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An estimation of fluoride release from various dental restorative materials at different pH: *In vitro* study

Abstract

Background: In the mid of 1980s, the cariostatic effect of fluoride ions on enamel caries had been demonstrated in many studies. The use of fluoride releasing dental restorative materials has seen increasing from many years for the specific purpose of leaching of fluoride into the surrounding tissues to inhibit secondary dental caries as well as prevention of caries in the newly erupted tooth. In the dental caries, acidic environment causes the demineralization of tooth structure and also affect the restorative margins of dental restoration. **Aim:** various restorative materials show different behavior in different pH conditions of oral cavity. Thus, the aim of this study was to evaluate the fluoride release of the various restorative materials at different pH. **Design:** In this *in vitro* study, 30 samples of each dental restorative material were prepared and grouped into five with six samples in each group as per the pH of the solution 4.3, 4.6, 5.0, 5.5, and 6.2. All the samples were subjected to alternate cycling of the demineralizing solution (6 h) and remineralizing solution (18 h) for 15 days. **Results:** the fluoride release was measured by using fluoride ion specific electrode and digital ion analyzer. The result showed that the fluoride release rate was significantly higher in first day and reduced after third day to nearly constant level. At pH 4.3, the fluoride release was highest and lowest at pH 6.2. **Conclusion:** the Amalgomer CR showed the highest fluoride release among all the experimental dental restorative materials.

Key words

Cariostatic, fluoride, pH, restorative materials

Introduction

The cariostatic efficacy of fluoride has been a major component of the preventive pedodontic practices. Fluoride has been added to solutions and gels for topical applications. Fluorides releasing dental restorative

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materials have been developed with the same consideration and have been a subject of interest by research workers.

Nowadays several fluoride-containing dental restorative materials are available in the market including glass-ionomers, resin-modified glass-ionomer cements, polyacid-modified composites (compomers), composites and amalgams as advocated by Annette Wiegand *et al.* (2007). The composites have better aesthetic characteristics than glass ionomer cements but they do not release fluoride.

The fluoride releasing property of dental restorative materials depends on the physical and chemical environment, that is, saliva in vivo and the storage media *in vitro*. It also depends on physical and chemical

properties of dental restorative materials like solubility, porosity of the set restorative material mass, the chemical reaction during setting of the material as proposed by De Schepper *et al.*, 1991. Most of the studies comparing fluoride release among various dental restorative materials have confirmed the effect of storage media on fluoride release. In the present study the storage media with different pH 4.3, 4.6, 5.0, 5.5, and 6.2 have been used as demineralizing solutions and a neutral solution with pH 7.0 have been considered as a remineralizing solution. The different pH solutions have been used in attempt to simulate the oral environment in this study.

This study was envisaged to evaluate the fluoride release from various dental restorative materials at different pH to confirm the preventive measures of various dental restorative materials in oral environment.

Materials and Methods

Solution preparation
The demineralizing and remineralizing solution were prepared in triple filter distilled water. The demineralizing solutions contained 2.0 mmol/L Calcium and Phosphorus, in acetate buffer 75 mmol/L. The pH of the solution was maintained using pH paper and pH measuring electrode. The remineralizing solution contained Ca 1.5 mmol/L, P 0.9 mmol/L, KCl 150 mmol/L in Tris buffer 20 mmol/L, pH 7.0.

Sample preparation
The study comprised of six dental restorative materials, namely Amalgomer CR, Fuji II, Fuji IX, Beautifil II, Dyract extra, and Coltene Synergy. The thirty cylindrical samples of each dental restorative material were prepared using a Teflon mold, having a diameter of 5 mm and height of 3 mm. All the six dental restorative materials were manipulated according to the manufacturer instructions. Polymerization of the light cured dental restorative materials was performed using a light cure unit [GNATUS OPTILIGHT] at both sides of the sample, with a light intensity of 500 mW/cm². Conventional restorative material's initial hardening was done under pressure applied over samples for 10 min. Samples were prepared at room temperature (24 ± 12°C) and controlled relative humidity (50 ± 5%).

All the samples ($n = 180$) were stored in a covered plastic container at room temperature, for 4 h.

Experimental groups/subgroups: each experimental group was further divided into five subgroups, according to pH of demineralizing solutions with six samples ($n = 6$) in each subgroup. The samples were selected randomly from each experimental group in respective subgroups.

pH cycling of test samples at different pH
Samples in each subgroup were submitted to their respective pH level of demineralizing solution in polypropylene test tubes. Each test tube contained 2 mL of the demineralizing solution. The test tubes containing samples were subjected to constant shaking for 6 h in shaking table of orbital shaker, placed in incubator at 37 ± 1°C.

The samples of each subgroup were removed from the test tubes with the help of tweezers, rinsed with distilled water, dried with blotting paper (or tissue paper) and submitted to the next set of test tubes with each tube containing 2 mL of remineralizing solution. These test tubes were subjected to constant shaking for next 18 h in shaking table of orbital shaker and placed in incubator at 37 ± 1°C. The solutions were collected daily, identified and stored in polypropylene containers at 4°C to measure fluoride release. The procedure was continued for 15 days.

Estimation of fluoride release
Fluoride ion estimation was performed by ion selective electrode method using Expandable ion analyzer (Thermo Orion Model 940) and combination fluoride electrode (Thermo Orion Model 96 -09) previously calibrated with a series of standard solutions(0.02 to 0.32 or 1 to 16 ug F /mL). TISAB III was added to the solution at a ratio of 1:10, and fluoride concentration was measured while the solution was being constantly shaken. The fluoride concentration of the solutions were separately measured, the result of the demineralizing and remineralizing solutions were recorded, finalizing a 24 h period and thus one cycle of the pH cycling completed.

Results

Statistical analysis
The fluoride release (ppm) of various dental restorative materials at different pH over fifteen days was analyzed by using Pearson correlation analysis and simple linear regression analysis. A two tailed (a = 2), probability (P) value $P < 0.05$ was considered to be statistically

significant. MS EXCEL (MS Office 97-2003) and Graph Pad Prism (version 5) were used for the analysis.

Total fluoride release

The total fluoride release (ppm) of various dental restorative materials at different pH over 15 days was summarized in Table 1. All materials showed linear decrease in fluoride with increasing pH. The correlation analysis revealed a significant ($P < 0.01$) and inverse (negative correlation) relation between pH and fluoride release. However, the correlations of fluoride release among materials were found to be positive and significant ($P < 0.05$ or $P < 0.01$). The regression coefficient or the slope (b) of best fit regression line that indicates the rate of change were summarized in Table 2 and for each dental restorative material, the best fit regression line was also summarized graphically [Figure 1]. The Amalgomer CR showed the maximum fluoride release, the release was recorded 2.3, 1.3, 4.7, 2.1, and 7.0 fold more respectively than the respective release of Fuji IX, Fuji II, Beautiful II, Dyract extra and Coltene synergy.

The higher coefficient of determination (R^2) value of each material [Figure 1] indicates that the fluoride release of Amalgomer CR, Fuji IX, Fuji II, Beautiful II, Dyract extra and Coltene synergy can be estimated significantly from pH that can account total variation in fluoride release of these materials respectively by 92%, 90%, 99%, 96%, 99%, and 93%.

Discussion

Fluoride release in the oral environment from different dental restorative materials is an important parameter to establish their anticariogenic role. This study was therefore designed to compare fluoride release over a period of 15 days from six dental restorative materials under variable pH conditions.

In the present study all the samples of dental restorative materials were prepared with a dimension of 5 mm × 3 mm to allow maximum surface area exposed; as required in vitro study. The fluoride release was dependent on exposed surface area and not on sample weight. It was also confirmed by Williams.[1]

Cury JA (2001) proposed that saliva protects enamel at a pH higher than 5.5 and dentin at a pH higher than 6.5. Considering the difference between the critical pH of enamel and dentin, the demineralizing solutions were prepared in a pH range from 4.3 to 6.2. Featherstone and others[2] revealed that there is a correlation of process of alternate demineralization and remineralization phenomena with the development of caries in high cariogenic situation in vivo. The alternate cycling of demineralizing and remineralizing solution was used in this study to reproduce a dynamic situation of dental caries.

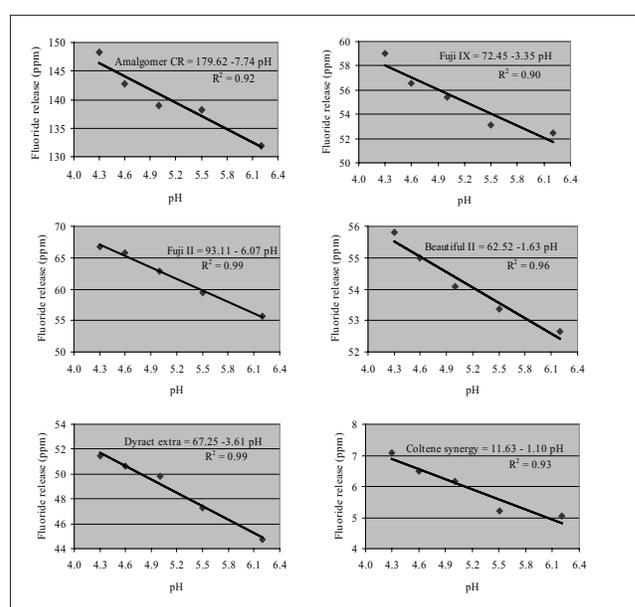In the present study the Amalgomer CR, Fuji II and Fuji IX did not show significance difference in fluoride

Table 1: Total fluoride release (ppm) of various dental restorative materials at different pH

Materials	pH 4.3	pH 4.6	pH 5.0	pH 5.5	pH 6.2
Amalgomer CR	148.33	142.69	138.91	138.24	131.87
Fuji IX	59.03	56.53	55.44	53.12	52.48
Fuji II	66.70	65.72	62.81	59.38	55.63
Beautifil II	55.79	54.98	54.07	53.36	52.64
Dyract extra	51.45	50.60	49.78	47.29	44.74
Coltene synergy	7.07	6.50	6.15	5.22	5.04

Table 2: Regression ($n = 5$) analysis summary of total fluoride release (ppm) of various dental restorative materials at different pH

Variables	Coefficients (β)	t value	P value	CI for β	
				Lower 95%	Upper 95%
Amalgomer CR	-7.74	6.02	0.009	-11.83	-3.65
Fuji IX	-3.35	5.26	0.013	-5.37	-1.32
Fuji II	-6.07	21.90	0.000	-6.95	-5.19
Beautiful II	-1.63	8.36	0.004	-2.25	-1.01
Dyract extra	-3.61	14.15	0.001	-4.42	-2.80
Coltene synergy	-1.10	6.21	0.008	-1.66	-0.54

Figure 1: Best fit regression ($n = 5$) lines of total fluoride release (ppm) of each dental restorative materials at different pH

release after third day. Beautifil ll, Dyract extra, Synergy did not show any significance difference between any days. Thus, decrease in fluoride release rate reached on constant level after third day. Since the process apparently occurred only during the first day after placement, the amount of fluoride loss may be associated with the setting and maturation reactions of the materials as evidenced by DeShepper and others.[3] Thus, the length of time for evaluating the amount of fluoride released would be important when comparing results among different restorative materials.

In this study the fluoride release values of Dyract extra seen were significantly smaller than glass ionomer and Beautifil II. This result is in agreement with the findings of Forsten[4] who compared Dyract cem with other products. Synergy composite material showed significantly less fluoride release than all experimental dental restorative materials. These findings were also confirmed earlier by Temin and Csuros.[5] It seems that fluoride compound added to the composition of composite resin lead to low fluoride release, whereas in glass ionomers, powder consist of fluoride compounds and fluoride is being released when acid base reaction is taking place during hardening.

Amalgomer CR was found to have significantly highest fluoride releasing capacity among the all experimental dental restorative materials in the present study. Like other materials in this study, it also showed the maximum release of fluoride on the first day then declining sharply thereafter and reaching near to constant level after a week. Amalgomer CR has good fluoride releasing value at various pH values of storage media.

The coarse ceramic particles reinforced in glass ionomer of Amalgomer CR may contribute to its high fluoride release. This is supported by DeSchepper and Others[3] who observed that the coarse silver alloy particles in Argion (a metal reinforced glass ionomer), which are not bound to the cement matrix, result in an increase in the microporosity of the cement, thus increasing the effective surface area available for elution of fluoride.

Fluoride forms a major constituent of many glass ionomer cements but it has no role in the cement forming process as predicted by Crisp and Wilson.[6] The other components such as calcium and aluminum that are involved in cement-matrix formation and play a basic role in hardening and strengthening the ionomer salt hydrogel as postulated by Crisp *et al.*.[7] When

powder and liquid are mixed together or during setting the ions leachable glass is decomposed by proton attack on the surface thus fluoride ions are liberating fluoride from glass particles. Swift and Dogan[8] reported that fluoride is disposed throughout the matrix region of set cement homogeneously. Fluoride in the matrix is available for elution for long period after setting as predicted by Crisp *et al.*.[7]

In this study, Fuji IX showed significantly less fluoride release than Fuji II at low pH but with pH increase it did not show significant difference. In conventional glass ionomers, fluoride release is related to the presence of fluoride compounds and their interaction with polyalkenoic acids as stated by Crisp and Wilso. [6] Thus, the difference in fluoride release level of these conventional glass ionomer materials could be the result of differences in composition or in powder – liquid ratio.

The result of the present study has shown a difference in the amount of fluoride releasing capacity of the six experimental dental restorative materials which may be attributed to several intrinsic variables like formulation and filler of the material.

Conclusion

This study led to conclusion that different pH values of storage media affect the fluoride release of experimental dental restorative materials that are emphasized as

- At low pH experimental dental restorative materials release the maximum amount of fluoride, preventing the demineralization of teeth.
- Fluoride burst occurs on first day decreasing sharply and reaching a constant value after third day.
- The Amalgomer CR showed the highest fluoride release among all the experimental dental restorative materials.

The results of the present study are of clinical importance especially to pediatric dental practices where early recognition of caries and its prevention is greatly required. The Amalgomer CR releasing higher concentration of fluoride may be advised for the restoration of tooth in highly susceptible child patients to dental caries.

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