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Fluoride Release from Restorative Materials Coated with an Adhesive

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The retention of both fluoride resins and resin-modified glass ionomer cements to dental tissues can be improved by the association of an adhesive system which promotes the bonding between the resin component and dentin, forming a hybrid layer. The purpose of this investigation was to evaluate if the presence of the adhesive, being part of the hybrid layer composition, interfered with the fluoride released to tooth tissues. The restorative materials studied were: Vitremer (3M), Heliomolar (Vivadent) and Z100 (3M) using an adhesive application (Scotch Bond MultiPurpose Plus - 3M). Ten discs of each material were prepared: 5 were covered with the adhesive and 5 were not. The discs were immersed in individual flasks containing artificial saliva which was changed daily. Fluoride release was measured at days 1, 5, 10, 15 and 20 by a fluoride combined electrode (9609 BN - Orion) coupled to an ion analyzer (SA-720 Procyon). One-way ANOVA and the Student-Newman-Keuls test were applied to compare the materials. The results showed that the use of a dental adhesive significantly decreased the fluoride release of Vitremer and reduced the fluoride release of Heliomolar to undetectable levels with the methodology used.

Key Words: fluoride release, resin-modified glass ionomer cement, composite resin, adhesives.

INTRODUCTION

Since their development in the early 1970's, glass ionomer cements have been modified in order to improve physical properties and facilitate application (1). The addition of resin increases versatility and the advantages of glass ionomer cements are their biocompatibility, adhesion to the tooth structure, hydrophilic nature, low solubility in the oral environment and fluoride release (1).

Fluoride release from both conventional and modified glass ionomer cements is similar (2) and confers an anticariogenic potential. This effect is of value in preventing secondary caries, even under conditions of high caries risk (3-5). The pattern of fluoride release is similar in most glass ionomer cements (6): a high initial rate followed by a fast decrease from 24 to 48 h, reaching a constant level lasting up to 2 years (7). Some studies also show the capacity of being recharged by the uptake of fluoride from toothpastes and solutions (3,7,8).

Resin compounds are materials with greater enamel and dentin bond strength than glass ionomer cements (9). Composite resin materials containing fluoride fillers such as YbF_3 have been developed (3,10). However, fluoride release from these materials is significantly lower than that achieved with glass ionomer cements (8). To increase enamel and dentin bond strength, reducing microleakage, resin-modified glass ionomer cements were developed. Although greater than conventional cements, the adhesion of resin-modified glass ionomer cements is still not equivalent to modern composite resins associated to adhesive systems which promote hybridization (11). Hybridization is a diffuse transition zone between the bulk resin phase and the underlying mineralized dentin. This is called the hybrid layer or the resin-reinforced layer because there is a diffuse penetration of resin into the deminer-

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alized matrix where the monomer envelops exposed collagen fibrils. When properly created, it is uniformly continuous and provides a strong union between resin and dentin (12). In addition, it creates a condition by which fluid or microorganisms cannot enter the dentinal tubules, thus eliminating both microleakage and postoperative sensitivity.

In order to increase dentin bond strength of resin-modified glass ionomer cements, White (11) suggests the association of adhesive agents with these materials. Pereira et al. (13), comparing the use of resin-modified glass ionomer cements with or without hybridization, found a significant increase in shear bond strength when associated with hybrization, thus improving clinical performance.

Mukai et al. (5) observed that resin-modified glass ionomer cement increased fluoride in dental tissues adjacent to restorations *in vivo*. These beneficial effects would be lost if the hybrid layer is impervious to fluoride ions.

Sano et al. (14) reported a special kind of leakage in a porous zone of the hybrid layer basal portion, calling it nanoleakage. Most microleakage studies involve measuring the magnitude of movement of a tracer molecule through a gap between restorative materials and the wall of cavity preparations. The microscopic study by Sano and coworkers (14) examined the migration of silver nitrate into the interface between dentin and five different dentin bonding agents used to restore class 5 cavities, in the absence of gap formation. Several different leakage patterns were seen, however they all indicated leakage within the hybrid layer when viewed by scanning electron microscopy. Thus, silver ions penetrated into demineralized dentin of the hybrid layer in all cases suggesting that fluoride ions which have similar dimensions to silver ions (15) can also penetrate the hybrid layer.

The purpose of the present study was to evaluate fluoride release from restorative materials using an adhesive system which, being part of the hybrid layer composition, could act as a barrier against fluoride access to tooth tissues near restorations.

MATERIAL AND METHODS

The restorative materials used in this study were one resin-modified glass ionomer cement (Vitremer), one resin with fluoride in its composition (Heliomolar) and one resin without fluoride (Z100, negative control) (Table 1). Ten discs (6.0 mm in diameter x 2.0 mm thick, area: 0.94 mm^2) were prepared of each material in a cylinder teflon mold. The glass ionomer cement was mixed according to manufacturer recommendations and placed in the mold with a Centrix syringe between two glass plates. The resins were placed in the mold in bulk and pressed between the two glass plates. Orthodontic wire was incorporated into the materials before setting to suspend the samples in the testing medium. All materials were light cured (L1500, 3M, St. Paul, MN, USA) in three different positions, each side for 40 s in order to ensure the complete cure of the material. Subsequently, five discs of each material were chosen at random and covered with one layer of Scotch Bond MultiPurpose Plus adhesive (SBMPP; 3M) that was light cured in three different positions on both sides for 20 s each (test group). The other discs without the adhesive protection formed the control group. The discs were then stored at 100% relative humidity and 37^{o}C for 24 h. Each sample was subsequently suspended in a plastic flask containing 4 ml of artificial saliva with the following composition: 0.400 g NaCl (no. 1), 0.400 g KCl (no. 7), 0.795 g $CaCl_2H_2O$ (no. 75), 0.690 g $NaH_2PO4.H_2O$, 0.005 g $Na_2S.9H_2$, 0.200 g NaN_3. Every twenty-four hours for 20 days, the discs were dried with filter paper and immediately immersed in new 4 ml of artificial saliva. The flasks were maintained at 37^{o}C without agitation.

Fluoride release was measured at 1, 5, 10, 15, and 20 days after buffering 1 ml of saliva with an equal volume of Tisab II (total ionic strength adjustor), with the following composition: 57 ml acetic acid, 58 g NaCl (no. 1), 4 g CDTA (no. 23), NaOH in an amount to complete 1000 ml. The measurements were made with a fluoride ion-specific electrode (9609 BN, Orion Re-

Table 1. Materials used in this study.

Material	Product	Batch No.	Manufacturer
Glass ionomer cement	Vitremer	19930823	3M Dental Products St. Paul, MN, USA
Composite resin	Z100	5904A3	3M Dental Products St. Paul, MN, USA
Composite resin	Heliomolar	052715	Ivoclar Vivadent Schaan, Liechtenstein
Adhesive system	Scotch Bond MultiPurpose Plus	19950112	3M Dental Products St. Paul, MN, USA

search Inc., Boston, MA, USA) and an ionanalyzer (Orion SA-7200) after previous calibration with fluoride standard solutions of 1 and 10 ppm (16). Two samples of each tube containing artificial saliva were measured. If the variation between these two samples was higher than 5%, a third sample was measured. Before making fluoride measurements, three vials containing artificial saliva and Tisab II were tested to determine baseline fluoride concentration of the artificial saliva. The mean baseline concentration (0.034 ppm) was subtracted from each concentration obtained from the samples. Total fluoride release was divided by the area of the disc, giving all recorded units in µg F/cm^2.

Means and standard deviations were significantly correlated (r = 0.9355), suggesting a log transformation to control data heterogenity. Data were analyzed by one-way ANOVA to compare the different materials at each time period. The Student-Newman-Keuls test was used to identify group differences for each ANOVA.

RESULTS

Table 2 reports the fluoride release of the restorative materials after exposure to artificial saliva for 24 h, 5, 10, 15 and 20 days. The fluoride released from Heliomolar + SBMPP, Z100 and Z100 + SBMPP was considered nonexistent because it was less than 0.026 µgF/cm^2, which is the limit of the electrode sensitivity.

At 24 h, Vitremer released significantly more fluoride than the other products studied, followed by the group in which this material was covered by the adhesive. The fluoride-containing resin Heliomolar released significantly less fluoride than the other two. At day 5, fluoride release decreased with significant differences among all 3 groups. At the subsequent days (10, 15 and 20) Vitremer continued to release significantly more fluoride than the others. At these time points, there was no significant difference between Vitremer + SBMPP and Heliomolar.

Vitremer presented an initial high fluoride release without significant statistical differences during the first five days of the experiment. After that, the fluoride release decreased significantly. In the other two groups (Vitremer + SBMPP and Heliomolar), the decrease in fluoride release was statistically significant after the first day compared to the subsequent time points.

DISCUSSION

The *in vitro* model used in this study intended to reproduce an *in vivo* situation that associates the hybridization process with a resin-modified glass ionomer cement. Despite the fact that this technique is not indicated by the manufacturer, it would offer the advantages of increased retention and a consequent decrease in microleakege, thus reducing the risk of secondary caries. However, this model diverges from the clinic situation because the fluid resin layer over the discs was probably thicker, more homogeneous and less porous than the resin component of the hybrid layer *in vivo*.

Dentin is an organic tissue that also makes the hybrid layer not totally impervious. Probably the existing difference is the presence of a porous zone in the basal portion of the hybrid layer (14). The results of this study show a statistical difference in fluoride release between the groups treated and their controls. There-

Table 2. Mean fluoride release and standard deviation, in µgF/cm^2, of the different materials studied, after 24 h, 5, 10, 15 and 20 days.

Material	24 h	5 days	10 days	15 days	20 days
Vitremer	0.8878 ± 0.2587[a]	0.7570 ± 0.1292[a]	0.5270 ± 0.1038[a]	0.2003 ± 0.0407[a]	0.1310 ± 0.0251[a]
Vitremer +SBMPP	0.2230 ± 0.1354[b]	0.0895 ± 0.0247[b]	0.0110 ± 0.0117[b]	0.0365 ± 0.0033[b]	0.0110 ± 0.0067[b]
Heliomolar	0.1115 ± 0.0846[c]	0.0305 ± 0.0084[c]	0.0110 ± 0.0056[b]	0.0230 ± 0.0018[b]	0.0010 ± 0[b]
Heliomolar+SBMPP*	-	-	-	-	-
Z100*	-	-	-	-	-
Z100+SBMPP*	-	-	-	-	-

*The values found did not reach the sensitivity limit of the electrode (0.026 µgF/cm^2). Data with different letters are statistically different comparing groups ($p<0.05$). SBMPP = Scotch Bond MultiPurpose Plus adhesive.

fore, the adhesive layer reduced but did not prevent fluoride passage. In the case of the resin Heliomolar, the adhesive reduced fluoride release to such an extent that it could not be detected by the methodology used.

Araújo et al. (17) also described less fluoride release when they covered Vitremer with an adhesive. However, they refer to this "coverage" as if it could function as a protective barrier against dehydration and that, in the clinic, this bonding agent could be lost due to wear produced by chewing, occlusal grinding and toothbrushing. However, they believed that after a period of time of exposure to this environment, fluoride release could be similar to the unprotected Vitremer.

The findings of the present study are in agreement with Burgess et al. (18) who reported that a layer of adhesive considerably reduces fluoride release and that the application of the light-cured primer used with Vitremer did not inhibit this release. Extrapolating, these authors believe that the primer would not prevent the fluoride passage to the cut dentin surface.

Tam et al. (19) evaluated the effect of the intermediary adhesive agents on the development of caries adjacent to resin-modified glass ionomer cement restorations. In the groups in which only acid and primer were applied, fluoride penetrated deeper into the dentin. The additional application of an adhesive layer reduced this penetration but definitely did not prevent it. The presence of microgaps or of some material between the glass ionomer cement and the tooth surface could, theoretically, affect the fluoride penetration to the adjacent tooth tissues. Components and fluids present at these intermediary gaps or in this material layer could potentially restrict or promote fluoride passage. Thus, the use of dentin adhesive agents was suggested to mediate the union of the ionomer and dentin. It is interesting to highlight the results of Lee et al. (20) who demonstrated that an adhesive system is not susceptible to an *in vitro* chemical breakdown (hydrolysis) caused by artificial saliva. When resin-modified glass ionomer cement was compared to fluoride-containing resin, the use of an adhesive system, *in vitro*, completely annulled the fluoride release of the resin. Yet, Tam et al. (19) found that everytime glass ionomer cement was tested, including with hybridization, there was an apparent zone of sound dentin between the material and the body of the lesion. In the resin, the lesion continued directly from the restoration.

The application of an adhesive significantly reduced the fluoride passage from the resin-modified glass ionomer cement Vitremer and the amount of fluoride released from Heliomolar was so reduced that it was undetectable by the methodology used. However, fluoride release was still observed in the Vitremer group to a lesser extent. The minimal quantity of fluoride necessary to be clinically significant to inhibit the caries process is not well established (17); however, the use of hybridized glass ionomer cements still presents the capacity of fluoride release which is advantageous in relation to the use of a fluoride-containing resin. Taking into account that increased adhesion and reduced microleakage could lead to better benefits clinically than the fluoride release *per se*, it could be inferred that the hybridization process used with resin-modified glass ionomer cements is a technique to be considered. However, research is still necessary, including *in vivo* studies, to verify the feasibility of the technique.

RESUMO

Miranda LA, Weidlich P, Samuel SMW, Maltz M. Liberação de flúor de materiais restauradores cobertos com adesivo. Braz Dent J 2002;13(1):39-43.

A retenção aos tecidos dentais, tanto de resinas fluoretadas, quanto de cimentos de ionômero de vidro modificados por resina, pode ser melhorada pela associação de um sistema adesivo, promovendo a formação da camada híbrida. Esta técnica traria vantagens desde que a presença do adesivo, sendo parte da camada híbrida, não funcionasse como uma barreira contra a passagem de íons flúor até a superfície dental próxima às restaurações. O objetivo do presente estudo foi avaliar a liberação de flúor dos materiais restauradores Vitremer (3M) e Heliomolar (Vivadent), tendo como controle a resina composta Z100 (3M), através de uma aplicação de um adesivo (Scotch Bond Multi Purpose Plus - 3M). Dez discos de cada material foram preparados: 5 foram cobertos com adesivo, enquanto os demais não. Os discos foram imersos em frascos separados contendo saliva artificial, sendo esta trocada diariamente. As medidas de liberação de flúor foram feitas nos dias 1, 5, 10, 15 e 20 por um eletrodo combinado de flúor (9609 BN - Orion) coplado a um analisador de íons (A-720 Procyon). Testes ANOVA de uma via e SNK foram aplicados aos resultados para comparar os perfis de cada material. Os resultados mostraram que, com a metodologia empregada, o uso de um adesivo dental foi capaz de reduzir significantemente a liberação de flúor do material Vitremer e reduzir a níveis não detectáveis o flúor liberado pela resina fluoretada.

Unitermos: liberação de flúor, cimento de ionômero de vidro modificado por resina, resina composta, adesivos.

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