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The Efficacy of Nanohydroxyapatite and High Concentration Fluoride Containing Toothpastes on Dentin Hypersensitivity: A Scanning Electron Microscopy and EDX Study

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This study examined the changes in dentinal tubules after treatment with a toothpaste containing nanohydroxyapatite and another containing high concentration fluoride which is known to strengthen teeth. Experimental tooth samples were prepared by exposing the dentin surfaces using a pH cycling model to simulate the oral environment. The two toothpastes were applied to the dentin surfaces. After treatment, the teeth were exposed to pH cycling for 3 or 6 days. Using scanning electron microscopy, initial occlusion of the dental tubules was observed in the high concentration fluoride group. By Day 6, the number of occluded tubules increased in the nanohydroxyapatite group. Analysis of the levels of calcium (Ca), phosphorous (P) using electromagnetic radiation spectroscopy revealed that the proportion of Ca and P atoms prominently increased in both groups for 6 days. These findings demonstrated that a toothpaste containing nanohydroxyapatite effectively occluded dentinal tubules. This suggests that nanohydroxyapatite may be effective for dentin desensitization.

Keywords : component analysis, dentinal tubule, energy-dispersive electromagnetic radiation spectroscopy (EDS), scanning electron microscope (SEM), magnetic analysis

1. Introduction

Dental caries is a chronic disease characterized by the demineralization of dental hard tissues. Carious lesions occur when bacteria metabolize carbohydrates to produce organic acids, which subsequently lead to demineralization of the tooth surface [1, 2]. During this process, calcium (Ca) and phosphate (P), which are key mineral components of teeth, are released. Together with the dissolution of organic components, such as proteins follows, ultimately resulting in the structural destruction of the tooth. Although Ca and P ions from saliva can partially counteract demineralization through remineralization, this process is limited. Various agents, including fluoride, have been utilized to promote remineralization of the dental tissues [3]. The replenishment of Ca and P ions lost during demineralization facilitates the growth of hydroxyapatite crystals, the principal inorganic component of enamel, thereby contributing to remineralization. To supply Ca and P ions to the tooth surface, agents such as fluoride, hydroxyapatite, and tricalcium phosphate have been tried. Ongoing efforts are directed toward developing strategies to ensure stable and effective ion delivery to the enamel surface for remineralization [4, 5]. Fluoride, which is the most widely used agent for dental remineralization, can be delivered through various methods, among which toothpaste remains the most common and convenient vehicle. In addition to fluoride, toothpastes typically contain auxiliary components, such as abrasives, binders, humectants, and flavoring agents. Certain formulations may also incorporate Ca compounds, sodium fluoride, or high concentration fluoride ions to promote the formation of calcium fluoride deposits. The incorporation of fluoride minimizes the reaction time required for binding with Ca, thereby enhancing the synergistic Ca-F ionic effect. Clinically, topical fluoride application to the tooth surfaces aims to promote remine-

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ralization, inhibit enamel demineralization, and consequently, increase enamel surface microhardness.

Fluoride presents limitations due to its short duration of action and the potential risk of toxicity following ingestion. To overcome these drawbacks, alternative or adjunctive agents have been researched and developed [6, 7]. Recently, remineralizing agents incorporating nano-sized tricalcium phosphate and hydroxyapatite have been developed to promote enamel remineralization. These nano-scale materials exhibit distinct thermodynamic, optical, electrical, and chemical properties compared to conventional micro-sized particles. Due to their reduced particle size and increased surface area, nanoparticles facilitate faster release and greater accessibility of Ca and P ions, thereby enhancing their remineralization potential [8, 9].

This study aimed to compare the remineralizing effects of a high fluoride toothpaste and a toothpaste containing nano-sized remineralizing agents on enamel and dentin by applying a pH-cycling model that stimulates the oral environment.

2. Materials and Methods

2.1. Sample preparation

Sound human premolars were sectioned at the root portion using a hard tissue cutting machine and examined under a stereomicroscope (SZ-CTV, Olympus, Tokyo, Japan). A total of 30 tooth crowns were sectioned sagittally using a hard-tissue cutter (Minitom, Struers, Denmark) and embedded in paraffin blocks, each measuring 5.1 cm × 5.1 cm × 3.0 cm. The exposed dentin surfaces were polished with abrasive papers ranging from 400 to 1800 grit to produce a uniform surface.

2.2. Treatment group division

To evaluate the remineralization effects, the specimens were divided into four groups as follows. 1. The negative control group received no treatment and was stored in artificial saliva (Xeromia, OSSTEM, Seoul, Republic of Korea). 2. The positive control group was demineralized by demineralizing solution (Biosesang, Seongnam, Republic of Korea) and subsequently stored in artificial saliva. 3. The high fluoride concentration experimental group was treated with Curaprox toothpaste (1450 ppm). 4. The nano-hydroxyapatite experimental group was treated with Apapro (Sangi Co. Ltd, Tokyo, Japan). All experimental specimens were stored in artificial saliva after the respective treatments.

2.3. pH-cycling model

Each specimen was treated with the respective nano-hydroxyapatite and Curaprox toothpaste for 5 minutes, rinsed, and then immersed in a demineralizing solution for 3 hours, followed by rinsing. Subsequently, the specimens were immersed in artificial saliva for 2 hours to induce remineralization. The application of the agents was repeated for 5 minutes, after which the specimens were again immersed in the demineralizing solution for 3 hours, and then rinsed. Finally, the agents were reapplied for 5 minutes, and the specimens were stored in artificial saliva. The degree of remineralization in each experimental group was evaluated after 3 and 6 days. These processes aimed to reproduce the changes in the oral cavity.

2.4. Scanning electron microscopy

To examine surface changes in the specimens of each experimental group, scanning electron microscopy (SEM, JSM-IT500, JEOL, Tokyo, Japan) was employed. Depending on the degree of remineralization, the relative roughness and smoothness of the specimen surfaces were evaluated. The dentin surfaces were observed at ×1000 magnification.

2.5. Energy-dispersive electromagnetic radiation spectroscopy

Energy-dispersive electromagnetic radiation spectroscopy was used to analyze the main enamel and dentin surface components. The surfaces were completely dried at room temperature, coated with platinum, and then fixed on a stand using adhesive tape. Surface spot sizes were detected at 400 μm, at an acceleration voltage of 0.1 eV and 15 kV. The Ca and P levels were expressed as average weight % per surface of enamel or dentin. The X-ray photon energy associated with the radiation frequency was determined using the following equation:

$$E = h\nu$$

h = Planck's constant (6.626×10^{-34} J/s or 4.135×10^{-15} eV/s)
ν = frequency

The photon energy wavelength (λ) was obtained using the following equation.

$$\lambda = 1.240\times10^{-6}/E$$

The degree of remineralization in demineralized tooth specimens was quantitatively analyzed based on their major components. Energy-dispersive electromagnetic radiation spectroscopy was used to evaluate the concentrations of Ca and P. The ionic concentrations were expressed as percentages to assess compositional changes according to the treatment period in each experimental group. This experimental method was detailed in a previous study [10].

2.6. Statistical analysis

For statistical evaluation, data were analyzed using SPSS, version 20.0 (SPSS, Chicago, IL, USA). To assess the significance of surface compositional analysis, one-way analysis of variance (ANOVA) was performed at a significance level of $\alpha = 0.05$, followed by Tukey's post hoc test.

3. Results

3.1. Morphological analysis of dental samples' surfaces

The degree of dentin remineralization was evaluated after 3 and 6 days of treatment. At Day 3, the Curaprox group exhibited dentinal tubule occlusion, whereas the nano-hydroxyapatite group showed less occlusion. However, by Day 6, the nano-hydroxyapatite–treated specimens demonstrated greater dentinal tubule occlusion and smoother surfaces than those in the Curaprox group. These findings indicate that nano-hydroxyapatite exerts a more pronounced remineralization effect over a longer period (Fig. 1 and 2).

3.2. Quantification of mineral atomic components

To evaluate the release of Ca and P, comparisons were made between the negative and positive control groups. Differences were observed between the control and experimental groups, and between the two experimental groups themselves. Compared with the negative control, both experimental groups showed increased Ca and P at Day 6. However, no significant differences were noted between days 3 and 6. In contrast, when compared with

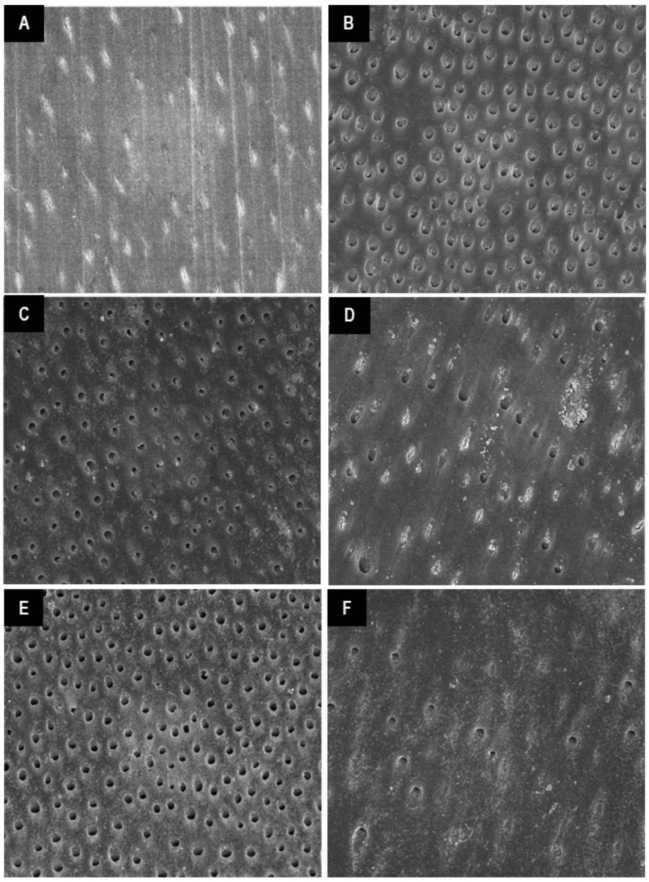

Fig. 1. SEM micrographs (1000×) of dentin specimens in the experimental groups. (A) Negative control. (B) Positive control. (C) 1450 ppm Curaprox toothpaste (high concentration fluoride) for 3 days. (D) 1450 ppm Curaprox toothpaste (high concentration fluoride) for 6 days. (E) Nanohydroxyapatite toothpaste for 3 days. (F) Nanohydroxyapatite toothpaste for 6 days.

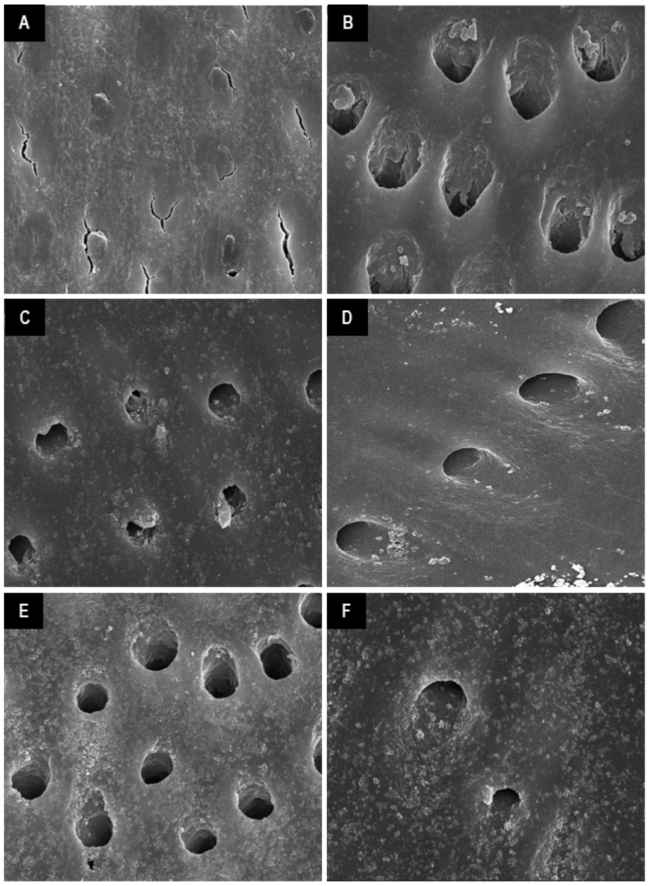

Fig. 2. SEM micrographs (5000×) of dentin specimens in the experimental groups. (A) Negative control. (B) Positive control. (C) 1450 ppm Curaprox toothpaste (high concentration fluoride) for 3 days. (D) 1450 ppm Curaprox toothpaste (high concentration fluoride) for 6 days. (E) Nanohydroxyapatite toothpaste for 3 days. (F) Nanohydroxyapatite toothpaste for 6 days.

Table 1. Compositional changes in dentin mineral elements according to treatment.

Experimental group	N	P (Mean±SD)	p	Ca (Mean±SD)	p
Negative control	15	45.55±0.60[a]	<0.001	54.45±0.60[a]	<0.001
Positive control	15	43.97±0.73[b]		52.94±0.73[b]	
Curaprox 3 days	15	48.67±0.88[a]		55.33±0.22[a]	
Curaprox 6 days	15	49.04±3.72[c]		58.96±0.58[c]	
Nanohydroxyapatite 3 days	15	47.19±0.51[a]		52.82±0.51[a]	
Nanohydroxyapatite 6 days	15	49.05±1.29[c]		60.20±1.29[c]	

[a,b,c)]Difference letter indicates statistics significant difference by One-Way ANOVA ($p < 0.05$).

the positive control, both experimental groups exhibited increased Ca and P levels from Day 3, and by Day 6, these values were significantly higher than those of both the negative and positive control groups ($p < 0.05$).

4. Discussion

Dentin hypersensitivity can be treated using various dental substances, including fluoride and desensitizing agents. The development of nanohydroxyapatites has added another solution [11]. Nanohydroxyapatite is the most biocompatible and bioactive material and has earned wide approval in medicine and dentistry recently. Nano-sized particles are similar to the hydroxyapatite crystals found in enamel. Moreover, synthetic nanohydroxyapatite has more solidity and stability than the tooth structure [4, 12]. In recent years, numerous studies have demonstrated the potential of nanohydroxyapatite in recovering dental hard tissues [13-15]. In contrast, its effectiveness in reducing dentin hypersensitivity is less researched. Additionally, comparative studies on the dentin desensitizing effect of nanohydroxyapatite and fluoride are few. In the present study, the dentin tubule occlusion effect of nanohydroxyapatite was investigated and compared with the effects of high-concentration fluoride for 3 and 6 days. A pH-cycling model was applied to reproduce the pH changes in the oral cavity throughout the day, which has a significant influence on mineral homeostasis [16]. Leal *et al.* [11] in their in vitro study, they compared the root dentin demineralization effects of nanohydroxyapatite and 5000 μg F/g concentration dentifrice, focusing on XRD, FTIR, and TEM. In another study, 3 toothpastes containing nanohydroxyapatite demonstrated significant dentinal tubule occlusion and evidence of mineral deposition on the dentin surfaces [17]. In another study, which examined the remineralization effects of nanohydroxyapatite versus fluoride toothpaste on artificial enamel lesions over 10 days, the nanohydroxyapatite-containing toothpaste demonstrated greater efficacy than the fluoride toothpaste [18]. In comparison, our SEM results revealed that dentinal tubule occlusion occurred earlier in the high- concentration fluoride treatment group than in the nanohydroxyapatite group. However, at Day 6, dentinal tubule occlusion increased in the nanohydroxyapatite group compared to the high-concentration fluoride group. These results were consistent with a previous SEM study that demonstrated greater dentinal tubule occlusion using nanohydroxyapatite toothpaste than a dentin desensitizing toothpaste containing 1450 ppm fluoride [15]. Considering these, the dentinal tubule occlusion effect appears more rapidly in high-concentration fluoride, but the degree of occlusion is higher in nanohydroxyapatite, suggesting different clinically applicable methods. Analysis of mineral atomic components by EDS showed that the proportions of mineral substances, such as Ca and P were changed before and after treatment. Our EDS analysis using an electron microscope revealed changes in Ca and P, the main mineral substances of dentin. As shown in Table 1, the ratio of Ca and P atoms decreased in the positive control. After treatment using high-concentration fluoride and nanohydroxyapatite toothpastes for 3 and 6 days, the proportion of Ca and P atoms increased in a time-dependent manner. Contrary to our results, a previous study tested hydroxyapatite, fluoride, and bioactive glass toothpastes, and concluded that the remineralization effectiveness of hydroxyapatite toothpaste was inferior to that of fluoride toothpaste [19].

This study compared the dentinal tubule occlusion effect of a nanohydroxyapatite toothpaste and a high-concentration fluoride toothpaste through SEM and EDS. While numerous studies on the enamel remineralization effect of nanohydroxyapatite exist, few studies have examined the use of nanohydroxyapatite for dentin hypersensitivity. Our findings are significant, demonstrating the potential of nanohydroxyapatite agents for dentin hypersensitivity.

5. Conclusion

This study, using SEM, demonstrated the effectiveness of nanohydroxyapatite and high concentration-fluoride-containing toothpastes in occluding dentinal tubules in a time-dependent manner. Additionally, Ca and P, the main atomic components of dentin, markedly accumulated in a time-dependent manner in both experimental groups. Taken together, this study found that nanohydroxyapatite has potential as an agent for dental hypersensitivity.

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