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Original Research

Fluoride Release, Radiopacity and Rugosity in Glass Ionomer Surfaces Subjected to pH Cycling

Liberação de Flúor, Radiopacidade e Rugosidade de Superfície de Cimentos de Ionômero de Vidro Submetidos à Ciclagem de pH

Liberación de Fluoruro, Radiopacidad y Rugosidad Superficial de Cementos de Ionómero de Vidrio Sometidos a Ciclos de pH

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Abstract
The aim of this study was to evaluate fluoride release, radiopacity and rugosity in glass ionomer surface of four restorative glass ionomer cements subjected to pH cycling. Six specimens from each experimental group (Ketac N100, Vitremer, Riva Light Cure, Fuji IX, Riva Self Cure) were first immersed in 2mL of demineralizing solutions (De) for six hours and then placed in the remineralizing solution (Re) for eighteen hours, this cycle repeated for 7 days. Radiopacity and roughness were measured at the beginning and end of cycling. The fluoride released in the Des- and Re- solutions was analyzed at 24-hour intervals over the 7 days. The data were submitted to two-way ANOVA and Fisher's PLSD test ($p<0.05$). The Ketac N100 group showed lower roughness and greater radiopacity before and after pH cycling, when compared to the other groups ($p<0.05$). All glass ionomer cements evaluated showed greater fluoride release on the first day of cycling. The Riva Light Cure, Vitremer and Fuji IX groups had a higher mean fluoride release during the 7 days of cycling when compared to Ketac N100. It was concluded that Ketac N100 presented, before and after cycling, lower roughness and greater radiopacity when compared to the other groups. In addition, it presented fluoride released higher than Riva Light Cure (SDI-L) and Riva Self Cure (SDI-S), but similar to Vitremer (VIT) and Fuji IX (F9).
Descriptors: Glass Ionomer Cement; Radiography; Fluoride; pH.
Resumo
O objetivo do presente estudo foi avaliar a liberação de flúor, radiopacidade e rugosidade na superfície de ionômero de vidro de quatro cimentos de ionômero de vidro restauradores submetidos à ciclagem de pH. Seis espécimes de cada grupo experimental (Ketac N100, Vitremer, Riva Light Cure, Fuji IX, Riva Self Cure) foram primeiramente imersos em 2mL de soluções desmineralizantes (De) por seis horas e então colocados na solução remineralizante (Re) por dezoito horas, este ciclo repetido por 7 dias. A radiopacidade e rugosidade foram medidas no início e no final da ciclagem. O flúor liberado nas soluções Des- e Re- foi analisado em intervalos de 24 horas ao longo dos 7 dias. Os dados foram submetidos à ANOVA bidirecional e ao teste de Fisher PLSD ($p<0,05$). O grupo Ketac N100 apresentou menor rugosidade e maior radiopacidade antes e após a ciclagem de pH, quando comparado aos outros grupos ($p<0,05$). Todos os cimentos de ionômero de vidro avaliados apresentaram maior liberação de flúor no primeiro dia de ciclagem. Os grupos Riva Light Cure, Vitremer e Fuji IX apresentaram maior liberação média de flúor durante os 7 dias de ciclagem quando comparados ao Ketac N100. Concluiu-se que o Ketac N100 apresentou, antes e após a ciclagem, menor rugosidade e maior radiopacidade quando comparado aos demais grupos. Além disso, apresentou liberação de flúor maior que o Riva Light Cure (SDI-L) e o Riva Self Cure (SDI-S), mas semelhante ao Vitremer (VIT) e ao Fuji IX (F9).
Descritores: Cimento ionômero de vidro; Radiografia; Fluoreto; pH.
Resumen
El objetivo del presente estudio fue evaluar la liberación de fluoruro, la radiopacidad y la rugosidad en la superficie de ionómero de vidrio de cuatro cementos restauradores de ionómero de vidrio sometidos a ciclos de pH. Primero se sumergieron seis muestras de cada grupo experimental (Ketac N100, Vitremer, Riva Light Cure, Fuji IX, Riva Self Cure) en 2 ml de soluciones desmineralizantes (De) durante seis horas y luego se colocaron en la solución remineralizante (Re) durante dieciocho horas. Este ciclo se repitió durante 7 días. Se midieron la radiopacidad y la rugosidad al principio y al final del ciclo. El fluoruro liberado en las soluciones Des- y Re- se analizó a intervalos de 24 horas durante los 7 días. Los datos fueron sometidos a un ANOVA de dos vías y a la prueba Fisher PLSD ($p<0,05$). El grupo Ketac N100 mostró menor rugosidad y mayor radiopacidad antes y después del ciclo de pH, en comparación con los otros grupos ($p<0,05$). Todos los cementos de ionómero de vidrio evaluados mostraron una mayor liberación de fluoruro el primer día de ciclado. Los grupos Riva Light Cure, Vitremer y Fuji IX mostraron una mayor liberación promedio de fluoruro durante los 7 días de ciclado en comparación con Ketac N100. Se concluyó que Ketac N100 presentó, antes y después del ciclo, menor rugosidad y mayor radiopacidad en comparación con los otros grupos. Además, presentó mayor liberación de flúor que Riva Light Cure (SDI-L) y Riva Self Cure (SDI-S), pero similar a Vitremer (VIT) y Fuji IX (F9).
Descriptores: Cemento de Ionómero de Vidrio; Radiografía; Fluoruro; pH

INTRODUCTION

Since its introduction by Wilson & Kent[1,2] and the main clinical developments by McLean & Wilson[3-5] glass ionomer cements have been widely used in preventive and restorative dentistry. One of the great qualities of these materials is the release of fluoride over time both in *in vivo*[6-8] and *in vitro* conditions[9-15].

The release of fluoride from glass ionomer cement is considered the likely cause of less dental demineralization around direct restorations[16-19]. Some factors can influence the release of fluoride from ionomeric cements: type and commercial brand, fluorine content of the glass, handling of the material and the acidity of the environment[20,21]

This may suggest that changes in the pH of the environment where glass ionomer cement restorations are observed may lead to variations in the release of fluoride from these materials and consequently a greater or lesser effect on the demineralization and remineralization process (dental caries).

Studies analyzing the release of fluoride in solutions with an acidic pH[22,23] observed a difference in release when compared to other study media such as saliva and water. Therefore, *in vitro* assessments of fluoride release from restorative materials should take into account the change in pH in dental plaque[22,24,25].

When evaluating the anticariogenic activity of materials that release fluoride, a situation closer to clinical conditions must be imitated, in which there is always a dynamic of demineralization and remineralization[22,26]. Several protocols have been used according to the experimental design with variations in the pH of the solutions[18,19,26-28]. This could influence the material's ability to release fluoride and its effect on the *in vitro* caries process. In the oral cavity, the pH of the plaque medium during the demineralization and remineralization process undergoes variations, which are not considered in *in vitro* tests. Thus, with the aim of observing the release of fluoride as close to oral reality, where the pH variation is dynamic, and to support methodologies that simulate cariogenic challenge through pH cycling, we believe it is valid to evaluate the release of fluoride from restorative glass ionomer cements in demineralizing and remineralizing solutions.

Composite resin finishing and polishing procedures are important steps in restorative dentistry. A polished surface minimizes plaque accumulation, gingival irritation and color changes in the composite, contributing to better aesthetics and minimizing the possibility of caries recurrence. According to Bollen et al[29], there would be a surface roughness limit for bacterial adhesion (Ra=0.2µm), above which it would result in the accumulation of bacterial plaque, consequently increasing the risk of caries and periodontal inflammation. Therefore, maintaining a restoration with low surface roughness values becomes essential in achieving a lasting restorative process. The objective of this study was to evaluate the release of fluoride, calcium, phosphorus, radiopacity and surface roughness of restorative glass ionomer cements subjected to demineralizing and remineralizing solutions.

MATERIAL AND METHOD

- *Preparation of test specimens*

The materials tested are listed in Table 1. 18 specimens from each experimental group were manufactured following the manufacturer's recommendations, which were obtained from metallic matrices with a central hole measuring 5mm in diameter and 2mm in height. and coupled to the stainless-steel wire. For Fuji IX and Riva Self Cure materials, pressure was maintained until the material hardened. As for the Ketac N100, Vitremer and Riva Light Cure materials, photopolymerization was carried out using an Ultraled photopolymerizing device (Dabi Atlante), for a period of 40 seconds, on the upper and lower surfaces. After hardening, excess was carefully removed.

Table 1. Distribution of tested materials, according to their classification and manufacturers

Material (abbreviation)	Manufacturer	Material Classification
Ketac N100 - KN	3M Espe	Resin-modified glass ionomer cement with nanoparticles
Vitremer - VIT	3M Espe	Resin modified glass ionomer cement
Riva Ligth Cure – SDI-L	SDI	Resin modified glass ionomer cement
Fuji IX – F9	GC Corporation	conventional glass ionomer cement
Riva Self Cure – SDI – S	SDI	conventional glass ionomer cement

Roughness and pH cycling for release of F, Ca and P from GICs (Source: Research Data)

To analyze the surface roughness, the specimens were taken individually to the SJ-401 portable roughness meter (Mitutoyo). The roughness standard used will be Ra, which represents the arithmetic mean between recorded peaks and valleys. A cut-off of 0.25mm was used, necessary to maximize the filtering of surface waviness, and on each surface, three readings were taken in different positions and the arithmetic mean was calculated before and after the aging procedures.

The specimens were placed in tubes containing 2 mL of demineralization (DE) or remineralization (RE) solutions[26]. Initially, the specimens were stored for 6 hours in DE solution (2.0 mmol/L Ca and P in 0.075 mol/L acetate buffer, at pH 4.7). Then, the samples were transferred to new tubes containing RE solution (Ca 1.5 mmol/L, P 0.9 mmol/L, KCl 0.15 mol/L in cacodylate buffer 0.02 mol/L, at pH 7 ,0) remaining for 18 hours. The

tubes were left under constant agitation (TE-420 Orbital shaking table – Tecnal, Piracicaba, SP, Brazil), at a temperature of 37 ºC. These procedures were repeated for 15 days. The specimens were washed with deionized water and dried with absorbent paper before being immersed in a new solution. The solutions were collected daily, identified and stored in polypropylene test tubes at 4°C to read the F, Ca and P released. After this process, the specimens were taken again to the roughness meter to analyze the final surface roughness.

- *Analysis of F, Ca and P in DE and RE solutions*

To measure fluoride in DE and RE solutions, a specific electrode (9409 BN, Orion Research, Inc., Beverly, MA, United States of America) and reference microelectrode were used, previously calibrated with standard solutions of 0.0625 to 16 mg F/mL, coupled to an Orion 720 A ion analyzer (Orion Research). For dosages, 0.5 mL of DE and RE solutions were pipetted and 0.5 mL of TISAB II (total ionic strength adjusting buffer) were added. Readings were taken under constant agitation on a magnetic stirrer (TE-081, Tecnal). These values were converted to µg F/cm^2. The P released by the materials was measured using the colorimetric method[27], in 96-well plates (Flat-bottom cell culture plate - Model 92096 - TPP, Switzerland). Aliquots of 0.02 mL of the DE or RE solutions were used for readings on a microplate reader (PowerWave 340, Biotek, Winooski, VT, United States of America), at a wavelength of 660 nm. The concentration of P determined in the DE and RE solutions was subtracted from the amount of phosphorus existing in the DE (57.5 ±7.6 µg P/mL) and RE (26.9 ±2.7 µg P/mL) solutions and the final value considered as that arising from P present in the solutions. These values were converted to µg HMP/cm^2. The F and P concentrations of DE and RE solutions were determined separately. Subsequently, the results of the DE and RE solutions were added (DE + RE), completing a period of 24 hours and a cycle of the methodology used (pH cycling), in the 15 days analyzed. The calcium concentrations of the DE and RE solutions were determined by the colorimetric method using Arsenazo III method in 96-well plates (Flat-bottom cell culture plate - Model 92096 – TPP, Switzerland). Aliquots of 0.005 mL of DE and RE solutions were used for readings on a plate reader (PowerWave 340, Biotek), at a wavelength of 650 nm. These values were converted to µg Ca/cm^2.

- *Digital radiographic images*

Digital radiographic images (storage phosphor plate, Digora, Soredex, Orion Corporation, Helsinki, Finland) obtained with a GE-100 X-ray unit (50 kVp, 10 mA and 12 pulses, General Eletric, WI, USA) were used to determine the radiographic density of the specimens, before and after cycling. The target-to-film distance was set to 30 cm. The digital radiographic images were made using an aluminum wedge (Alcan Alumínio do Brazil S.A., Porto Alegre, RS, Brazil) containing 10 steps with different thicknesses (1=0.55; 2=1.05; 3=1.50; 4=2.1; 5=3.05; 6=4.05; 7=5.05; 8=6.05; 9=6.9 mm and 10=7.97) and the radiopacity values were thus expressed in aluminum equivalents (mm Al). Two 2-mm thick longitudinal sections of enamel and dentin were cut from extracted human molar tooth with a low-speed diamond saw. These specimens were stored in water until the moment of use. Three specimens of different materials, two teeth sections, aluminum stepwedge and metal code letter/number were placed on the phosphor plate.

The images were analyzed using the Digora 1.51 software (Orion Corporation Soredex, Helsinki, Finland). A standardized area (in pixels) was established for each item analyzed: specimen (12x34 pixels); enamel (12x12 pixels); dentin (12x12 pixels); and aluminum step wedge (78x20 pixels). The radiographic density was measured three times and arithmetic mean values were obtained, corresponding to the radiopacity value of the item.

RESULTS

In table 2, lower initial and final roughness was observed for the KETAC N100 group when compared to the other groups ($p<0.05$). The Fugi IX group showed a statistical difference between the initial and final roughness after seven days of cycling ($p<0.05$).

Table 2. Dosages of F, Ca and PO_4^{-3} in each glass ionomer group

Material	Fluoride (µg/L)	Calcium (µg/L)	Phosphate (µg/L)
Vitremer	7,4 (6,6) a	-15,8 (26,4) b	-3,0 (10,2) b
Fuji IX	7,1 (7,9) a	-26,4 (23,8) a	-6,7 (7,4) a
Ketac N100	3,7 (4,1) b	-26,1 (11,0) a	-2,0 (6,5) b
Riva Light	7,1 (8,4) a	-16,8 (23,7) b	-2,5 (6,5) b
Riva Self	3,6 (2,5) b	-25,9 (17,1) a	-1,6 (5,4) b

Source: Research Data

After pH cycling, the VITRIMER, FUJI IX and Riva Light groups showed a higher mean fluoride release when compared to KETAC N100 and Riva. All groups showed greater fluoride release on the first day of cycling, decreasing after this period and remaining constant from the third day onwards.

According to figure 1, the surfaces of all groups remained the same after pH cycling, presenting no statistical difference before and after. The VIT group presented the highest roughness values among all groups, while KN presented the lowest. The SDI-L, F9 and SDI-S groups presented intermediate values, falling between the VIT and KN range.

KETAC N100 showed greater initial and final radiopacity, statistically differentiating itself

from the other groups (p<0.05). pH cycling did not influence the radiopacity of the tested materials. The other groups did not show a statistically significant difference between them, remaining in the same radiopacity range (Figure 2).

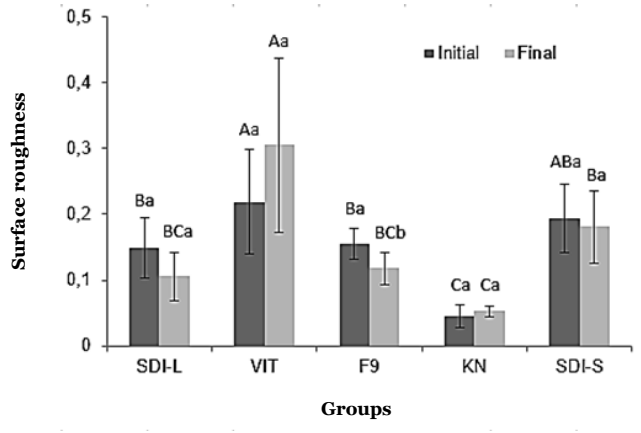

Figure 1. Surface roughness in each glass ionomer cement group before and after pH cycling.

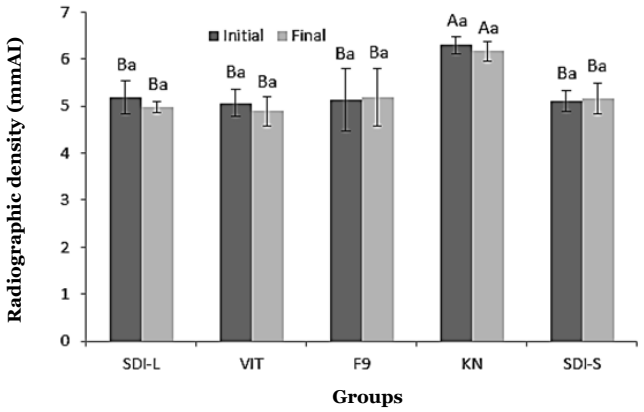

Figure 2. Radiographic density (mmAl) in each glass ionomer cement group before and after pH cycling.

DISCUSSION

All groups maintained the initial roughness of their surfaces after pH cycling, indicating that the conditions for biofilm formation were not changed between variations.

In comparison between groups, Vitremer (VIT) demonstrated considerable surface roughness, inherent to this material from the beginning, increasing its biocompatibility and providing an environment more conducive to the formation of biofilms and microbial colonization; on the other hand, there was a high release of F, which has antimicrobial properties. Differently, the Ketac N100 (KN) group presented the lowest roughness values, at the same time as it presented low F release. The Riva Light (SDI-L) and Fuji XI (F9) groups presented low final roughness and high release. of F. Finally, the SDI-S group demonstrated intermediate final roughness with low release of F. Considering exclusively these characteristics of the material, it is possible to know its behavior under pH variations in the oral cavity.

The level of radiopacity of the material can indirectly influence oral health, allowing good monitoring of the integrity of the glass ionomer through imaging tests, facilitating diagnoses and safe procedures. In this way, the groups maintaining their radiopacities unchanged, even after the pH variation (4.7), also implies significant changes in their compositions and structures, simultaneously in which there was release of F, PO_4^{-3} and Ca, whose presence in saliva modulates the formation of biofilm and consequent caries lesion, also helping to remineralize dental tissue and reduce its demineralization.

The analysis of the formation and composition of biofilms in cements in relation to the properties of the materials could provide important information about the impact of roughness and release amounts of F, PO_4^{-3} and Ca, however, despite these limitations in the study, the physical and chemical properties of cements were measured.

CONCLUSION

The radiopacity and roughness of each ionomer did not vary significant and there was greater release of F in the Vitremer, Fuji IX and Riva groups Light, depending on the pH decrease.

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CONFLICT OF INTERESTS

The authors declare no conflict of interest.

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