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THE POTENTIAL OF FLUORIDE-RELEASING DENTAL RESTORATIVES TO INHIBIT ENAMEL DEMINERALIZATION: AN SEM STUDY

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A b s t r a c t: *Objectives*: The study was aimed at determining the effectiveness of fluoride-releasing materials (conventional and resin-modified glass-ionomers, compomer and fluoride-releasing composite resin) in inhibiting demineralization of restored teeth in an artificial caries medium.

Methods: A total of 72 teeth (36 deciduous and 36 permanent) were used and Class V cavities were prepared on each tooth. These cavities were restored with or without conditioning (except for the composite, where all specimens were conditioned). The teeth were then stored in artificial saliva for periods of 1, 6, 12 and 18 months before being exposed to an acidic artificial caries gel and examined by SEM.

Results: In the absence of a restoration, teeth were found to undergo enamel demineralization. Conventional glass-ionomer cements were found to inhibit this significantly. The resin-modified glass-ionomer generally had little effect, except for the18 month specimens, which also showed distinct zones of inhibition. The compomer showed no inhibition, and the fluoride-releasing composite resin showed only limited signs of inhibition.

Conclusions: Glass-ionomers, both conventional or resin-modified, are more effective at protecting the tooth against further decay than either compomers or fluoride-releasing composites, with the best protection of all being provided by conventional glass-ionomers. The nature of the tooth had no influence on these outcomes.

Key words: Dental restoratives, artificial caries, electron microscopy, fluoride release.

Introduction

Dental caries is known to occur when the equilibrium between demineralization and remineralization at the tooth surface is shifted in favour of demineralization. The caries process includes infection by micro-organisms, leading to acid production as a result of the metabolism of carbohydrates by these organisms within the oral biofilm. Acids, of which lactic acid is the main one, attack the mineral phase of the tooth, leading to demineralization, with subsequent degeneration of the organic component and formation of a cavity [1]. Before the development of the cavity, a carious lesion looks like a white spot with a relatively intact, mineral-rich, but porous surface. It covers a subsurface area with a reduced mineral content [2].

Restorative materials are placed to repair frank cavities in teeth that have been damaged by caries. Modern materials are typically designed to be resistant to secondary caries and to micro-leakage at the edges, properties they possess on account of their ability to release fluoride and to be bonded to the prepared tooth surface. Margins of restorations are of particular importance, and lack of integrity of these may significantly increase the risk of secondary caries [3, 4, 5]. Composite restorations are much in demand on account of their excellent aesthetics, but because they undergo polymerization shrinkage on setting, they are associated with marginal leakage, and this leads to bacterial penetration and further damage to the tooth [6]. Secondary caries is, in fact, the most frequent indication for replacement of all types of restoration [3, 7] and the limited durability of dental restorations means that some patients are in continuous restorative cycles that result in larger and larger restorations and more complex therapeutic measures.

As a means of protection against recurrent caries, fluoride-releasing restorative materials have been developed. Of these, the most important are the glass-ionomer cements and their hybrids (resin-modified glass-ionomer cements and polyacid-modified composite resins; so-called "compomers"). By releasing fluoride, these materials offer protection to the hard dental tissues[8] and the surrounding micro-environment [9, 10].

There are a number of mechanisms by which release of fluoride protects the teeth. First, the presence of small amounts of fluoride in the saliva reduces the solubility of the mineral phase of the tooth mineral. Second, fluoride incurporated into the mineral phase leads to the formation of a thin layer of fluor-apatite, which is less soluble even at low values of pH than hydroxyapatite. Third, fluoride may interfere with the metabolism of cariogenic bacteria by inhibiting essential enzyme-mediated processes. All of these mechanisms shift the demineralization/remineralization equilibrium back in favour of remineralization [11, 12].

Glass-ionomers, for example, have been reported to contribute to the remineralization on incipient enamel lesions *in vitro* [13]. Such studies on the effects of fluoride on dentine reveal that low fluoride concentrations may lead to hypermineralization of dentine [14, 15]. In fact, the choice of the restorative material can be crucial in determining whether demineralization or remineralization occurs in the dentine tissue surrounding a restoration. Incipient caries-like lesions under glass-ionomer restorations have been found to remineralize and even to hypermineralize, whereas amalgam and composite restorations have been shown to be predominantly associated with further remineralization of the specimens [12]. The distinct zone of interaction found between the glass-ionomer cement and hard dental tissues contributes to the adhesion and high resistance to microleakage of glass-ionomer cements restorations.

The present study was carried out to determine the detailed effects of fluoride-releasing restorative materials on the progress of demineralization. It employed an artificial caries medium of the type frequently used in such research [15] and involved studies of the appearance of the demineralization inhibition zone in deciduous and permanent teeth filled with different fluoride-releasing dental restorative materials.

Materials and methods

A total of 72 teeth (36 deciduous and 36 permanent) were used in this investigation. They were extracted either due to exfoliation (deciduous teeth) or for orthodontic reasons (young permanent teeth). After extraction, the surfaces of the teeth were cleaned, the radices cut with a diamond bur with water cooling in the level of the cemento-enamel junction, and the remnants of the pulpal tissue discarded. Class V cavities were prepared on each tooth using diamond bur and turbine with water cooling. After preparation, the teeth were divided into five groups at random and filled with one of five different materials, as shown in Table 1.

For most of the materials, each of the groups was divided into two subgroups; the first was conditioned, and the other left unconditioned. In the composites group, by contrast, all of the specimens were conditioned. The conditioning and the filling were carried out in accordance with the manufacturer's instruction; these are also listed in Table 1.

The teeth were stored in artificial saliva designed for use in dental materials testing [19]. The composition is given in Table 2.

The prepared teeth were examined after 1 month, 6 months, 12 months and 18 months. The specimens, after the storage time interval, were placed in an acidic artificial caries gel, prepared according to the method of Arends *et al.* [20]. It consisted of: 6% by weight hydroxyl-ethyl cellulose; 0.1 mol/l lactic acid and 1.0 mol/l NaOH, adjusted to pH = 4.5. The teeth were stored in this gel for 5 days at 37˚C. The extent of the dentine demineralization was then determined with SEM investigation.

Table 1 – Табела 1

Restorative materials employed
Реставраторски материјали употребени во студијата

Material	Type	Conditioning	Manufacturer
Fuji IX	Conventional glass-ionomer cement	1. GC Cavity Conditioner (application 10 sec., rinsing and soft drying)	GC, Japan
Fuji II LC	Resin-modified glassionomer cement	1. GC Cavity Conditioner (application 10 sec., rinsing and soft drying)	GC, Japan
EGIC	Conventional glassionomer cement	Poly (acrylic) acid (ex. Aldrich, Poole, Dorset, UK) (application 10 sec, rinsing and soft drying)	Experimental material
Dyract AP	Polyacid-modified composite resin (compomer)	1. 37% phosphoric acid (application15 sec. on enamel, 5 sec. on dentine, rinsing) 2. Prime&Bond NT (*first layer* – application 30 sec., elimination of the surplus with air blow, polymerization 10 sec.; *second layer* – application, elimination of the surplus, polymerization 10 sec.)	Dentsply, Konstanz, Germany
Unifil flow	Fluoride-releasing composite	GC Unifil Bond (*first layer* – self-etching primer – application, 20 sec., drying 5 sec., not rinsing; *second layer* – bonding-application and polymerization)	GC, Japan

Table 2 – Табела 2

Components of the artificial saliva
Состав на артифициелната плунка

components	concentration (g l^{-1})
NaCl	0.50
$NaHCO_3$	4.20
$NaNO_3$	0.03
KCl	0.20

Specimens were prepared for SEM examination as follows: After drying, the teeth samples were mounted onto aluminium stubs with conducting carbon cement and subsequently coated with a thin layer of gold in a sputter coater (Model Edwards 150B). The specimens were viewed and evaluated under High Resolution Scanning Electron Microscope (Model Cambridge Stereoscan 360, Cambridge Instruments Co. UK) at magnifications of × 1000, × 3000 and × 5000, and images recorded.

Results

In the absence of a restorative material, teeth exposed to artificial caries gel were found to undergo varying degrees of enamel demineralization (Figure 1). Under these circumstances, the crystals building up the enamel rods were not densely packed and lacked distinct borders.

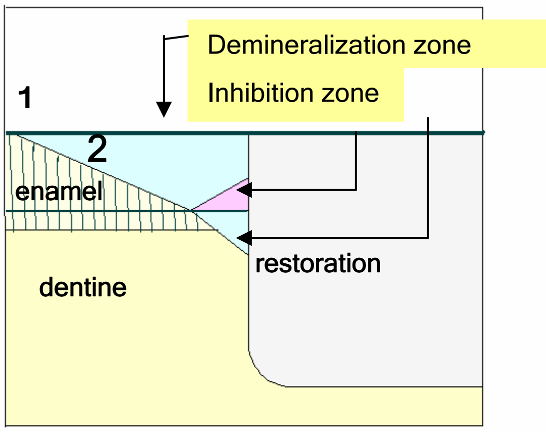

Figure 1 – Schematic presentation of the experimental model: 1. transitional zone between sound and demineralized enamel, 2. completely demineralized enamel
Слика 1 – Шематски приказ на експерименталниот модел: 1. транзициона зона меѓу цврстиот и деминерализираниот емајл, 2. комплетно деминерализиран емајл

By contrast, in the presence of glass-ionomer cement, there were zones with defined enamel prisms (Figures 2–4), and evidence that demineralization had been significantly inhibited. Figure 2 shows the results for the experimental glass-ionomer, where these effects were apparent, despite the loss of the restorative from the cavity. Figure 3 shows the results for the conventional glass-ionomer Fuji IX and Figure 4 for the resin-modified glass-ionomer Fuji II LC. In the case of Fuji IX, there is evidence of enamel decomposition with development of porosity, but these specimens also show mineral deposition close to the base of the restoration. By contrast, the specimens restored with Fuji II LC shown no signs of remineralization, though the specimens stored for

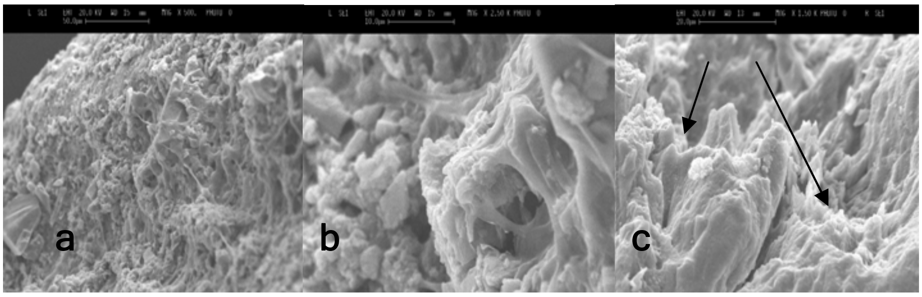

Figure 2 – SEM images of demineralized enamel with (a,b) thin and irregular enamel rods with indistinct borders and (c) wide inter-rod zones. The arrows point towards the basements of the fractured enamel rods
Слика 2 – Микрофотографии на деминерализираниот емајл добиени со SEM со (a, b) тесни и ирегуларни емајлови призми со нејасни граници и (c) широки интерпризматски простори. Стрелката е насочена кон базите на фрактурираните емајлови призми

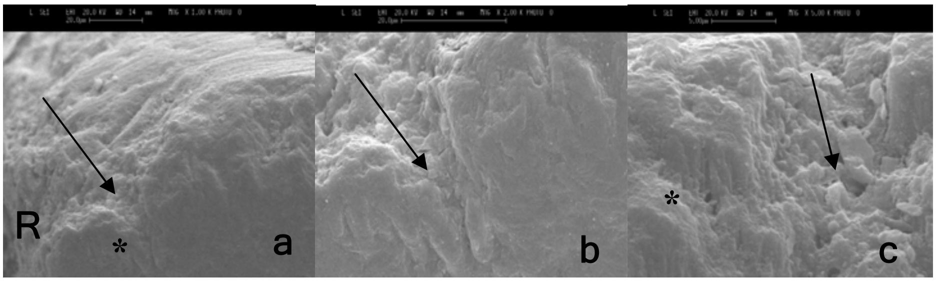

Figure 3 – Restored with experimental glass-ionomer (restoration lost). There is evidence of inhibition of demineralization inhibition and of mineral deposition.
Слика 3 – Реставрација со експериментален глас-јономер цемент (реставрацијата е загубена). Се забележува инхибиција на деминерализацијата и депозиција на минерали

18 months were somewhat different from the others, and did show a significant inhibition of demineralization (Figure 5).

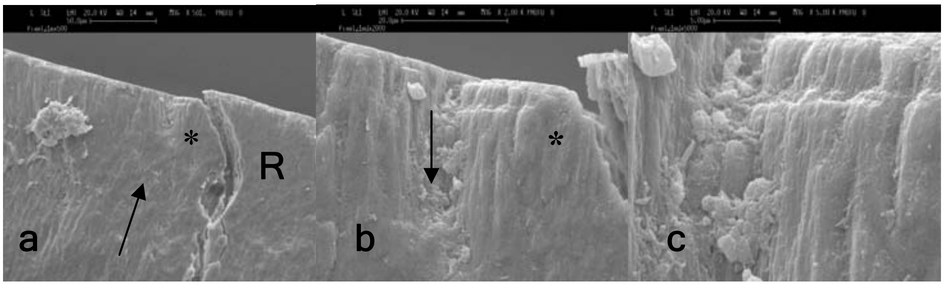

Figure 4 – Restored with Fuji IX glass-ionomer cement, with arrows showing the demineralized areas. (Note: the marginal gap resulted from desiccation during preparation of the specimen for the SEM)

Слика 4 – Реставрација со Fuji IX глас-јономер цемент, со стрелки кои се насочени кон деминерализираните зони. (Забелешка: маргиналниот јаз е резултат од исушување за време на подготовката на примероците за SEM)

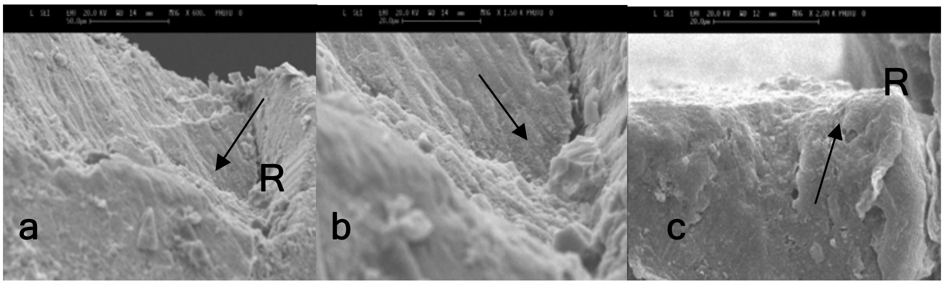

Figure 5 – Restored with Fuji II LC, a, b. showing a wedge-shaped area of demineralization adjacent to the restoration and no signs of remineralization; c. Fuji II LC at 18 months, with a distinct inhibition zone indicated by arrows

Слика 5 – Реставрација со Fuji II LC, a, b покажува клинеста зона на деминерализација близу до реставрацијата без знаци на реминерализација; c. Fuji II LC по 18 месеци, со јасна зона на инхибиција прикажана со стрелка

Figure 6 shows the results for the compomer Dyract AP. In these specimens, there is no evidence of inhibition of demineralization adjacent to the filling, unlike the results observed for the conventional glass-ionomers. Figure 7 shows the results for the fluoride-releasing composite resin Unifil Flow, and here there are only slight signs of inhibition of demineralization. There is some similarity with the observations in Figure 1, in that the enamel rods are indistinct, and the inter-rod spaces are wide.

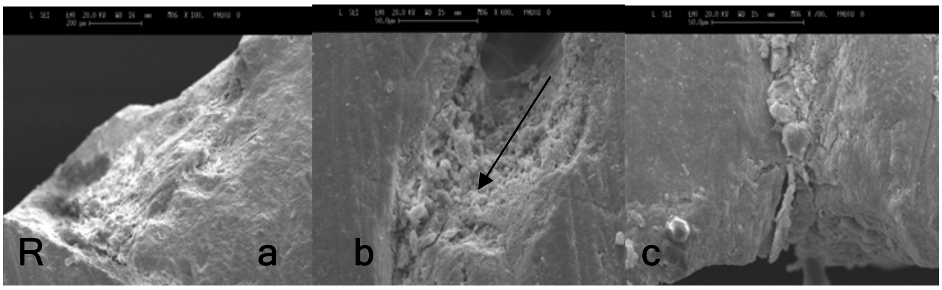

Figure 6 – Restored with Dyract AP and showing complete absence of inhibition of demineralization
Слика 6 – Рес̅шаврација со Dyract AP кај која се забележува ком̅пле̅шно о̅шсус̅шво на инхибиција на деминерализација

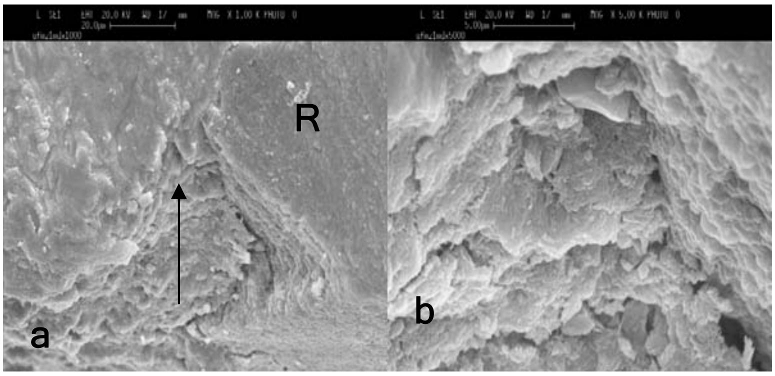

Figure 7 – Restored with Unifil Flow and showing demineralization adjacent to the restoration
Слика 7 – Рес̅шаврација со Unifil Flow која ̅покажува деминерализација во близина на рес̅шаврација̅ша

Discussion

Since the introduction of the fluoride-releasing restoratives, there have been several studies showing that they inhibit caries within enamel structures. Enamel demineralizes at a much slower rate than dentin, therefore, fluoride-releasing restoratives are especially effective at inhibiting caries at enamel margins [18, 21].

The enamel adjacent to the restorations displayed two different types of behaviour. This SEM study indicated that the basic enamel structure, with hydroxyapatite crystals forming rods and inter-rod enamel, could be damaged, making the borders of the enamel rods indistinct and the inter-rod zones hardly visible. Alternatively, the rods can become very thin with wide inter-rod zones. This featureless and amorphous appearance of enamel has been shown in the completely decomposed zones.

The enamel is formed during two main phases of ameloblastic activity, the formative secretory phase when the matrix is laid down and the maturation phase when the mineralization mainly occurs. The enamel crystals are laid down during the secretory stage, and the matrix architecture is probably the primary determinant for their orientation and growth. During the maturation stage, the crystals increase in width and thickness [22]. There are chemical and structural differences between primary and permanent dentine. In particular, it has been shown that permanent dentine is more mineralized than primary dentine [23, 24]. However, in our study we used young immature permanent teeth in which the maturation process is not yet complete, which meant that we could not recognize any difference between the extent of demineralization or its inhibition between the deciduous and permanent teeth. Also, we observed no differences between the conditioned and unconditioned samples.

In our study, glass-ionomer cements were found to inhibit demineralization but, by contrast, compomers did not. We attribute this to the fact that primers are needed to bond the compomers in place surface and these form a layer that blocks diffusion of fluoride into the dentine surface [18]. The composite system in the present study was more satisfactory, because both the adhesive and the composite resin (Unifil Bond and Unifil Flow) were fluoride-releasing, and were thus able to provide fluoride ions to the dentine surface below the restoration. This enabled them to maintain the integrity of the cavity wall. However, despite this, there was still a significant amount of demineralization. Hence, despite their fluoride release, they were not effective in preventing the appearance of caries. This may be because their release pattern differs from that of glass-ionomers, and is steady over time, rather than showing a large initial burst of fluoride release.

Overall, the results suggest that conventional glass-ionomer cements provide the best protection against demineralization of all the materials examined. This suggests in turn that it is not release of fluoride alone which inhibits demineralization, but the overall pattern of fluoride release with time, and the ease of fluoride transport into the enamel immediately surrounding the restoration. Further research is needed to fully elucidate the factors that contribute to effective anti-caries properties in restorative materials, and the reasons for the observed superiority of conventional glass-ionomer cements in this regard.

Conclusions

Zones of inhibition of demineralization were found around conventional glass-ionomer restorations after ageing and exposure to artificial caries medium in both deciduous and young permanent teeth. For resin-modified glass-ionomer restorations, such zones of inhibition were only apparent in specimens aged for 18 months. By contrast, there were no such zones under any circumstances around either compomers or fluoride-releasing composite resin. Also, no differences were observed between primary and permanent teeth, or between conditioned or unconditioned surfaces. We therefore conclude that glass-ionomers, both conventional or resin-modified, are more effective at protecting the tooth against further decay than either compomers or fluoride-releasing composites, with the best protection of all being provided by conventional glass-ionomers.

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Р е з и м е

ПОТЕНЦИЈАЛОТ НА ФЛУОРОСЛОБОДУВАЧКИТЕ ДЕНТАЛНИ РЕСТАВРАТОРСКИ МАТЕРИЈАЛИ ВО ИНХИБИЦИЈАТА НА ДЕМИНЕРАЛИЗАЦИЈА НА ГЛЕЃТА: СТУДИЈА SEM

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Цел на трудот: Студијата има за цел да ја одреди ефикасноста на флуорослободувачките дентални реставраторски материјали (конвенционален и глас-јономер цементи модифицирани со смола, компомер и флуорослободувачки композит) во инхибиција на деминерализацијата на реставрираните заби во артифициелна плунка.

Методи: Вкупно 72 заба (36 млечни и 36 трајни) беа употребени и беа препарирани кавитети од V класа на секој заб. Кавитетите беа реставрирани со и без кондиционирање (освен кај композитот, каде што сите примероци беа кондиционирани). Забите беа складирани во артифициелна плунка во времетраење од 1, 6, 12 и 18 месеци пред изложувањето на артифициелен гел кој предизвикува кариес и набљудувани под SEM.

Резултати: Без присуство на реставрација, забите се подложни на деминерализација. Конвенционалниот глас-јономер цемент врши значајна инхибиција на деминерализацијата. Глас-јономерот модифициран со смола има мал ефект, со исклучок на примероците по 18 месеци, кои покажуваат јасни зони на инхибиција. Компомерот не покажа знаци на инхибиција на деминерализацијата, додека композитот покажа само ограничени знаци на инхибиција.

Заклучоци: Глас-јономерите, конвенционалните и модифицираните со смола се поефикасни во заштитата на забите од понатамошен развој на кариесот, за разлика од компомерите или флуорослободувачките композити, иако најуспешна заштита даваат конвенционалните глас-јономер цементи. Природата на забот немаше влијание на исходот.

Клучни зборови: дентални реставраторски материјали, артифициелен кариес, електронска микроскопија, ослободување на флуориди.

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