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Can Dental Restoratives be Re-Charged with Fluorides?.

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ABSTRACT

The ability to take-up and re-release ions in the surrounding medium makes the glass-ionomers as "rechargeable reservoirs" and they therefore offer a possibility to regenerate the fluoride release by repetitive application of topical fluoride agent or fluoride dentifrice. The purpose of the study was to explore the possibility and the extent of the probable re-charge of glass-ionomers, compared to compomers and composite resins. A total number of 90 teeth, 45 deciduous and 45 permanent were used in this investigation. Class V cavities were prepared on each tooth and restored with glass-ionomers, compomers and composite resins. The teeth were stored in artificial saliva for 1 month. Four cycles of refluoridation were performed: after 1, 2, 3 and 4 weeks. The concentration of the fluoride was determined by spectrophotometry and the aluminium level was determined by atomic absorption spectrophotometry. The conventional and resin-modified glass ionomer cement restoratives are able to regenerate the fluoride release and therefore enhance the long-term anticariogenic ability by displaying a considerable level of fluoride recharging potential. This was not the case with the polyacid-modified composite resin and the fluoride releasing composite resin, since they exhibited a low level of fluoride uptake and re-release. The fluoride re-charge results in subsequent increase of the aluminium release, probably as a result of preservation of the electro-neutrality.

Keywords: fluoride, aluminium, glass-ionomer cement, compomer, composite resin

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INTRODUCTION

It has been commonly suggested that the fluoride release is one of the most favorable properties of the dental restorative materials. In fact, the selection of the restorative material to be used for dental restoration can be influential for the appearance of demineralization or remineralization around the restoration. In vitro studies about the effects of fluorides on the dentin reveal that small quantities of fluoride can lead to hypermineralization of dentin [1, 2]. Incipient carious-like lesions close to glass ionomer cement restorations are found to remineralize or even hypermineralize [3,4,5,6], while amalgam or composite restorations are predominantly associated with further demineralization of the samples [4].

When the glass ionomer cement is set, the released fluoride may originate from: the unreacted glass particles; the siliceous gel phase (which is a result of the acid-base reaction and covers the glass particles); the polycarboxylate matrix where the fluoride ions are strongly complexed with the metal ions; and, finally, the liquid in the pores where the fluoride ions are loosely bound and can move easily [7]. The fluoride release results from two different processes, which appear simultaneously. The short-term release is associated with the leaching of the loosely bound fluoride from the cement matrix [7, 8]. Long-term release is a result of diffusion controlled phenomena, where the concentration gradient is the moving force for the release. The concentration gradient results from the balance between the leaching of the glass particles in the cement and the diffusion of the leached fluoride through the cement matrix [9]. Numerous studies were performed in order to determine the concentration of fluoride released from glass ionomer cements [10-14], nevertheless, the precise mechanism of fluoride release is not yet fully elucidated.

Apart from fluoride, the glass ionomer cements release several different matrix forming cations [13, 15] in the ambient solution, namely monovalent (sodium and fluoride), divalent (calcium, strontium, barium and zinc), trivalent (aluminium) or complex forming ions (silica and phosphorus) [16-18].

When three- or tetra- valent metal cations as aluminium, iron, zirconium etc. are near to fluoride ions, they form stable complexes. Aluminium fluoride is one of the stable complexes usually created with the fluoride anions. Several different forms of aluminium fluoride can be detected in different Al/F ratio- in fact, one aluminium ion in the complex structure can be surrounded by maximum of six octahedrally placed fluoride ions [19]. The aluminium release reaches a peak during the first day after setting [14]. With the maturation of the glass ionomer cement, the aluminium release is decreased, because the aluminium ions from the surface are washed out and the rest of the ions remain entrapped (embedded) deep into the cement matrix [14].

Additionally, the ability to take-up and re-release ions in the surrounding makes them “rechargeable reservoirs” [20] and they therefore offer a possibility to regenerate the fluoride release by repetitive application of topical fluoride agent or fluoride dentifrice [21]. It has, also, been implied by Xu et al. [21] that the materials with higher fluoride content release and uptake higher amounts of fluoride, but that they are associated with lower mechanical strength. In this manner, Forsten [22] states that the polyacid modified composite resins and fluoride containing composites did not show recharging potential.

The progression of the carious lesion is influenced by factors as the chemical adhesion of the enamel and dentin of the cavity walls [23], as well as the morphological and structural differences between the deciduous and permanent teeth, and even by the fact that the thickness of dentine of the deciduous teeth is lower than in the permanent ones [24]. However, the young permanent teeth present a separate category, with completely different characteristics. Their most intriguing feature is the incomplete enamel mineralization. Precisely, when the permanent teeth appear in the mouth, they have to face a continuation of the mineralization process, which completes a few years after their eruption. During this period of post-eruptive maturation, about 10% of minerals are being incorporated in the subsurface layers of the enamel, mostly fluorides [25].

Having in mind the properties of the fluoride releasing dental restoratives and the specifics of the deciduous and the young permanent teeth, the purpose of this study was to compare the fluoride and aluminium ion release and uptake (and additionally) their re-release from several different types of fluoride releasing dental restorative materials, and also, to compare this parameters in deciduous and young permanent immature teeth.

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MATERIAL AND METHODS

A total number of 90 teeth, 45 deciduous and 45 permanent were used in this investigation. Indication for extraction was the exfoliation of the deciduous teeth and orthodontic reasons for the young permanent teeth. After the extraction, the surface of the teeth was 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 were discarded. Class V cavities were prepared on every tooth using diamond bur and turbine with water cooling. After the preparation, the teeth were divided into five groups at random, and filled with one of five different materials, given in Table 1. Each of the groups, consisting of 10 deciduous and 10 young permanent teeth, was divided in two subgroups; the first was conditioned, and the other one left unconditioned. In the group with the composites (5 deciduous and 5 young permanent immature teeth), all of the specimens were conditioned. The conditioning and the filling was performed according to the manufacturers' instruction, as listed in Table 1.

The teeth were stored into artificial saliva [26] used for dental materials testing according to the British Standards Institution, BS 7115, part 2, BSI, London, 1988. The composition of the artificial saliva is given in Table 2.

Table 1: Materials used

Material	Type	Manufacturer	Conditioning option
Fuji IX	Conventional GIC	GC, Japan	Cavity conditioner (GC, Japan)
Fuji II LC	Resin-modified GIC	GC, Japan	Cavity conditioner (GC, Japan) Aldrych (polyacrylic) acid
Experimental GIC	Conventional GIC	Experimental material	H_3PO_4(37%) then Prime& Bond NT (Dentsply)
Dyract AP	Polyacid-modified composite resin	Dentsply De Trey, Germany	Unifil Bond (GC, Japan)
Unifil Flow	Fluoride-releasing composite resin	GC, Japan	

Table 2: Components of the artificial saliva

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

The teeth were stored in artificial saliva for 1 month. The samples were washed in deionized water, dried with a laboratory tissue and stored in 0, 2% NaF for 5 minutes. Then, the samples were washed again and stored in artificial saliva for 1 week. Four cycles of refluoridation were performed: after 1, 2, 3 and 4 weeks. After each measurement, the artificial saliva from each sample was replenished and the level of fluoride and aluminium ions determined.

The concentration of the fluoride was determined by spectrophotometry and the aluminium level was determined by atomic absorption spectrophotometry.

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Aluminium was determined directly by flame atomic absorption spectrophotometry (Varian Atomic Absorption Spectrophotometer 10BQ) with addition of substances for decreasing the ionization and the procedure involved initially preparing a calibration curve. For this, a series of standards in the range of 0.00-5.00 mg/l were prepared by dissolution of the standard compound in water. Subsequently, 1.0 ml NaCl in 10ml standard solution was added, the absorbance measured and calibration curve constructed. Determination was then carried out as follows: In 100ml from the sample, 0.5 ml HNO_3 and 5.0ml HCl were added, heated to reduce the volume to 10ml without boiling. After cooling 1.0ml NaCl was added and the absorbance and concentrations measured. The amount of aluminium was then calculated according to the equation mg/lAl= 100a, where a = concentration of aluminium in 10ml on the calibration curve.

Fluoride in artificial saliva was determined by spectrophotometry (Cintra 6) after isolation of the fluorides with distillation. These techniques are based on observation of colour change by chemical reaction between fluoride ion and an indicator (in this case SPADNS). The procedure required initial creation of a calibration curve. This used a series of standards with concentrations from 0.00 to 1.40 mg/l prepared with a dilution of 50ml using standard fluoride solution (1ml=0.01mg F^-). Determination involved distillation, as follows: 400ml water was put in a flask for distillation and 200ml of concentrated sulphuric acid was added, this solution was boiled to 180˚ C and afterwards cooled to 100 ˚C. The specimen was diluted in distilled water and 300ml of this solution was added to the distillate. The solution was boiled to 180˚C again. Finally, 50 ml of the distillate were put into Nessler pipe and mixed with 10 ml of SPADNS (with addition of acidic cyrconil), after which the absorbance was determined. If the concentration of aluminium was above 3mg/l, then the reading of the results was delayed for 3 hours, as has been suggested previously [27]. This is because of possible interaction of fluoride with aluminum in solution, an effect that diminishes with time, and allows true fluoride levels to be determined. The fluoride level was calculated by substituting into the following equation:

$$\text{Fluoride mg/l} = 50A/V,$$

Where,

A = amount of fluoride (mg) measured by spectrophotometry,
V = volume of the specimen (ml).

RESULTS

The results for the aluminium and fluoride release after four cycles of refluoridation are presented in the Tables 3-7.

Table 3: Aluminium and fluoride release from Fuji IX restorations after refluoridation

Fuji IX		1st week	2nd week	3rd week	4th week
		MEAN(SD)	MEAN(SD)	MEAN(SD)	MEAN(SD)
Al (mg l^{-1})	Deciduous teeth ***ab**	28,06 **c** (1,59)	10,46 **c** (0,68)	28,94 **c** (3,69)	9,45 (0,38)
	Conditioned deciduous teeth **ab**	24,36 (3,89)	10,63 (0,62)	27,52 (4,50)	10,01 **c** (0,82)
	Young permanent teeth ***b**	12,06 (0,61)	10,00 (0,13)	10,14 (1,22)	9,11 (0,52)
	Conditioned young permanent teeth **b**	10,74 (1,49)	11,40 (0,50)	10,04 (0,80)	12,94 (0,56)
F (mg l^{-1})	Deciduous teeth *****	17,58 **c** (1,66)	10,92 **c** (0,20)	22,40 **c** (3,60)	7,35 (3,60)
	Conditioned deciduous teeth **b**	27,36 **c** (3,73)	10,10 (0,78)	23,82 (0,88)	9,56 (0,88)
	Young permanent teeth ***a**	11,72 (1,46)	10,06 **c** (0,12)	10,26 (0,47)	10,58 (0,47)
	Conditioned young permanent teeth ***ab**	10,40 (0,81)	12,51 **c** (0,81)	10,02 **c** (1,00)	10,27 (1,00)

*****statistically significant values for each group after the four measurements (ANOVA p<0,05 i Post hoc Tukey HSD test)

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a- Statistically significant differernce between the conditioned and unconditioned samples (t-test for independent samples p<0,05)
b- statistically significant difference between deciduous/ young permanent teeth and conditioned deciduous./ conditioned young permanent (t-test for independent samples p<0,05)
c- statistically significant difference between the results after 1 month of storage in artificial saliva and after refluoridation (t-test for independent samples p<0,05)

Table 4: Aluminium and fluoride release from Fuji II LC restorations after refluoridation

Fuji II LC		1st week	2nd week	3rd week	4th week
		MEAN(SD)	MEAN(SD)	MEAN(SD)	MEAN(SD)
Al (mg l^{-1})	Deciduous teeth***ab**	28,88 **c** (4,34)	10,65 (0,45)	18,58 (1,59)	9,18 **c** (0,42)
	Conditioned deciduous teeth **ab**	19,10 (0,89)	9,80**c** (0,25)	17,46 (1,31)	9,13 **c** (0,34)
	Young permanent teeth ***b**	12,26 (1,12)	9,46 **c** (0,28)	12,74 **c** (0,81)	10,14 (0,43)
	Conditioned young permanent teeth **b**	13,04 **c** (0,88)	12,01 (0,06)	9,94 (0,93)	11,48 (0,44)
F (mg l^{-1})	Deciduous teeth *	19,88 (2,07)	10,12 (1,64)	26,74 (1,64)	9,97 (0,13)
	Conditioned deciduous teeth **b**	21,28 (3,86)	9,51 (1,37)	17,66 (1,37)	9,52 (0,26)
	Young permanent teeth ***a**	13,92 **c** (2,01)	9,62**c** (2,11)	11,26 **c** (2,11)	9,64**c** (0,26)
	Conditioned young permanent teeth ***ab**	12,22**c** (0,61)	10,90 (2,52)	11,84 (2,52)	11,12 (0,21)

*statistically significant values for each group after the four measurements (ANOVA p<0,05 i Post hoc Tukey HSD test)
a- Statistically significant differernce between the conditioned and unconditioned samples (t-test for independent samples p<0,05)
b- statistically significant difference between deciduous/ young permanent teeth and conditioned deciduous./ conditioned young permanent (t-test for independent samples p<0,05)
c- statistically significant difference between the results after 1 month of storage in artificial saliva and after refluoridation (t-test for independent samples p<0,05)

Table 5: Aluminium and fluoride release from EGIC restorations after refluoridation

Experimental glass ionomer cement		1st week	2nd week	3rd week	4th week
		MEAN(SD)	MEAN(SD)	MEAN(SD)	MEAN(SD)
Al (mg l^{-1})	Deciduous teeth***ab**	18,50 (2,24)	8,61 (0,28)	16,00 (2,75)	7,07 (0,42)
	Conditioned deciduous teeth **ab**	24,58 (3,09)	12,08 **c** (0,39)	27,66 (1,76)	10,90 **c** (0,49)
	Young permanent teeth ***b**	18,58 (1,60)	11,61 (1,20)	18,04 (0,60)	10,60 (0,96)
	Conditioned young permanent teeth **b**	27,34 (3,32)	14,39 (0,77)	27,08 (1,49)	14,73 **c** (0,86)
F (mg l^{-1})	Deciduous teeth *	17,34 (1,41)	8,73 **c** (0,90)	18,94 (1,12)	7,93 (0,27)
	Conditioned deciduous teeth **b**	24,62 **c** (1,23)	11,84 **c** (0,63)	27,76 **c** (1,29)	11,67 **c** (0,54)
	Young permanent teeth ***a**	19,74 (0,75)	12,50 (0,80)	20,02 **c** (1,14)	10,84 (0,33)
	Conditioned young permanent teeth ***ab**	24,82 (1,03)	14,03 (0,36)	28,40 (1,32)	12,00 (1,44)

*statistically significant values for each group after the four measurements (ANOVA p<0,05 i Post hoc Tukey HSD test)
a- Statistically significant differernce between the conditioned and unconditioned samples (t-test for independent samples p<0,05)
b- statistically significant difference between deciduous/ young permanent teeth and conditioned deciduous./ conditioned young permanent (t-test for independent samples p<0,05)
c- statistically significant difference between the results after 1 month of storage in artificial saliva and after refluoridation (t-test for independent samples p<0,05)

Table 6: Aluminium and fluoride release from Dyract AP restorations after refluoridation

Dyract AP		1st week	2nd week	3rd week	4th week
		MEAN(SD)	MEAN(SD)	MEAN(SD)	MEAN(SD)
Al (mg l^{-1})	Deciduous teeth***ab**	14,28 (1,82)	10,87 (0,31)	17,14 (1,83)	10,67 (0,48)
	Conditioned deciduous teeth **ab**	12,92 **c** (0,77)	10,00 (0,17)	14,24 **c** (2,17)	11,57 **c** (1,14)
	Young permanent teeth ***b**	15,52 (0,99)	11,74 **c** (0,33)	14,82 (1,99)	12,04 (0,07)
	Conditioned young permanent teeth **b**	13,02 **c** (0,90)	12,56 (0,28)	10,70 (0,45)	13,76 (0,44)
F (mg l^{-1})	Deciduous teeth *	14,48 (2,06)	10,53 (0,29)	17,14 (0,92)	10,94 (1,10)
	Conditioned deciduous teeth **b**	14,58 **c** (0,75)	10,90 **c** (0,62)	14,24 (2,28)	10,61 (0,37)
	Young permanent teeth ***a**	17,06 c (0,63)	12,82 (0,30)	14,82 (2,53)	12,13 (0,69)
	Conditioned ***ab** Young permanent teeth	12,86 **c** (0,65)	13,64 **c** (0,38)	10,70 (0,82)	13,20 (0,56)

*statistically significant values for each group after the four measurements (ANOVA p<0,05 i Post hoc Tukey HSD test)

a- Statistically significant differernce between the conditioned and unconditioned samples (t-test for independent samples p<0,05)

b- statistically significant difference between deciduous/ young permanent teeth and conditioned deciduous./ conditioned young permanent (t-test for independent samples p<0,05)

c- statistically significant difference between the results after 1 month of storage in artificial saliva and after refluoridation (t-test for independent samples p<0,05)

Table 7: Aluminium and fluoride release from Unifil Flow restorations after refluoridation

Unifil Flow		1st week	2nd week	3rd week	4th week
		MEAN (SD)	MEAN (SD)	MEAN (SD)	MEAN (SD)
Al (mg l^{-1})	Deciduous teeth	12,12 (1,49)	4,60 (0,29)	9,30 (1,34)	5,19 **c** (0,80)
	Young permanent teeth*	10,28 (1,10)	10,74 (0,40)	10,24 (1,75)	9,99 (0,43)
F (mg l^{-1})	Deciduous teeth***b**	11,78 **c** (4,46)	5,95 (0,97)	8,02 (1,41)	5,68 **c** (0,28)
	Young permanent teeth **b**	11,52 (0,88)	10,83 (0,35)	8,84 (1,11)	10,90 (0,92)

*statistically significant values for each group after the four measurements (ANOVA p<0,05 i Post hoc Tukey HSD test)

a- Statistically significant differernce between the conditioned and unconditioned samples (t-test for independent samples p<0,05)

b- statistically significant difference between deciduous/ young permanent teeth and conditioned deciduous./ conditioned young permanent (t-test for independent samples p<0,05)

c- statistically significant difference between the results after 1 month of storage in artificial saliva and after refluoridation (t-test for independent samples p<0,05)

After the refluoridation, the samples restored with Fuji IX showed increase in the fluoride level in the artificial saliva after 1 and 3 weeks, while after 2nd and 4th weeks there was a declination. Statistically significant differences (p<0,05) appeared between the four measurements in the deciduous and young permanent immature teeth and with the conditioned young permanent immature teeth with fluoride.

The values for the fluoride and aluminium release from Fuji II LC restorations after refluoridation between the deciduous and young permanent teeth when aluminium was determined and between all the teeth when fluoride was determined showed statistically significant differences. As with Fuji IX, there is an elevation in the levels of aluminium and fluoride after the 1st and the 3rd week, and a slight decline after the 2nd and 4th week.

The experimental glass ionomer cement demonstrated the same trend of increasing the level of both tested ions (aluminium and fluoride) in the 1st and the 3rd week as the previous materials.

The quantity of aluminium and fluoride released from Dyract AP after refluoridation shows the same trend as the previous materials, but it is not so obvious, having in mind the fact that the fluoride and aluminium increase after the 1st and the 3rd week was lower compared to the one by the glass ionomer cements. Statistically significant differences appear between the conditioned and unconditioned samples and between the deciduous and young permanent immature teeth (for aluminium release).

The samples restored with Unifil Flow show statistically significant differences between the young permanent immature teeth when determining aluminium and between the deciduous teeth when determing fluoride. Additionally, the levels of both aluminium and fluorides are lower compared to the previous materials.

DISCUSSION

The ability of the materials to recharge (uptake) and re-release fluorides after professional application or usage of fluoride dentifrices is especially important for long-term, protracted preservation of therapeutic concentrations of fluorides in the solution around the interface between the tooth enamel and the material.

The release of fluorides from refluoridated materials is lower than in the freshly mixed ones. Additionally, the level of fluorides released after exposure to fluoride solutions depends on the concentration of the solution. This means that exposure of the samples to solutions with different concentrations can not completely restore the initial fluoride release.

It is highly important that the materials can be recharged during a long-time interval and that significant and constant presence of fluorides can be sustained in their environment, because it is a mode to raise the fluoride level in the saliva and around the neighboring hard dental substances [3, 4, 20]. Even small quantities released from the restoration can act in the protection against the dental decay, because the caries-preventive effect is highest when small quantities of free fluoride ions are persistently present in the oral environment.

According to Creanor et al., repeated incubations with concentrated fluoride solutions resulted in higher stable fluoride release [28]. A study performed by Suljak et al. [29] proved that the glass ionomer cement restorations are capable of recharging fluorides from the environment, which becomes obvious after a short period of increasing of the fluoride level of the examined solution. But, in vivo, the fluoride recharge is limited by a thin film originating from the saliva, which is formed on the surface of the cement [30].

There are attempts to describe the mechanism of fluoride recharge, but it is still speculative. According to Damen et al. [31], a part of the water present in the glass ionomer cements can evaporate, and it is marked as “loosely bound”. The other part, so called “tightly bound” water is complexed with the cations and the matrix. The loosely bound water and the liquid in the porosities of the cement diffuse passively in the environment. The fast recharge and release of fluoride are based on the mechanism of diffusion in and out of the porosities in the cement filled with liquid. Because of the fast release, a higher basic concentration around the glass ionomer cements can be expected only after frequent exposure to high doses of fluorides.

According to Forsten [22], the fluoride treatment did not affect the compomers and the fluoride-releasing composite resins, which was proved in our study. Our results point to the fact that the influence of the repeated exposure to sodium fluoride solution did not lead to significant escalation of the level of fluoride in the artificial saliva in the compomer and fluoride-releasing composite resin’ samples. But, Attar et al. [32] found that the compomers can be recharged, although they have significantly lower refluoridation potential than the glass ionomer cements. It is also obvious that the glass ionomer cements have a certain refluoridation potential. Namely, our results prove that after 1 week significant levels of fluorides were re-released, and the level was the same, and even higher than the basic fluoride release after 1 month. This refers that most of the fluoride gained from the exposure of the teeth samples to sodium fluoride solution is probably incorporated in the porosities of the cement (and, probably into the tooth enamel), so later, even in the 3rd week there is a decrease in the release (namely, increase of the fluoride level in the artificial saliva). The significant differences which appeared between the deciduous and the young immature permanent teeth are evidence of the incorporation of the fluorides into the tooth enamel.

The differences that appeared between the conditioned and unconditioned samples could be a result of the reduction of the spaces reachable for adsorption of the fluoride ions after the partial demineralization.

The fluoride recharge, definitely, results in increased aluminium release. Preservation of the electro-neutrality seems to be the probable mechanism. Namely, the fluoride as electronegative element, extracts the aluminium (as a cation) from the cement.

CONCLUSIONS

The conventional and resin-modified glass ionomer cement restoratives are able to regenerate the fluoride release and therefore enhance the long-term anticariogenic ability by displaying a considerable level of fluoride recharging potential. This was not the case with the polyacid-modified composite resin and the fluoride releasing composite resin, since they exhibited a low level of fluoride uptake and re-release. There were insignificant changes in the initial fluoride release after four cycles of refluoridation. The teeth conditioning prior to the glass ionomer cement restoration' placement results in elevated fluoride re-release (or, more precisely, diminished uptake), which might be a result of decreasing the number of places reachable for adsorption. The fluoride re-charge results in subsequent increase of the aluminium release, probably as a result of preservation of the electro-neutrality.

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