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ANALYSIS OF ANION AND CATION RELEASE FROM GLASS IONOMER CEMENTS USING ION CHROMATOGRAPHY

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ABSTRACT: The aim of this study was to determine the amount of the anions (F^-, Cl^-, PO_4^{3-}, and SO_4^{2-}), cations (Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}), and silica released from four different glass ionomers stored in ultra-distilled water. The materials were a novel restorative glass ionomer (Glass Carbomer™ Fill), a high-viscosity glass ionomer (EQUIA Forte®), a resin-modified glass ionomer (Photac™ Fil Quick Aplicap™), and a traditional glass ionomer (Riva Self Cure). Twelve cylindrical specimens, 10 mm in width and 2 mm in depth, were prepared from each material. Each sample was placed in a sterile polypropylene tube with a cover, and 50 mL of ultra-distilled water (18 megaohm-cm) was added. Each tube was rinsed twice daily. After 14 days, the samples were removed from the solutions. The amount of anions and cations in the solutions were analysed using ion chromatography. Statistical analysis, for two group comparison, was performed using one-way ANOVA, Duncan's multiple range test, and the independent samples *t*-test ($p<0.05$). The resin-modified glass ionomer released the highest amount of F^-, Cl^-, PO_4^{3-}, Na^+, and silica ($p=0.001$). The lowest F^-, Na^+, and K^+ release was detected in the traditional glass ionomer.

Keywords: Anion; Cation; Chromatography; Glass ionomer; Silica

INTRODUCTION

Fluoride (F) can be found in the earth's crest, the groundwater, and the air, into which it may be released by industrial activities.[1-3] A high intake of F, from drinking water, food, air, and dental products, can adversely affect many organ systems and impair human health.[4-7]Although the WHO recommend an upper limit for F in drinking water of 1.5 mg/L, they also allow countries to set their own national standards or local guidelines. The country standard for India is 1 mg/L with the rider the "lesser the fluoride the better, as fluoride is injurious to health," for Senegal, West Africa, it is 0.6 mg/L, and for community water fluoridation in the USA the recommended level is 0.7 mg/L. Some restorative materials, especially glass ionomer cements (GICs), contain F.[8-11] GICs consist of a powder (fluoroaluminosilicate glass) and a liquid (polyacrylic acid). The powder part of GICs is prepared by melting AlF_3, CaF_2, SiO_2, Al_2O_3, NaF, and $AlPO_4$ at temperatures ranging from 1,200 to 1,550ºC. A homogeneous glass is formed, after the melting process. Micron-level dust particles are obtained by grinding this glass.[8] GICs have been used for different purposes, such as restorative filling, pit-and-fissure sealing, luting, lining, and as base cements depending on the particle size.[8,9] In the course of time, they have been modified, mainly by the addition of resin components, to improve their mechanical properties, without an adequate evaluation of F release.[10,11] Resin-modified glass ionomer (RMGI) is set through an acid–base reaction and polymerisation with light.[12-14] In addition, manufacturers also produced high-viscosity glass ionomers (HVGI) that can be used in the posterior region, while retaining the basic features of the conventional GICs. The physical and chemical properties of the conventional GICs have been improved by adding glass particles of different sizes, by altering the proportions of the powder and liquid parts, and by changing the amounts of Ca^+ and

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Al^{3+} ions.[15] Glass Carbomer™ Fill (GCF) is a novel restorative filling material that incorporates F and nano-hydroxyapatite particles. GCF demonstrates excellent clinical performance in posterior teeth, and is superior to many traditional and high-viscosity GICs because of its nano-sized powder particles and the fluoride–hydroxyapatite structure. The manufacturer also claims that the material becomes stronger when the total particle surface in contact with the Glass Carbomer™ liquid increases.[16,17]

GICs are known sources of low F release into the mouth, for long periods.[18] F can inhibit demineralisation as well as bacterial colonisation and bacterial acid production.[19] In addition to F, other anions and cations can be released from GICs into an aqueous solution.[10,11] The anions and cations released from these materials may also be useful for secondary caries prevention, remineralisation, and prevention of bacterial colonisation. Therefore, the importance of the released anions and cations is quite substantial.[10,18,19]

In the literature, there is little documentation on the amounts of anions and cations released from of GICs when their physical and mechanical properties are changed. The aim of this study was to compare the amount of anions and cations released from a novel restorative glass ionomer (GCF), a high-viscosity glass ionomer (HVGI), a resin-modified glass ionomer (RMGI), and a traditional glass ionomer (TGI). The null hypothesis was that the amount of anions and cations released did not differ significantly between the types of glass ionomer.

MATERIALS AND METHODS

Two ion chromatography (IC) systems, one for anions and the other for cations, were used for the analysis because of their accepted advantages. They have a multi-anion or multi-cation analytical capability and only detect ions.[20] Complex compounds cannot be estimated by this method.[20,21] This matter is important because only free F ions can enhance tooth resistance to secondary caries attacks. In addition, the silica release from each material was evaluated.

Sample preparation: Four different capsuled glass ionomer restorative materials, of A2 color, were used in this study; GCF (GCP Dental, Ridderkerk, Netherlands), HVGI (EQUIA Forte®, GC Corporation, Tokyo, Japan), RMGI (Photac™ Fil Quick Aplicap™, 3M ESPE, St. Paul, MN USA), and TGI (Riva Self Cure, SDI Limited, Bayswater, VIC, Australia). The materials and contents are shown in Table 1.

Twelve cylindrical specimens, 10 mm in width and 2 mm in depth, were prepared from each material according to the manufacturer's instruction at 23±2ºC.[11] Polymerisation was carried out with light-curing devices for the GCF and RMGI materials (Table 2). After polymerisation, the samples were polished with a low-speed hand device and polishing burs. The samples were kept at 95% humidity and 37ºC for 24 hours in an incubator.[10,11] Then, each sample was placed in a sterile polypropylene tube with a cover, and 50 mL of ultra-distilled water (18 megaohm-cm) was added. All samples were stored at 37ºC for 14 days in an incubator. Each tube was rinsed twice daily. After 14 days, the samples were removed from the solutions, and the latter were analysed for anions and cations using two IC systems.

Table 1. Tested materials (PT=polymerization type)

Material	Composition	Color	Manufacturer	Lot number	PT
Glass Carbomer Fill (novel glass ionomer)	Fluoroaluminosilicate glass, apatitie, polyacids	A2	GCP Dental, Ridderkerk, Netherlands	7310972	Light and heat
Equia Forte (high viscosity glass ionomer)	Fluoroaluminosilicate glass, polyacrylic acid powder, pigment, polyacrylic acid, distilled water, polybasic carboxylic acid	A2	GC, Corporation Tokyo, Japan	1610251	Chemical
Photac Fil Quick Aplicap (resin-modified glass ionomer)	Fluoroaluminosilicate glass, surface modified with 2-propenoic acid, 2-hydroxyethyl-methacrylate(HEMA)	A2	3M ESPE, St. Paul, USA	552651	Light
Riva Self Cure (traditional glass ionomer)	Fluoroaluminosilicate glass, acrylic acid polymers, tartaric acid	A2	SDI Limited, Bayswater, VIC, Australia	B121211 1EG	Chemical

Table 2. Light curing devices used in this study

Group	Device	Manufacturer	Light Intensity (mW/cm^2)	Curing time (sec)
Glass Carbomer Fill	GCP CarboLED thermo-cure lamp	GCP Dental, Ridderkerk, Netherlands	1,200	60
Resin-modified glass ionomer	Elipar S10	3M ESPE, St. Paul, USA	1,200	20

Anion tests: A Dionex 5000 ion chromatography system (Thermo Scientific, Dionex IonPac, USA) was used to measure the levels of the anions (F^-, Cl^-, PO_4^{3-}, SO_4^{2-}) released from the glass ionomers. Before the measurements were made, the device validation was performed with a standard mixture (High-Purity Standards, North Charleston, USA). The standard mixture (100 µL) was injected into the device and the calibration peaks were obtained. A different peak was formed for each anion.

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The amount of anions (ppm=mg/L) were determined by using the peak area or height.[20] The conditions of the Dionex 5000 ion chromatography during the analysis process are specified in Table 3.

Table 3. Device conditions of the two ion chromatography

Parameter	İon chromatography system	
	Dionex 5000	Dionex 1000
Analytical column	Dionex IonPac AS19 (4X250mm) (Thermo Scientific, Dionex IonPac, USA) (Lot: 014-27-023)	Dionex IonPac CS12A (4X250mm) (Thermo Scientific, Dionex IonPac, USA) (Lot: 012-26-020)
Guard colomn	Dionex IonPac AG19 (4X50mm) (Thermo Scientific, Dionex IonPac, USA) (Lot:013-29-082)	Dionex IonPac AG 19 (4X50mm) (Thermo Scientific, Dionex IonPac, USA) (Lot:010-21-137)
Suppressor current	112 mA	53 mA
Total signal duration	<2 µsec	<2 µsec
Cell temperature	30°C	30°C
Flow	1 mL/min	1 mL/min
Autosample injection volume	100 µL	25 µL

Cation tests: A Dionex 1000 ion chromatography system (Thermo Scientific, Dionex IonPac, USA) was used to measure the levels of the cations (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}) released from the four different glass ionomers. Firstly, device validation was performed with a standard mixture (AccuStandard, IC-MCA-06-1, 125 market Street, New Haven, USA). Secondly, the standard mixture (25 µL) was injected into the device and calibration peaks were obtained. A different peak was formed for each cation. The amount of cations (mg/L) were determined by using the peak area or height.[20] The conditions of the Dionex 1000 ion chromatography during the analysis process are shown in Table 3.

Statistical analysis: The statistical analysis, for two group comparisons, was performed using the one-way ANOVA, Duncan's multiple range test, and the independent samples *t*-test. (SPSS 17 for Windows, SPSS Inc., Chicago, IL, USA) with the significance level set at $p<0.05$.

RESULTS

The mean and standard deviation of the amount of each anion released are shown in Table 4.

Table 4. The amounts of the anions released from the glass ionomers. Values are expressed as mean±SD (ppm)

Material	Anion			
	F^-	Cl^-	SO_4^{2-}	PO_4^{3-}
Glass Carbomer Fill	7.92±1.12[b]	0.19±0,07[b]	0.92±0.8[a]	0.55±0.08[b]
High-viscosity glass ionomer	5.98±0.37[c]	0.12±0.02[c]	0.23±0.02[b]	0
Resin-modified glass ionomer	9.09±0.27[a]	0.25±0.1[a]	0.005±0.002[b]	0.64±0.06[a]
Traditional glass ionomer	2.21±0.45[d]	0.16±0.04[bc]	0.05±0.02[b]	0

[a,b,c,d] In the same anion group: the difference between the group averages of different lowercase letters is significant (p<0.05).

Each material released F^-, Cl^-, and SO_4^{2-}. The amount of F^- was significantly different between the glass ionomers ($p=0.001$). RMGI released the highest amount of F^-, followed by GCF, HVGI, and TGI. RMGI also released the highest amount of Cl^- while HVGI released the lowest amount ($p=0.001$). The highest value of SO_4^{2-} release was observed in the GCF group ($p=0.001$) and other materials were similar in their release levels. There was a statistically significant difference between RMGI and GCF in terms of PO_4^{3-} release ($p=0.007$), whereas no PO_4^{3-} release was detected for TGI and HVGI groups.

There was a statistical difference between the groups in terms of cation release ($p=0.001$; Table 5).

Table 5. The amounts of the cations released from the glass ionomers. Values are expressed as mean±SD (ppm)

Material	Cation			
	Na^+	K^+	Mg^{2+}*	Ca^{2+}
Glass Carbomer Fill	9.45±0.55[b]	0.24±0.06[c]		0.63±0.02[a]
High-viscosity glass ionomer	7.81±0.46[c]	4.46±0.32[a]		0.21±0.05[c]
Resin-modified glass ionomer	16.03±0.21[a]	0.4±0.11[b]	0.51±0.08	0.14±0.03[c]
Traditional glass ionomer	4.44±0,46[d]	0.16±0.05[c]		0.32±0.06[b]

[a,b,c,d] In the same anion group: the difference between the group averages of different lowercase letters is significant (p<0.05). * No statistical analysis

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The amount of Na^+ released was significantly different between the groups. RMGI released the highest amount of Na^+, followed by GCF, HVGI, and TGI. The highest value of K^+ release was observed in the HVGI group, TGI and GCF groups were similar. Mg^{2+} release was observed only in the RMGI group. GCF released the highest amount of Ca^{2+}. The tested materials did not release NH_4^+. The highest silica release was found in the RMGI group, the lowest release was HVGI (p=0.001; Table 6).

Table 6. The amounts of silica released from the glass ionomers. Values are expressed as mean±SD (ppm)

Material	Silica
Glass Carbomer Fill	9.95 ± 1.46^{b}
High-viscosity glass ionomer	6.3 ± 2.40^{d}
Resin-modified glass ionomer	15.6 ± 1.6^{a}
Traditional glass ionomer	8.07 ± 0.27^{c}

^{a,b,c,d} There is a statistical difference between different lowercase letters ($p<0.05$).

DISCUSSION

GICs are suitable materials for developing further. Different types of capsuled glass ionomers have been developed to ensure an ideal powder:liquid ratio. Newly introduced GCF, HVGI, and RMGI are examples of these materials.[10,11,22] All types of GICs release F,[10,11,22,23] and the antibacterial and cariostatic properties of GICs are associated with the amount of F released.[9] GICs may release not only F but also some anions and cations.[9-11] Calcium, which is a cation, has similar antibacterial and cariostatic effects to F. Therefore, the amount of anions and cations released from the materials is very important for dental health.

In relation to time, there are two processes for F release from GICs: the fast elution process and the long-term release process. Previous studies reported that the highest amount of F release from GICs occurred during the first day.[10,11,18] In GICs, there is an acid–base reaction resulting in the leaching of F ions to form a polysalt matrix.[9] In addition, unreacted glass particles may cause a rapid release after mixing,[23] which may be responsible for the fast elution process. This phenomenon is known as the "burst effect."[18] There is also a much smaller but steady release that can last for a long period of time.[11] These release processes may be valid for other anions and cations. Forss[24] reported that similar release activities in some ions, such as sodium, aluminium, and calcium, and in silicon had been observed. In the present study, F^-, Cl^-, PO_4^{3-}, SO_4^{2-}, Na^+, K^+, Mg^{2+}, Ca^{2+}, and Si release from the GICs were observed. When all the results of the present study were considered together, there were statistically significant differences between the type of GICs. Therefore, the null hypothesis was rejected.

Kuhn and Wilson[25], in relation to the release mechanism, stated the presence of three mechanisms for F release from GICs: diffusion through pores and microfractures, mass diffusion, and superficial rinse. Superficial rinse can be related to the particle size of the GICs. Glass particles are milled under dry conditions to a size less than 20 µm. As a result, the surface may be coated with a fine dust that can be washed off and dissolved when in contact with a liquid. Other release mechanisms may be related to long-term release processes. GICs absorb water over time and ion release may occur from the cracks and the body of the GICs and may require more time than estimated.[25] A number of studies have reported that ion release was reduced to constant levels at about 14 days.[18,26,27] Therefore, this study determined the cumulative amount of anions, cations, and silica released over 14 days. Although the same conditions (specimen geometry, temperature, storage medium, and polishing procedure) were established for all the samples, differences were observed in the amounts of ions released. These findings may be related to multiple factors, such as the chemical and physical characteristics of the GICs, the powder:liquid ratio, the solubility of the glass particles, the mixing time, and the type of polymerisation.[11,26]

In release studies, different storage media, such as distilled water, artificial saliva, human saliva, saline, and acidic solutions, have often been used.[18,28,29] Some studies have suggested that saliva or pH-cycling models may better simulate the oral environment and, consequently, be more appropriate for studying ion release from GICs. In spite of this fact, the use of ultra-distilled water (since there are no existing ions in this medium) is considered to give an accurate estimate of the ions released.[30] In addition, the pattern of F release in water was similar to that in artificial saliva or acidic solutions.[18,29] Therefore, ultra-distilled water is acceptable as a storage medium for *in vitro* studies, and, in this study, it was preferred for the release of all the ions and silica.

Ion chromatography and the ion-selective electrode (ISE) are the traditional methods used to measure ion release, especially for F.[31] The ISE measures the total released F ions (free and complexed). Therefore, it is difficult to distinguish between free F ions and F complexes released from GICs. IC estimates the concentration of free F ions.[20] This distinction is important because only free F ions can prevent secondary caries attacks in caries active persons.[10] In this study, two different IC systems were used and only the free ions were measured. The total amount of ions (free and complexed) may be more than the amount detected by the IC method.

Previous studies have shown that TGI released more F than RMGI.[32,33] TGIs are only chemically polymerized and may release more F, whereas RMGIs are set by both an acid-base and a light polymerisation reaction. In other studies, Okte et al.[10] and Gandolfi et al.[23] stated that RMGIs showed significantly greater cumulative fluoride release. In a study by Forss, it was reported that for some of the light curing GICs, the weight gain (water absorption) was large.[24] Therefore, ion release may increase. There are inconsistencies between the results of previous studies. The differences in the findings are mainly related to the extrinsic factors present in each study. In this study, RMGI released a greater amount of F^-, Cl^-, $_4^{3-}$, and Na^+ than other materials. Furthermore, RMGI was the only glass ionomer with the capability of releasing Mg^{2+}. Paschoal et al.[26] showed that nanofilled-RMGI presented a similar cumulative F release pattern to RMGI, and they stated that there was no incremental effect of nanoparticles on the cumulative F release profile. On the other hand, Hasan et al.[22] stated that incorporation of nano-sized particles into glass ionomers can further enhance the F release. They claimed that GCF and TGI exhibit a similar F release and uptake behavior, although GCF releases more F than TGI.

In the current study, GI with nano-sized particles (GCF) released the highest amount of Ca^{2+}. The calcium release from glass ionomers is also very important because F precipitates as calcium fluoride on the tooth surface. This acts as a protective layer and F is released when a drop in pH occurs.[34] In this study, very small amounts of Cl^-, SO_4^{2-}, and K^+ were detected in all the GIs. The effects of these ions on the mouth and hard tissues are unknown. HVGI and TGI contain PO_4^{3-} but no PO_4^{3-} is released into solutions. Advances in material technology and the light curing of other materials can cause this situation.

Even though glass ionomers contain less Na^+ than F^-, Wilson et al.[35] found sodium to be eluted more than F. The results of this study were mainly in line with their conclusions. The most released cation was Na^+ and the most released anion was F^-. As a result, the RMGI with high F^- release and GCF with high Ca^{2+} release may be used as a good alternative to TGIs for bacterial elimination and the prevention of secondary caries attacks.

CONCLUSIONS

This study demonstrated that the amounts of F^-, an anion, and Na^+, a cation, released were significantly higher than the amounts released of the other ions. The newly developed glass ionomer (GCF), which contains nanoparticles, was found to be promising in terms of F^- release compared to the other materials, except for RMGI.

The composition and chemical and physical characteristics of GIs are important for the release of ions and silica. New glass ionomers with a high calcium content can be developed for bacterial elimination and the prevention of secondary caries attacks. There is a need for further *in vivo* and *in vitro* studies.

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