Migration of fluoride ions from the permanent teeth into saliva in children with glass ionomer cement restorations: an in vitro study

In vitro migracija jona fluora u pljuvačku iz stalnih zuba dece sa restauracijom od glas-jonomer cementa

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Abstract

Background/Aim. Glass ionomer cements (GIC) belong to the group of polycarboxyl cements, and one of the principal characteristics of these materials is their anticariogenic potential of fluorine release into saliva and enamel-dentin substance. The aim of this study was to examine the content of released fluorine from GIC restorations (Fuji IX, GC, Japan) of young permanent teeth in the medium of artificial saliva and similar releases in the same medium by the restorations of these teeth treated with a low concentration fluoride solution.

Methods. We examined 12 premolars extracted from orthodontic reasons. The GIC restored teeth were divided into the group treated daily with low concentration fluoride solution (334 ppm) and the control, not treated group. The samples of artificial saliva were analyzed for fluorine ion content using an ion selective electrode.

Results. Our comparative analysis of the mean values using the Student’s t-test demonstrated a statistically significant difference in fluorine ion concentration in artificial saliva of fluoridated and non-fluoridated teeth with GIC restorations after 14 and 21 days, while the difference detected after 7 days was with no statistical significance.

Conclusion. The results of this in vitro study indicated that low-concentration fluoride solutions could serve to refluoridate GIC fillings and contribute to an increased fluorine content in saliva. The process of refluoridation of GIC fillings should be advised 2–3 weeks after the restoration, since the release of fluorine from GIC fillings diminishes in time.

Key words: dental cements; saliva, artificial; fluorides; child.

Apstrakt

Uvod/Cilj. Jedna od najznačajnih karakteristika glas-jonomer cementa (GJC) je antikariogeni potencijal oslobađanja fluorida u pljuvačku i glezndentinsku supstancu. Cilj ove studije bio je praćenje sadržaja oslobodjenih jona fluora iz restauracija od GJC (Fuji IX, GC, Japan) na mladim stalnim zubima u medijumu veštačke pljuvačke, kao i praćenje ovog oslobađanja u istom medijumu, iz restauracija kod pomenutih zuba tretiranih niskokoncentrovanim rastvorom fluorida.

Metode. U istraživanju je korišćeno 12 premolara ekstrahovanih iz ortodontskih razloga. Zubi restaurisani glas-jonomer cementom bili su podeljeni u grupu koja je svakodnevno tretirana rastvorom niskokoncentrovanog fluorida (334 ppm) i kontrolnu grupu koja nije tretirana fluoridima. Uzorci veštačke pljuvačke su analizirani na sadržaj jona fluora primenom jon selektivne elektrode.

Rezultati. Komparativnom analizom srednjih vrednosti Studentovim t-testom utvrđena je statistički značajna razlika između koncentracije jona fluora u veštačkoj pljuvački, fluorisanih i nefluorisanih zuba sa GJC ispunom posle 14. i 21. dana (p < 0,05), dok je analiza posle 7 dana pokazala da razlika postoji, ali bez statističke značajnosti.

Zaključak. Rezultati istraživanja ove in vitro studije pokazuju da niskokoncentrovan fluoridni rastvor može poslužiti za refluorizaciju GJC ispana i time doprineti povećanju fluoridnog sadržaja u pljuvački. Proces refluorizacije GJC ispana predlaže se posle 2–3 nedelje od restauracije, pošto se oslobađanje fluora iz GJC ispana vremenom smanjuje.

Ključne reči: zub, cement; pljuvačka, veštačka; fluoridi; deca.

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Introduction

Glass ionomer cements (GIC) belong to the group of polycarboxyls, introduced in dentistry by Smith in the late 1960s in order to improve the adhesiveness of restoration materials to hard dental tissues.

The development of GIC in the last decade has witnessed some significant changes in the composition of the glass component of powder and in polycarboxyl acids of the fluid component. Original GICs were based on SiO2-Al2O3-CaF2-PO42-AlF3-Na3AlF6 composition. According to Wilson and McLean, the Al2O3/SiO2 ratio should be 1 : 2 or more, with fluoride component content reaching even 23%. The typical chemical composition of a GIC powder is shown in Table 1. More recently, GICs supplemented with Sr, Ba, and Zn have become commercially available.

Table 1
Composition of glass ionomer cement powder according to Wilson and McLean

<table>
<thead>
<tr>
<th>Substances</th>
<th>Quantity (ppm)</th>
<th>Mass percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO2</td>
<td>41.9</td>
<td>35.2</td>
</tr>
<tr>
<td>Al2O3</td>
<td>28.6</td>
<td>20.1</td>
</tr>
<tr>
<td>AlF3</td>
<td>1.6</td>
<td>2.4</td>
</tr>
<tr>
<td>CaF2</td>
<td>15.7</td>
<td>20.1</td>
</tr>
<tr>
<td>NaF</td>
<td>9.3</td>
<td>7.6</td>
</tr>
<tr>
<td>AlPO4</td>
<td>3.8</td>
<td>12.0</td>
</tr>
</tbody>
</table>

In spite of the fact that mechanical properties of these materials limit their usefulness as the materials for final cavity closure in all fillings, its anticariogenic property made GIC one of the most attractive materials in pedodontics. These materials release fluorine ions, chemically attach to hard dental tissues, possess thermal compatibility with the enamel, biocompatibility, and low cytotoxicity, and can be used with milk teeth.

Fluorine ions released from the restoration materials contribute to the reduction of caries via the physical-chemical and biologic pathways, with anticariogenic potential of the materials largely depending on the amount of released fluorine, as well as on the duration of that release.

Studies have shown that GICs are the most effective fluorine-releasing materials, but also that in the situation of continued presence of fluorides in the mouth cavity, these materials show the ability to uptake them.

The aim of this study was to establish the presence of released fluorine ions from GIC restorations in young permanent teeth in the medium of artificial saliva, and similar release in the same medium from restorations of these teeth treated with low-concentration fluoride solutions.

Methods

The study was performed in the Department of Preventive and Pediatric Dentistry, Dentistry Clinic the Department of Pharmacy - Analytical Chemistry, the Institute of Histology and Embryology, and the Public Health Institute of the Faculty of Medicine in the town of Niš.

In this in vitro study, we used 12 young, permanent, healthy premolars extracted from orthodontic reasons, kept after extraction in physiologic solution for one month. The study was performed in three phases: teeth extraction and preparation; teeth incubation and fluoridation, and determination of fluorine ion concentration.

Phase I: Teeth extraction and preparation

After extraction, the surfaces of all the teeth were cleaned, roots were cut off with a metal cutter at the level of enamel-cement borderline, and the remaining pulp tissue was removed. The average mass of the studied tooth samples was 0.627 ± 0.105 g.

From the vestibular aspect, class V cavities were prepared, 3 × 2 × 2 mm in size, using a diamond drill, and all the teeth were restored with GIC (Fuji IX, GC, Japan) following the manufacturer’s guidelines. After GIC binding, the excess material was removed and the teeth were washed under the current of distilled water and placed in 100 mL of artificial saliva. Chemical composition of the artificial saliva solution is shown in Table 2.

Table 2
Chemical composition of an artificial saliva solution

<table>
<thead>
<tr>
<th>Substance</th>
<th>Concentration (mol/dm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaHCO3</td>
<td>1.5 × 10⁻²</td>
</tr>
<tr>
<td>KCl</td>
<td>2.0 × 10⁻²</td>
</tr>
<tr>
<td>KHCO3</td>
<td>1.5 × 10⁻²</td>
</tr>
<tr>
<td>Lactic acid</td>
<td>10.0 × 10⁻¹</td>
</tr>
</tbody>
</table>

The pH value of artificial saliva was around 6.7, as the closest approximation to physiologic values for saliva in the mouth cavity. Fluorine ion concentration in the artificial saliva was 0.071 ppm.

All glass-ionomer cement restored teeth (GICrT) were divided into two groups with six teeth each. The first group was treated daily with low concentration fluoride, while in the second group there was no fluoridation. Both groups were further divided into three subgroups with two teeth each, according to the experiment duration (7, 14, and 21 days).

Phase II: Teeth incubation and fluoridation

The prepared teeth samples were treated with the solution of artificial saliva incubated at 37 °C during the aforementioned periods of time.

The first group of GICrT (six samples) was fluoridated with the fluoride solution (concentration of 334 ppm), composed of 10 mL of low concentration fluoride solution and 5 mL of artificial saliva solution. The teeth in this group were fluoridated daily, being submerged in this solution for 1 minute, and then washed for 5 seconds with distilled water, being returned after drying into the artificial saliva. After the planned treatment periods, the samples of artificial saliva were analyzed for fluorine content.

Phase III: Determination of fluorine ion concentration

The content of fluorine ions was determined by way of automated potentiometric titration using a ion selective fluoride electrode (Ma-5705, Iskra, Slovenia). The obtained fluo-
Fluorine concentrations in artificial saliva solution were expressed in ppm.

The obtained values were compared using the Student’s t-test for independent small samples, with the test statistical significance cut-off value of $p < 0.05$. Statistical analysis was done using the SPSS software (version 15).

**Results**

The concentrations of fluorine ions released into the artificial medium for non-fluoridated and daily fluoridated GICrT after 7, 14, and 21 days are shown in Table 3.

The release of fluorine from GICrT in the medium of artificial saliva was highest in the first week in both fluoridated (0.833 ppm) and non-fluoridated teeth (0.644 ppm). Cumulative values of released fluorine ions were highest after the day 21 for GICrT treated daily with fluoride solution (1.209 ± 0.067 ppm). The median values demonstrated the highest difference (0.328 ppm) for GICrT kept in the artificial medium for 21 days. By way of comparative analysis of $\bar{X}$, a statistically significant difference was found between fluoridated and nonfluoridated GICrT after 14 and 21 days ($p < 0.05$). Statistical analysis for non-fluoridated and fluoridated GICrT after 7 days demonstrated a difference, although not a statistically significant one ($p = 0.076$) (Figure 1).

<table>
<thead>
<tr>
<th>Concentration</th>
<th>Fluoridated GICrT</th>
<th>Non-fluoridated GICrT</th>
<th>$t$-test</th>
</tr>
</thead>
<tbody>
<tr>
<td>7 days</td>
<td>0.833 ± 0.069</td>
<td>0.644 ± 0.036</td>
<td>0.188</td>
</tr>
<tr>
<td>14 days</td>
<td>1.053 ± 0.030</td>
<td>0.848 ± 0.047</td>
<td>0.204</td>
</tr>
<tr>
<td>21 days</td>
<td>1.209 ± 0.067</td>
<td>0.881 ± 0.001</td>
<td>0.328</td>
</tr>
<tr>
<td>$\bar{X}$</td>
<td>1.032 ± 0.175</td>
<td>0.791 ± 0.118</td>
<td>0.241</td>
</tr>
</tbody>
</table>

*The mean value of released fluoride ions of all the teeth in the group; **The mean value of released fluoride ions in fluoridated teeth; ***The mean value of released fluoride ions in non-fluoridated teeth; f – fluoridated; nf – non-fluoridated

![Fig. 1 – Concentration of released fluoride ions (ppm) in the medium of artificial saliva after 7, 14, and 21 days in the fluoridated and non-fluoridated GICrT](image1)

All non-fluoridated GICrT released 0.791 ± 0.118 ppm of fluorine, while fluoridated GICrT released 1.032 ± 0.175 mg/dm$^3$ of fluorine. By way of comparative analysis of $\bar{X}$ of all the studied teeth, a statistically significant difference was established between the total fluorine concentration released by all fluoridated and nonfluoridated GICrT ($p = 0.0193$).

Comparing the concentration of released fluorine ions in the medium within the groups in Figure 2, we can see that the highest ion concentrations were released in the artificial saliva medium in the first week, in both fluoridated and nonfluoridated teeth (0.883 and 0.664 ppm, respectively). During the second week the observed release trend continued, although with a lower concentration of newly released fluorine ions compared to the first week in both studied groups (0.220 and 0.204 ppm, respectively). In the third week, compared to the second week, the trend of reduction of concentration of newly released ions of fluorine continued (0.156 and 0.033 ppm, respectively). These data demonstrated that the release was still present, with the lowest concentration of newly released fluorine ions in the third week, and with a lower reduction rate in fluoridated teeth (Figure 2).

Non-fluoridated group of teeth released on the average 0.29 mg/dm$^3$ of fluorine a week in the medium of artificial saliva, while fluoridated teeth released 0.40 mg/dm$^3$ of fluoride a week during this three week study.

**Discussion**

The use of restoration materials based on glass ionomers and the use of oral low concentration fluorides are among the best solutions to treat and prevent caries in chil-
Our results in the first week demonstrated that the concentrations of fluorine ions released into the artificial saliva medium in non-fluoridated GICrT were insignificantly higher compared to non-fluoridated teeth (0.188 ppm), which could be explained by an initial release of fluorine from non-fluoridated GICrT, and the statistical significance could not be confirmed \((p = 0.076)\). If we compare the differences between the groups of teeth after 14 and 21 days, we may say that the fluorine concentration in the medium rose after 14 days to 0.204 ml/dm\(^2\) and after 21 days to 0.328 mg/dm\(^2\) (the advantage on the part of fluoridated teeth).

In non-fluoridated and fluoridated GICrT, there was a statistically significant difference in the concentrations of released fluorine after 14 and 21 days \((p = 0.035\) and 0.020, respectively), demonstrating that with diminished fluoride concentration in GIC there was an increased affinity towards fluorine from the fluoride solution. GICs were able to uptake and release fluorine due to their high reactivity. It was established that the materials with higher initial release of fluorine were characterized by a higher reuptake ability; moreover, old, refluorinated GICs could never reach the initial level of fluorine release.\(^{12}\)

In our study, we used low concentration fluoride solution of 334 ppm, intended for everyday use. The studies showed that the degree of refluoridation was higher if more concentrated solutions were used, as well as with more frequent applications and lower pH of the environment (e.g., the conditions favoring the development of caries) \(^{13,14}\). However, most researchers thought that everyday use of low concentration solutions was more effective compared to highly concentrated weekly or biweekly solutions; it was not because of their stronger action or effectiveness, but because they additionally motivated users to maintain their oral hygiene.\(^{15}\)

The results of this study are compatible to other studies demonstrating the mode of fluoride release by GICs, characterized at first by an initial rapid release, and by rapid reduction of fluoride release afterwards \(^{16–18}\). Wieand et al.\(^{12}\) reported two mechanisms of fluoride release from GICs, the first being an abrupt reaction of dissolution of the external GIC layer, and the second being a slower one, involving a continued migration of ions from the deeper GIC layers.

In our study we used pure premolars, submersed in the solution of artificial saliva, representing just an experimental model, the characteristics of which could be markedly different from the real saliva. In the mouth cavity environment, higher viscosity of the saliva or accumulation of the dental plaque can both have an impact on the ion diffusion into or from GICs \(^{17}\). Moreover, ionic composition of saliva can have an impact on the migration of fluorine ions \(^{20}\).

**Conclusion**

In this *in vitro* study both groups of teeth restored with GIC released initially the highest concentration of fluorine ions in the medium of artificial saliva during the first week, with fluoridated GICrT releasing more. Comparing the values of released fluorine ions during the second and third week in the medium of artificial saliva, a higher value was established in fluoridated GICrT compared to non-fluoridated ones, the difference reaching a statistical significance.

From the obtained results, a conclusion may be drawn that oral, low concentration fluoride solutions can serve to refluoridate GIC fillings and thus increase the content of fluorine in saliva, plaque, and hard dental tissues adjacent to fillings, and on the other enamel surfaces of the real saliva. The process of refluoridation of GIC fillings should be informed 2–3 weeks after the restoration, since fluorine release from GIC fillings diminishes in time.

**References**


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