F into the deep dentin layer was below detectable levels. The physical barrier provided by the polymerized resin matrix remained during a 60-day period.

Conflict of Interest Declaration

The authors of this manuscript certify that they have no proprietary, financial or other personal interest of any nature or kind in any product, service and/or company that is presented in this article.

REFERENCES

1. Al-Sabbagh M, Brown A, & Thomas MV (2009) In-office treatment of dentinal hypersensitivity *Dental Clinics of North America* **53(1)** 47-60, viii.

2. Wefel JS (1994) Root caries histopathology and chemistry *American Journal of Dentistry* **7(5)** 261-265.

3. Gernhardt CR, Aschenbach K, Bekes K, & Schaller HG (2005) The effect of different desensitizing agents on initial demineralization of human root dentin *Quintessence International* **36(9)** 679-685.

4. Beltran-Aguilar ED, Goldstein JW, & Lockwood SA (2000) Fluoride varnishes. A review of their clinical use, cariostatic mechanism, efficacy and safety *Journal of the American Dental Association* **131(5)** 589-596.

5. Walter R, Duarte WR, Pereira PN, Swift EJ Jr, Heymann HO, & Arnold RR (2008) Effect of resin adhesive systems on root caries formation in vitro *Quintessence International* **39(1)** 33-37.

6. Daneshmehr L, Matin K, Nikaido T, & Tagami J (2008) Effects of root dentin surface coating with all-in-one adhesive materials on biofilm adherence *Journal of Dentistry* **36(1)** 33-41.

7. Orchardson R, & Gillam DG (2006) Managing dentin hypersensitivity *Journal of the American Dental Association* **137(7)** 990-998; quiz 1028-1029.

8. Han L, Okamoto A, Fukushima M, & Okiji T (2006) Evaluation of a new fluoride-releasing one-step adhesive *Dental Materials Journal* **25(3)** 509-515.

9. Kaneshiro AV, Imazato S, Ebisu S, Tanaka S, Tanaka Y, & Sano H (2008) Effects of a self-etching resin coating system to prevent demineralization of root surfaces *Dental Materials* **24(10)** 1420-1427.

10. Tajima K, Nikaido T, Inoue G, Ikeda M, & Tagami J (2009) Effects of coating root dentin surfaces with adhesive materials *Dental Materials Journal* **28(5)** 578-586.

11. Shinohara MS, De Goes MF, Schneider LF, Ferracane JL, Pereira PN, Di Hipolito V, & Nikaido T (2009) Fluoride-containing adhesive: Durability on dentin bonding *Dental Materials* **25(11)** 1383-1391.

12. Chan WD, Yang L, Wan W, & Rizkalla AS (2006) Fluoride release from dental cements and composites: A mechanistic study *Dental Materials* **22(4)** 366-373.

13. Hara AT, Queiroz CS, Freitas PM, Giannini M, Serra MC, & Cury JA (2005) Fluoride release and secondary caries inhibition by adhesive systems on root dentin *European Journal of Oral Science* **113(3)** 245-250.

14. Caves GR, Millett DT, Creanor SL, Foye RH, & Gilmour WH (2003) Fluoride release from orthodontic band cements—A comparison of two in vitro models *Journal of Dentistry* **31(1)** 19-24.

15. Silva KG, Pedrini D, Delbem AC, & Cannon M (2007) Effect of pH variations in a cycling model on the properties of restorative materials *Operative Dentistry* **32(4)** 328-335.

16. Peris AR, Mitsui FH, Lobo MM, Bedran-russo AK, & Marchi GM (2007) Adhesive systems and secondary caries formation: Assessment of dentin bond strength, caries lesions depth and fluoride release *Dental Materials* **23(3)** 308-316.

17. Rodrigues E, Delbem AC, Pedrini D, Cavassan L, & de Oliveira MS (2010) Enamel remineralization by fluoride-releasing materials *Brazilian Dental Journal* **21(5)** 446-451.

18. Xu HH, Moreau JL, Sun L, & Chow LC (2008) Strength and fluoride release characteristics of a calcium fluoride based dental nanocomposite *Biomaterials* **29(32)** 4261-4267.

19. McCabe JF, Carrick TE, & Sidhu SK (2002) Determining low levels of fluoride released from resin based dental materials *European Journal of Oral Science* **110(5)** 380-384.

20. Ferracane JL, Mitchem JC, & Adey JD (1998) Fluoride penetration into the hybrid layer from a dentin adhesive *American Journal of Dentistry* **11(1)** 23-28.

21. de Carvalho FG, Puppin-Rontani RM, Soares LE, Santo AM, Martin AA, & Nociti-Junior FH (2009) Mineral distribution and CLSM analysis of secondary caries inhibition by fluoride/MDPB-containing adhesive system after cariogenic challenges *Journal of Dentistry* **37(4)** 307-314.

22. AS AL-H, Armstrong SR, Xie XJ, & Wefel JS (2003) Effect of smear layer on root demineralization adjacent to resin-modified glass ionomer *Journal of Dental Research* **82(2)** 146-150.

23. Gernhardt CR, Koravu T, Gerlach R, & Schaller HG (2004) The influence of dentin adhesives on the demineralization of irradiated and non-irradiated human root dentin *Operative Dentistry* **29(4)** 454-461.

24. Johnson G, & Almqvist H (2003) Non-invasive management of superficial root caries lesions in disabled and infirm patients *Gerodontology* **20(1)** 9-14.