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Influence of pH and oxygen-inhibited layer on fluoride release properties of fluoride sealant

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ABSTRACT

Objectives: This study tested the hypothesis that the oxygen-inhibited layer on a light-cured methacrylate based resin and the pH of the storage medium would increase significantly the initial fluoride release and long-term release rate from fluoride dental sealant.
Methods: Forty-eight discs (16-mm diameter × 1-mm thick) were made from FluroShield (<5 wt% NaF) and Helioseal F (<30 wt% fluorosilicate glass) sealants. For each sealant, 24 discs were cured through a Mylar® strip that covered the surface and the remaining 24 discs were cured in air allowing formation of the oxygen-inhibited surface. Each specimen in the 24-disc groups was stored individually in 25-mL vials, and divided into four six-vial groups to receive 10 mL of pH4–pH7 (designation of pH 4–7) lactate buffer solutions. The buffer solutions were replaced periodically up to 121 days. The cumulative fluoride release over time was used to determine the coefficients for short-term and long-term release.
Results: Two-way ANOVA showed that the mean coefficient values for either sealant were significantly influenced by the curing condition ($p < 0.0001$) and pH ($p < 0.0001$), except for short-term release from NaF sealant. The duration of short-term release was much longer for the fluorosilicate glass sealant.
Conclusions: Both pH and the source of fluoride source incorporated in the sealant play significant roles in fluoride release.

1. Introduction

Fissure sealants were first introduced in late 1960s. The clinical procedure for this material involves the application of a fluid resin within the pits and fissures of the teeth after acid etching of the sites. Clinical studies of the caries-preventive effectiveness on permanent teeth have been reported.[1] Presently, the sealants can be chemical and light-cured resins, glass ionomer cements, or resins with fluoride.[2]

The concept of incorporating fluoride minerals in sealant to combine the benefits of extended topical fluoride application with the sealing of fissures was reported in the literature as early as 1972.[3] There are two basic modes of incorporating fluoride compounds in sealants, physically compounding fine fluoride minerals,[3,4] and bonding fluoride ions to the monomer matrix.[5,6] The ability of these materials to release fluoride[7–10] and to inhibit demineralization of the adjacent tooth structure *in vitro* are well documented.[11–13] However, clinical studies show that the retention rate of fluoride releasing sealants is about the same[14] or lower[15,16] than that of non-fluoride sealant. In addition, the development frequency of caries lesions associated with fluoride releasing sealants is comparable to that of non-fluoride sealant.[17–19]

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Specimen preparation for *in vitro* studies often consists of "sandwiching" the fluoride resins between two glass plates during the curing process. This procedure leaves resin-rich surface layers, which are removed before the fluoride release study. Clinically, sealant is cured in air, which leaves an oxygen-inhibited layer on the surface. The decision to leave or remove this oxygen-inhibited layer has been a subject of debate.[20] However, its effect on the fluoride release has not been fully investigated.

During a cariogenic challenge, the pH of the site decreases as a result of the metabolic activity of the microorganisms. It is generally believed that the cariostatic activity of fluoride depends mainly on the presence of fluoride in the liquid phase adjacent to and within the outer surface layer of a tooth at low pH values.[21] Thus, an increased fluoride release from fluoride sealant to the surrounding environment at a lower pH should be beneficial. While several studies have been reported on the effect of pH on the fluoride release from various types of glass ionomer cement,[22–26] compomer,[27,28] and filled resin,[29] the effect of pH on the release of fluoride from fluoride sealant has not been well characterized.

The purpose of this study was to investigate how the oxygen-inhibited layer affects the fluoride release behaviors from two sealants containing different fluoride sources. The hypothesis to be tested is that the oxygen-inhibited layer on the light-cured methacrylate base resin and the storage medium pH will increase significantly the initial fluoride release and long-term release rate from fluoride dental sealants.

2. Materials and methods

Arrays of disc molds (16 mm diameter and 1 mm thick) were made of an addition duplicating silicone (Henry Schein, Melville, NY). Two commercial fluoride sealants of different fluoride sources were used in the study (Table 1). Helioseal F (Ivoclar Vivadent, Amherst, NY) is based on fluorosilicate glass, and FluroShield (Dentsply-Caulk, Milford, DE) is based on NaF. The materials will be referred to by their fluoride filler type thereafter. They were injected into the disc molds up to the edge of the mold. One group of specimens was irradiated with a light-curing unit (Visilux 2, Model 5520 AA; 3M/ESPE, St. Paul, MN) for 30 s in air and is designated as the "open air group." The second group was cured through a Mylar® strip that covered the entire mold surface and is designated as the "covered group." After removal from the mold, the other side of the disc received another 30 s of light curing. Twenty-four specimens were prepared for each curing protocol. A total of 48 specimens were prepared for each material. Each specimen was weighed before immersing them in the storage medium.

Table 1 – Composition of materials used for fluoride release analysis

Material	Composition[a]	
	Resin	Fillers
Helioseal F	Dimethacrylate: 40–70%	Fluorosilicate glass: 10–30% SiO_2: 10–30%
FluroShield	UED-BisGMA: <40% Resin: <10% PENTA phosphate: <5% BisGMA: <5%	Glass filler: <30% Silica amorphous: <2% TiO_2: <3% NaF: <5%

[a] The information was taken from the Material Safety Data Sheet (MSDS) provided by the manufacturer.

Four buffer solutions based on the mixture of sodium acetate and acetic acid were prepared and adjusted to pH 4–7, which are designated as pH4–pH7. One disc was placed in a 25-mL vial, to which 10 mL of buffer solution was added, and the vial was stored at room temperature (25 ± 1 °C). Stainless steel wire loops were used to suspend the disc specimens from lying flat at the bottom of the vial. Six discs each were used for each pH buffer solution, curing condition and material combination. The buffer solutions were replaced periodically after exposure for 6 h, and 1, 4, 9, 16, 25, 36, 49, 64, 81, 100, and 121 days. The released fluoride ion concentration was analyzed using a fluoride-specific ion electrode and a digital pH/mV meter (Model 801A, Orion Research, Cambridge, MA). Total ionic strength adjustment buffer (TISAB) was used as a decomplexing agent for fluoride ion measurements.[30] Fluoride concentration reference solutions were prepared for each pH solution. The values were converted to cumulative released mass per unit surface area of the discs. The pH of replaced solutions was also measured.

The cumulative fluoride ion release data (Y in $\mu g/cm^2$) over time, *t*, were fit to the following equation[31]:

$$Y = \alpha\left[\frac{t}{t + t_{1/2}}\right] + \beta t^{1/2}$$

The equation suggests that there are two concurrent release mechanisms associated with fluoride release from filled resins. The first term of the equation ($\alpha \cdot [t/(t + t_{1/2})]$), representing short-term release, describes the leaching from glass particles or minerals within the surface layer of the sealants that becomes insignificant after a certain time period. The quantity α is an estimate of total release by the first mechanism (short-term release), and $t_{1/2}$ is the time at which 50% of α has been released, and *t* is the duration of the experiment. The second term of the equation ($\beta t^{1/2}$), represents long-term release, and is associated with the balance between erosive leaching of the dispersed filler particles and diffusion of fluoride through the organic matrix. The value of β is the coefficient for long-term Fickian release. The values of each coefficient were computed using analysis of variance (ANOVA) to investigate the effect of pH and type of fluoride source on long-term release rates.

At the conclusion of the experiment, each specimen was patted dry and weighed immediately. The percent weight change with respect to the pre-immersion weight was calculated. The pH of the replaced buffered solutions was measured with a pH electrode and a digital pH/mV meter. The changes in pH with respect to the pH of new buffer solution were calculated.

After removing the specimens from the final solution, one specimen was randomly selected from each group for examination by a scanning electron microscope (SEM; JSM