In Vitro Study of Dentinal Tubule Occlusion with Sol-gel DP-bioglass for Treatment of Dentin Hypersensitivity

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INTRODUCTION

Dentin hypersensitivity has long been a tiresome problem afflicting many patients, particularly and prevalently among those in the age groups of 20-29.9 years (34.9%) and 30-39.9 years (33.3%)1. The major symptom of dentin hypersensitivity is characterized by brief, sharp pain that occurs in response to thermal, tactile, osmotic, chemical, or evaporative stimuli and which cannot be attributed to any form of pathology or dental defect2. Dentin hypersensitivity is closely related to exposed dentinal tubules3. Previous studies have demonstrated that sensitive teeth have an increased number of patent dentinal tubules (35.6% compared to 9.3%)4. In addition, the diameter of dentinal tubules was wider (0.83 μm) in sensitive dentin than in non-sensitive dentin (0.43 μm)5. Many factors may contribute to the exposure of dentinal tubules, such as attrition from occlusal wear, abrasion from toothbrushing, dietary erosion, parafunctional habits, gingival recession, aging, chronic periodontal disease, tooth abnormally positioned in the arch, periodontal surgery, root preparation, and abractive lesions6.

The most widely accepted theory to explain the mechanism of dentin hypersensitivity is the hydrodynamic theory, which states that stimulus application causes a pressure change across the dentin. As a result of pressure change, rapid shifts of fluids—in either direction—take place within the dentinal tubules, followed by excitation of sensory nerves in the pulp-dentin border7. Therefore, either blocking the exposed dentinal tubules with chemical agents or reducing the excitability of the relevant sensory nerves with physical agents would effectively treat dentin hypersensitivity. The Poiseuille-Hagen equation demonstrates that the movement of dentinal fluid in a tubule is proportional to the fourth power of tubular radius and the pressure difference between two ends of a tubule8. Consequently, permanent blockage of exposed dentinal tubules and subsequent reduction of fluid flow should be an efficient strategy to treat dentin hypersensitivity.

Chemical agents that have been suggested included strontium chloride, sodium fluoride, formalin solution9, ferric oxalate10, calcium hydroxide, strannous hydroxide, calcium oxalate, ferric phosphate11, potassium nitrate12, visible light-cured materials13, and fluoride varnishes14, while physical agents included Nd:YAG laser15 and GaAlAs laser16. Although these materials yielded promising results, their therapeutic effects were relatively short-lived or decreased with time because these agents could be gradually removed by daily toothbrushing or acidic beverage drinking. Therefore, the treatment material must penetrate the dentinal tubules and occlude the orifices of dentinal tubules to produce a long-lasting seal.

In a previous study, we have used bioglass and Nd:YAP laser treatment to achieve 10 μm of sealing depth for dentinal tubules17. In another study, we prepared DP-bioglass through melting of related oxide precursors at 1410°C and mixed the DP-bioglass with 30% phosphoric acid to produce homogeneous occlusion on exposed dentinal tubules and 60 μm of sealing depth18. However, the DP-bioglass paste had to be placed on the dentinal surface for three days in

Keywords: Sol-gel, Bioglass, Dentinal tubules

DP-bioglass paste has been demonstrated to produce 60 μm of sealing depth on exposed dentinal tubules. However, the occlusive effect depended on a continuous placement of DP-bioglass paste on dentinal surface for three days. In a bid to fabricate highly reactive DP-bioglass particles, a sol-gel method was used together with HNO3. DP-bioglass paste has been demonstrated to produce 60 μm of sealing depth on exposed dentinal tubules. However, the occlusive effect depended on a continuous placement of DP-bioglass paste on dentinal surface for three days. In a bid to fabricate highly reactive DP-bioglass particles, a sol-gel method was used together with HNO3. DP-bioglass paste has been demonstrated to produce 60 μm of sealing depth on exposed dentinal tubules. However, the occlusive effect depended on a continuous placement of DP-bioglass paste on dentinal surface for three days. In a bid to fabricate highly reactive DP-bioglass particles, a sol-gel method was used together with HNO3. DP-bioglass paste has been demonstrated to produce 60 μm of sealing depth on exposed dentinal tubules. However, the occlusive effect depended on a continuous placement of DP-bioglass paste on dentinal surface for three days. In a bid to fabricate highly reactive DP-bioglass particles, a sol-gel method was used together with HNO3. DP-bioglass paste has been demonstrated to produce 60 μm of sealing depth on exposed dentinal tubules. However, the occlusive effect depended on a continuous placement of DP-bioglass paste on dentinal surface for three days. In a bid to fabricate highly reactive DP-bioglass particles, a sol-gel method was used together with HNO3.
37 °C, 100% humidity environment, and could not be washed off to achieve an occlusive effect. Therefore, it was neither suitable nor feasible for clinical application.

Sol-gel-derived bioglass is known to exhibit a large surface area and porosity compared with conventional melt glasses. As such, we set out to fabricate smaller bioglass particles via sol-gel processing and utilized HNO₃, NaOH, and H₃PO₄ as catalysts to shorten the reaction time in this in vitro study. Then, we compared the occlusive effect of sol-gel bioglass on patent dentinal tubules with two commercial products: One Coat Bond (Coltène/Whaledent Inc., NJ, USA) and Seal & Protect (Dentsply Caulk, Milford, DE, USA). In addition, the microstructure and phase transformation of sol-gel bioglass were examined by means of scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDX) and X-ray diffractometry (XRD) respectively.

MATERIALS AND METHODS

Synthesis of sol-gel DP-bioglass paste and thermogravimetric analysis (TGA)
The sol-gel-prepared DP-bioglass was based on a Na₂O·CaO·SiO₂·P₂O₅ system (Na₂O 8.1%, CaO 40%, SiO₂ 39.6%, P₂O₅ 12% in weight ratio), and three types of catalyst (HNO₃, NaOH, and H₃PO₄) were used (Groups A-C). Group D represented the DP-bioglass synthesized with conventional glass-melting method and served as the control group. Raw materials and the weight amounts used are listed in Table 1. Then, to each group was added 20 ml of 95% ethanol. However, Group A received additional 1 ml of nitric acid as catalyst, while the catalysts of Group B and C were included as reactants. The mixture was fiercely stirred at room temperature for 2 hours and subsequently placed in an oven at 60 °C for seven to 10 days to form a bioglass gel. The thermal behavior of bioglass gel was first recorded by a TA/SDT2960 (Thermal Analysis Instruments Inc., New Castle, DE) for thermogravimetric analysis to determine the optimal calcination temperature. Scanning temperature was from room temperature up to 1500 °C with a heating rate of 20 °C/min and N₂ flow rate of 90 ml/min. Total weight of the specimen for each thermal analysis was 20 mg, using Al₂O₃ as the reference powder.

All bioglass gel specimens were pulverized by a Spex 8000 alumina ball mill and then placed in a platinum crucible for heating in a SiC furnace up to 220 °C for 20 hours. Dried bioglass was pulverized again and heated in the furnace up to 800 °C for three hours. Subsequently, the powder was milled again to obtain homogeneous bioglass particles. Finally, the DP-bioglass powder was mixed with 30% phosphoric acid aqueous solution in a powder/liquid ratio of 0.05 g/0.1 ml to form a DP-bioglass paste.

Specimen preparation
One hundred and twenty extracted human third molars from 16- to 40-year-old individuals with informed consent at the National Taiwan University Hospital were used for this study. Crowns with caries, restoration, or fracture were discarded. Any remaining soft tissue was thoroughly removed from the tooth surface with a dental scaler (Sonicflex 2000, KaVo Co., Biberbach, Germany). All teeth were then stored in 4 °C distilled water containing 0.2% thymol to inhibit microbial growth until use.

While hydrated, crown dentin discs of 2-mm thickness were cut perpendicular to the long axis of the tooth by means of a low-speed diamond wafering blade (Isomet, Buehler Ltd., Lake Bluff, IL). Each cut specimen was immersed in 17% EDTA followed by two minutes of ultrasonic vibration to remove the smear layer, then rinsed with copious distilled water and dried with clean air. These dentin discs were divided into six groups (A to F) with 20 in each group. Groups A-D were designated for DP-bioglass paste treatment, while Groups E-F received One Coat Bond and Seal & Protect respectively. The compositions of One Coat Bond and Seal & Protect are listed in Table 2.

DP-bioglass paste was applied to the specimens for 10 minutes and rinsed with an air-water spray for 20 seconds. As for One Coat Bond and Seal & Protect, they were handled according to manufacturers’ instructions. Briefly, One Coat Bond was applied directly from the syringe onto a disposable brush and massaged into the specimens for 20 seconds followed by light air-drying. A conventional light-curing unit (Demetron Optilux 401, Danbury, CT, USA) was then used to cure the specimens for 30 seconds. Seal & Protect was applied and agitated over the surface of the specimens for 30 seconds followed by light air-drying for five seconds. Two increments of Seal & Protect were applied on each specimen, and each increment was cured with the same light curing equipment for a period of 20 seconds.

All specimens were subsequently placed in 37 °C, 100% humidity environment for three days and then examined by a SEM (Model S-800, Hitachi, Tokyo, Japan). Half of the specimens of each group were designated for occlusive percentage calculation of the tested material on the orifices of dentinal tubules. The remaining half of each group were designated for the sealing depth of dentinal tubules to be calculated from the dentinal surface to the bottom of tested material. A groove was prepared using tapered fissure bur on each specimen to facilitate the sectioning of each specimen with a chisel. Initially, the surface area of each specimen was equally divided
into three portions and the specimens were observed at ~50 magnification. Subsequently, one site from each portion was randomly selected. Each selected site was then examined at ~1000 magnification to measure the occlusive percentage or sealing depth. Occlusive percentage was defined as the number of the orifices of dentinal tubules occluded by tested material divided by the total number of the orifices of dentinal tubules at ~1000 magnification. For sealing depth measurement, the maximum value of each selected site at ~1000 magnification was chosen. Three values of occlusive percentage or sealing depth were obtained from each specimen and the mean value calculated. Ten mean values of 10 specimens in each group were calculated again to obtain the final mean value and standard deviation of occlusive percentage or sealing depth. Data obtained were subjected to one-way ANOVA and Tukey’s post hoc test with the level of significance set at 5%.

**X-ray diffraction (XRD) analysis**

The crystalline phases of DP-bioglass paste before and after mixing with phosphoric acid were determined by an X-ray powder diffractometer (Rigaku Denki Co. Ltd., Tokyo, Japan) with CuKα radiation and Ni filter. Scanning range was 10 degrees to 60 degrees, with a scanning speed of 4 degrees/min. To determine the contents of different phases, relative intensities of the characteristic peaks of each phase were used.

**RESULTS**

**Scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDX) examination**

The occlusive effect, microstructure, sealing depth, and Ca/P ratio of DP-bioglass paste were observed using SEM-EDX (Model S-800, Hitachi, Tokyo, Japan). Specimens were immersed in 2.5% cold glutaraldehyde in 0.1 mol/L cacodylate buffer at pH 7.4 for eight hours. All specimens were then serially dehydrated in graded ethanol solutions (50%, 60%, 70%, 80%, 90%, 95%, and 100% ethanol) at 45-minute intervals, critical point-dried by CO₂, mounted on aluminum stubs, sputter-coated with ~20 nm of carbon, and finally examined by a SEM at an accelerating voltage of 15 kV.

**Table 1** Materials and weight amounts used to synthesize sol-gel DP-bioglass. Groups A-C were catalyzed by HNO₃, NaOH, and H₃PO₄ respectively.

<table>
<thead>
<tr>
<th>Group</th>
<th>Raw Materials and Weight Amounts</th>
</tr>
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<tbody>
<tr>
<td>A</td>
<td>0.23g NaNO₂, 1.712g Ca(NO₃)₂·4H₂O, 1.354g Si(C₂H₅O)₄, 0.3079g (C₂H₅O)₃PO₄</td>
</tr>
<tr>
<td>B</td>
<td>0.108g NaOH, 1.712g Ca(NO₃)₂·4H₂O, 1.354g Si(C₂H₅O)₄, 0.3079g (C₂H₅O)₃PO₄</td>
</tr>
<tr>
<td>C</td>
<td>0.108g NaOH, 1.712g Ca(NO₃)₂·4H₂O, 1.354g Si(C₂H₅O)₄, 1.949g H₃PO₄</td>
</tr>
<tr>
<td>D</td>
<td>0.084g Na₂O, 0.4g CaO, 0.396g SiO₂, 0.12g P₂O₅</td>
</tr>
</tbody>
</table>

**Table 2** Compositions of One Coat Bond and Seal & Protect.

<table>
<thead>
<tr>
<th>Product</th>
<th>Manufacturer</th>
<th>Ingredients</th>
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<tbody>
<tr>
<td>One Coat Bond</td>
<td>Coltene/Whaledent Inc., NJ, USA</td>
<td>Hydroxyethyl methacrylate (HEMA), hydroxypropyl methacrylate, glycerol dimethacrylate, polyalkenoate methacrylized, urethane dimethacrylate, amorphous silica</td>
</tr>
<tr>
<td>Seal &amp; Protect</td>
<td>Dentsply Caulk, Milford, DE, USA</td>
<td>Di- and trimethacrylate resins (urethane dimethacrylate resin, polymerizable trimethacrylate resins), PENTA (dipentaerythritol pentaacrylate phosphate), nanofillers, amorphous silicon dioxide, photoinitiators, stabilizers, cetylamine hydrofluoride, triclosan, acetone</td>
</tr>
</tbody>
</table>
bioglass prepared with the catalysts of HNO$_3$, NaOH, and H$_3$PO$_4$ respectively. Groups catalyzed by HNO$_3$ and H$_3$PO$_4$ demonstrated drastic weight loss (about 70%) at 200°C, while DP-bioglass catalyzed by NaOH revealed drastic weight loss (about 78%) at 600°C. Beyond 800°C, all curves were smooth and no further weight loss was registered thereafter.

Scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDX) examination

DP-bioglass catalyzed by HNO$_3$ demonstrated a porous and irregular structure of 10-20 µm in size (Fig. 2(a)). Pore size was 200 nm-3 µm (Fig. 2(b)). Conglomeration (20-30 µm in size) of small particles (200 nm-1 µm) was observed in DP-bioglass catalyzed by NaOH (Figs. 2(c) and (d)). DP-bioglass catalyzed by H$_3$PO$_4$ exhibited a mass (10-20 µm in size) attached with many spherical particles (1-2 µm) (Figs. 2(e) and (f)). Figures 2(g) and (h) show the DP-bioglass fabricated from conventional melted glass, which exhibited a greater mass (30-60 µm in size) without pore formation.

Table 3 shows the element percentages of sol-gel DP-bioglass. Compared with the element percentage of DP-bioglass catalyzed by NaOH or H$_3$PO$_4$, the group catalyzed by HNO$_3$ revealed similar composition to conventional DP-bioglass (Na$_2$O 8.4%, CaO 40%, SiO$_2$ 39.6%, P$_2$O$_5$ 12% in weight ratio), indicating that the reaction catalyzed by HNO$_3$ was more complete without much remaining excess reactants.

X-ray diffraction (XRD) analysis

Figures 5(a)-(c) show the XRD diffraction patterns of sol-gel DP-bioglass catalyzed by HNO$_3$, NaOH, and H$_3$PO$_4$ respectively. Figure 5(d) was the XRD diffraction pattern of conventional melt-derived DP-bioglass. Although there were three characteristic peaks in the DP-bioglass catalyzed by HNO$_3$ (Fig. 5(a)), no particular compound could be identified when compared with the standard JCPDS card. Therefore, it was considered an amorphous pattern but its crystallization was better than that of conventional melt-derived DP-bioglass (Fig. 5(d)). Prominent crystalline peaks of DP-bioglass catalyzed by NaOH and H$_3$PO$_4$ could be traced at the positions of 2θ=23.1°, 24.2°, 30.6°, and the...
Table 3 Weight and atom percentages of sol-gel DP-bioglass with different catalysts. Groups A C were catalyzed by HNO$_3$, NaOH, and H$_3$PO$_4$ respectively.

<table>
<thead>
<tr>
<th></th>
<th>Group A</th>
<th></th>
<th>Group B</th>
<th></th>
<th>Group C</th>
<th></th>
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<tbody>
<tr>
<td></td>
<td>Weight%</td>
<td>Atom%</td>
<td>Weight%</td>
<td>Atom%</td>
<td>Weight%</td>
<td>Atom%</td>
</tr>
<tr>
<td>O</td>
<td>40.0</td>
<td>56.62</td>
<td>46.08</td>
<td>63.59</td>
<td>34.6</td>
<td>53.79</td>
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<td>3.12</td>
<td>3.00</td>
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<td>0.44</td>
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<tr>
<td>Si</td>
<td>22.23</td>
<td>17.92</td>
<td>22.66</td>
<td>17.81</td>
<td>18.36</td>
<td>16.25</td>
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<tr>
<td>P</td>
<td>4.43</td>
<td>3.24</td>
<td>0.55</td>
<td>0.39</td>
<td>3.77</td>
<td>3.02</td>
</tr>
<tr>
<td>Ca</td>
<td>25.26</td>
<td>14.27</td>
<td>27.59</td>
<td>15.20</td>
<td>42.7</td>
<td>26.50</td>
</tr>
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</table>

Fig. 2 (a)-(b) DP-bioglass catalyzed by HNO$_3$ demonstrated irregular structure (10-20 μm in size) with 200 nm-3 μm of pore size. (c)-(d) Conglomeration (20-30 μm in size) of small particles (200 nm-1 μm) was observed in DP-bioglass catalyzed by NaOH. (e)-(f) A mass (10-20 μm in size) attached with many spherical particles (1-2 μm) was found in DP-bioglass catalyzed by H$_3$PO$_4$. (g)-(h) Conventional DP-bioglass exhibited a greater mass (30-60 μm in size) without pore formation.
Fig. 3 Deep penetration (arrow) of DP-bioglass paste into dentinal tubules could be observed in DP-bioglass catalyzed by HNO$_3$, NaOH, H$_3$PO$_4$ ((a)-(b), (c)-(d), (e)-(f)). However, comparatively shallower penetration of conventional DP-bioglass paste, One Coat Bond, Seal & Protect, was found in the dentinal tubules ((g)-(h), (i), (j)).
major crystalline compound was identified as calcium phosphate monohydrate (Ca(H$_2$PO$_4$)$_2$·H$_2$O) (Figs. 6(a)-(c)). The XRD diffraction pattern of conventional DP-bioglass paste showed that the major crystalline compound was dicalcium phosphate dihydrate (CaHPO$_4$·2H$_2$O) with characteristic peaks at the positions of 2θ = 11.6°, 21.0°, 29.3° (Fig. 6(d)).

DISCUSSION

Bioglass is a highly biocompatible material which can induce osteogenesis in simulated body fluids\textsuperscript{20}. It has been suggested as an adjunct material to the treatment of tooth cracks or fractures because of its strong chemical bonding with hydroxyapatite\textsuperscript{21,22}. In a previous study, we demonstrated that an occlusive depth of 10 μm in dentinal tubules could be achieved

Fig. 4 (a) Percentages of tubular occlusion in Groups A-F were 65.4%, 53.2%, 54.6%, 2.1%, 51.3%, 41.2% respectively (DP: conventional DP-bioglass; OCB: One Coat Bond; S&P: Seal & Protect) (n=10). (b) Average depths of tubular occlusion in Groups A-F were 62.7 μm, 58.9 μm, 55.8 μm, 1.5 μm, 4.0 μm, 32.5 μm respectively (n=10).

Fig. 5 DP-bioglass catalyzed by HNO$_3$ (a) and conventional melt-derived DP-bioglass (d) exhibited an amorphous pattern. Major crystalline compounds of DP-bioglass catalyzed by NaOH (b) or H$_3$PO$_4$ (c) were CaSiO$_3$ and Ca$_2$SiO$_4$·3H$_2$O.

Fig. 6 (a)-(c) After mixing with phosphoric acid to form DP-bioglass paste, the major crystalline compound was identified as Ca(H$_2$PO$_4$)$_2$·H$_2$O. (d) The major crystalline compound of conventional DP-bioglass paste was Ca(HPO$_4$)$_2$·2H$_2$O.
through the melting of bioglass with Nd:YAP laser irradiation\(^1\). Then, we further extended the occlusive depth to 60 \(\mu\)m by the use of DP-bioglass paste\(^6\). However, for chemical reaction to persist and hence achieve a deep sealing depth, there must be sufficient ion supply from DP-bioglass by virtue of a continuous release of ions. To this end, DP-bioglass paste had to be placed on the dentinal surface for three days - and could not be washed off - in a 37\(\%\), 100\% humidity environment. This prolonged reaction was not practical in a clinical setting as DP-bioglass paste would be easily washed off during mastication or toothbrushing. With an improved method employed in this study, the reaction time was substantially shortened. DP-bioglass paste was applied to the dentin surface for only 10 minutes, rinsed out with an air-water spray for 20 seconds, and then kept in 37\(\%\), 100\% humidity environment. Indeed, this method was not only more feasible for a clinical setting, but that significant tubular occlusion as well as good sealing depth were achieved by this method (Fig. 4).

With the advent of sol-gel technology, nanoparticles with large surface area could be fabricated at relatively low temperatures. In this study, we utilized HNO\(_3\), NaOH, and H\(_3\)PO\(_4\) as catalysts to accelerate the silica sol-gel formation. The underlying effects of these catalysts were described as follows. First, the hydrolysis of tetraethoxy silane (TEOS) - the first step of silica sol-gel formation - can be dramatically promoted by an acid catalyst (e.g., HNO\(_3\) or H\(_3\)PO\(_4\)) or base catalyst (e.g., NaOH)\(^3\). Secondly, condensation reaction proceeds via a rapid formation of a charged intermediate by reacting with a proton (acid-catalyzed reaction) or a hydroxide ion (base-catalyzed reaction). Due to high condensation rate and interlinking of highly cross-linked polymers, a porous network is formed and bioglass gelation rapidly occurs in seven to 10 days. Excess reactants are left unreacted at the gel stage even though the weight is determined by the nominal composition of conventional DP-bioglass (Na\(_2\)O 8.4\%, CaO 40\%, SiO\(_2\) 39.6\%, P\(_2\)O\(_5\) 12\% in weight ratio). Finally, these excess reactants have to be removed to obtain homogeneous DP-bioglass paste after mixing with phosphoric acid, and thereby obtain a deep occlusive depth.

According to the TGA curves (Fig. 1), no further weight loss was observed above 800 \(\circ\)C. The sol-gel bioglass was heated in the furnace at 800 \(\circ\)C for three hours to ensure that no reactants were left. However, this process would cause aggregation of nanoparticles (Figs. 2(a)-(f)). Nevertheless, the average particle size of sol-gel bioglass was still smaller than that of conventional melt-derived bioglass (Figs. 2(g)-(h)). Moreover, sol-gel bioglass exhibited a porous structure with large surface area, which was probably due to the high condensation rate and vaporization of NO\(^{3+}\) and OH\(^-\) at 800 \(\circ\)C. These characteristics served to accelerate the reaction such that only 10 minutes of DP-bioglass paste application on the dentin surface was enough to attain a deep sealing depth (Figs. 3(a)-(f)). In comparison, only shallow penetration of conventional DP-bioglass was observed (Figs. 3(g)-(h)) if it were removed after merely 10 minutes of application.

In terms of catalyst effectiveness, it could be evaluated from the element percentages of prepared sol-gel DP-bioglass by SEM-EDX (Table 3). The group catalyzed by HNO\(_3\) revealed similar composition to conventional DP-bioglass, as compared with the element percentages of DP-bioglass catalyzed by NaOH or H\(_3\)PO\(_4\). In other words, the sol-gel reaction catalyzed by HNO\(_3\) yielded the least amount of remaining excess reactants.

In-office treatments for dentin hypersensitivity include dentin bonding agents and desensitizing agents\(^5\). In this study, two commercial products, One Coat Bond and Seal & Protect, were chosen as representative agents to compare with DP-bioglass in terms of percentage of tubular occlusion and average depth of tubular occlusion. Percentage of tubular occlusion is related to the effectiveness of desensitizing agents, and the average depth of tubular occlusion is related to the therapeutic longevity of desensitizing agents. Both evaluation parameters are important for assessing the efficacy of desensitizing agents. The average sealing depth of Seal & Protect in this study was 32.5 \(\mu\)m (Fig. 4(b)), which was greater than the 5-10 \(\mu\)m reported in another study\(^{25}\). Although Seal & Protect has been suggested as an effective agent in reducing dentin hypersensitivity and permeability\(^{26}\), average wear on the sealant was approximately 28 \(\mu\)m at each month\(^{27}\). In addition, results showed that DP-bioglass catalyzed by HNO\(_3\) (Group A) displayed significantly better results than Seal & Protect (Group F) in both evaluation parameters (Fig. 4).

One Coat Bond, of which the composition comprised HEMA, hydroxypropyl methacrylate, glycerol dimethacrylate, polyalkenoate methacrylated, urethane dimethacrylate, and amorphous silica, was a solvent-free and one-bottle adhesive. Its use in conjunction with resin-based composites in restoring non-carious cervical lesions of teeth has demonstrated high retention rate, reduced air sensitivity, and good marginal integrity in a one-year clinical evaluation\(^{28}\). The percentage of tubular occlusion and the average depth of tubular occlusion of One Coat Bond were not significantly different from those of sol-gel DP-bioglass (Groups A-C) (Fig. 4). However, it should be highlighted that One Coat Bond was basically composed of organic polymers. In other words, if One Coat Bond were used alone without a resin...
composite, its resistance to toothbrushing would be inferior to bioglass and its durability questionable.

The sol-gel DP-bioglass catalyzed by NaOH or H_3PO_4, composed mainly of crystalline compounds of CaSiO_3 and CaO_3SiO_2â¢3H_2O (Figs. 5(b) and (c)). On the other hand, the sol-gel DP-bioglass catalyzed by HNO_3 and conventional melt-derived DP-bioglass displayed an amorphous pattern (Figs. 5(a) and (d)). These porous particles with large surface area were dissolved rapidly by phosphoric acid, thereby releasing large amounts of calcium and phosphate ions. The other source of calcium ions was derived from the dissolution of peritubular dentin after remnant phosphoric acid entered dentinal tubules. The high ion concentration gradient hastened the diffusion of ions into dentinal tubules, whereby precipitation followed until the acidic ions were neutralized. This was probably the mechanism by which sol-gel DP-bioglass paste could result in a profoundly deep sealing depth with only 10 minutes of application. The ensuing storage in a 37°C, 100% humidity environment for three days was for complete precipitation and was similar to the oral environment.

Suge et al. used a calcium phosphate precipitation (CPP) method, of which the major component was CaHPO_4â¢2H_2O, to achieve 10 mm of sealing depth into the dentinal tubules. The component was identical to that found in conventional DP-bioglass paste (Fig. 6(d)). However, the CPP method needed a post-treatment solution (1 mol/L NaHCO_3 with 0.3 mol/L NaF) for neutralization and immediate calcium phosphate precipitation. Further, the sealing depth of CPP method was shallower than that of sol-gel DP-bioglass used in this study. Apart from the high reactivity of sol-gel DP-bioglass particles, other possible suspected reasons were that Ca(H_2PO_4)_{2-n}â¢H_2O, the major crystalline compound of sol-gel DP-bioglass paste (Figs. 6(a)-(c)), exhibited higher solubility than CaHPO_4â¢2H_2O. Moreover, the pH value of Ca(H_2PO_4)_{2-n}â¢H_2O was lower than that of CaHPO_4â¢2H_2O, and this helped to facilitate the movement of ions into a deeper site of dentinal tubules.

Although results obtained using dentin disks cannot be directly applied to the clinical efficiency of these agents in occluding dentinal tubules, it has been reported to be a useful in vitro screening method before expensive clinical trials. Further, Absi et al. have reported on a positive relationship between tubular patency and sensitivity. Therefore, the results of this study can be used as a basis to rationally evaluate DP-bioglass for clinical application. Nonetheless, future clinical studies are recommended for more conclusive evidence.

**CONCLUSION**

Highly reactive DP-bioglass particles were fabricated by sol-gel technology with various catalysts. As a result, the application time of DP-bioglass paste was drastically reduced to 10 minutes. Results obtained showed 53.2-65.4% of homogeneous covering on exposed dentinal tubules and 55.8-62.7 mm of tubular occlusion depth after storage in a 37°C, 100% humidity environment for three days.

**ACKNOWLEDGEMENTS**

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