THE EFFECT OF LOW-CONCENTRATION FLUORIDE SOLUTIONS ON FLUORIDE RECHARGE ABILITY OF CONTEMPORARY DENTAL RESTORATIVES AND ADHESIVES

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SUMMARY: The aim of this *in vitro* study was to evaluate the fluoride release ability, during a 30-day period and the fluoride recharge ability, after 0.02% and 0.04% NaF solution treatments, of five fluoride-releasing restorative materials (Fuji IX GP, Ketac N100, Dyract Extra, Beautifil II, and Wave) and three fluoride-releasing dental adhesives (Stae, Prime & Bond NT, and FL-Bond II). Eight disk-shaped specimens for each material were prepared. Fluoride release was measured using a fluoride ion-selective electrode daily for 30 days. For refluoridation procedures, the specimens were immersed in 0.02% NaF for 5 min. Fluoride measurements were carried out at 24 hr intervals for 5 days. This refluoridation regime was repeated twice, and measurements were again recorded. After 15 days, the same procedure was carried out using 0.04% NaF. During the 30-day period, Fuji IX GP released the highest amount of fluoride among the restorative materials while Prime & Bond NT was the highest among the dental adhesives. After NaF solution treatments, Fuji IX GP again ranked the highest in fluoride release among the restorative materials while FL-Bond II ranked the highest among dental adhesives. It was concluded that fluoride recharge abilities of fluoride-releasing dental materials depend on their compositions and setting mechanisms and that low-concentration fluoride solutions which are indicated for children affect fluoride re-release.

Keywords: Fluoride recharge ability; Fluoride-releasing adhesives; Fluoride-releasing restoratives; Low-concentration fluoride solutions.

INTRODUCTION

Initial carious lesions in restoration margins must be exposed to a crucial concentration of fluoride ions for a prolonged period of time to achieve the cariostatic effect.[1] Fluoride-releasing restorative materials, including conventional glass ionomer cements (GIC), resin-modified glass ionomer cements (RMGIC), polyacid-modified composite resins (compomers), fluoridated composite resins and composite resins containing pre-reacted glass ionomer fillers (giomers), have been introduced in order to release sustained amounts of fluoride ions to exhibit cariostatic action.[2] However, only restorative materials that release high amounts of fluoride ions have been shown to effectively inhibit the demineralization of tooth structure adjacent to restorative margins.[3]

Fluoride-releasing dental adhesive systems have been developed and are expected to inhibit secondary caries by both promoting adhesion to dental tissues and releasing fluoride ions. It has been recognized that dental adhesives play a vital role in sealing the margins of composite restorations. Fluoride-releasing

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dental adhesives seem to be especially attractive, because they are not only in close contact with the dentin margins of restorations, but may also partially diffuse into them.[4]

Due to the fact that fluoride levels leached from fluoride-containing materials decrease over time, the "recharging" of these materials with fluoride has been suggested to maintain a continuously increased level of fluoride release.[5] Fluoride mouthrinses have been used extensively in the past to prevent dental caries in children. Supervised, school-based, weekly rinsing programmes using 900 ppm F solutions remain a popular procedure in USA.[6] Mouthrinses containing 230 ppm F are available commercially for daily home use in some countries. Rinses containing 100 ppm F are also available and recommended for twice daily use.[7]

Although the procedure is not recommended for children under 6 years of age, due to the risk of acute and chronic fluoride ingestion, there are data implicating fluoride mouthrinse use by pre-school children as a risk factor for dental fluorosis because some young children might swallow substantial amounts.[7] For this reason, the use of low-concentration fluoride solutions in children has been recommended. Chow et al.[8] demonstrated that the effectiveness of a fluoride regimen depends less on the fluoride dose and more on the ability of the treatment to utilize fluoride efficiently for remineralization of tooth tissues.

The aim of this *in vitro* study was to evaluate the fluoride release ability, during a 30-day period and the fluoride recharge ability, after 0.02% and 0.04% NaF solution treatments, of five fluoride-releasing restoratives and three fluoride-releasing adhesives. The first null hypothesis of this study was that the restoratives and the adhesives investigated release similar amounts of fluoride during the experimental period. The second null hypothesis was that there is no difference in fluoride re-release after NaF solution treatments among the materials tested.

MATERIALS AND METHODS

Two GIC and three fluoride-releasing resin-based restoratives were investigated in the present study and a non-fluoride-containing composite resin was used as a control (Table 1). Additionally, three fluoride-releasing adhesives were also investigated, and a non-fluoride-containing adhesive was used as a control (Table 2).

Table 1. The tested restorative materials

Restorative material	Type	Manufacturer	Composition
Fuji IX GP Capsule	Highly viscous conventional GIC	GC Corporation, Tokyo, Japan	Dust: Fluoro-alumino-silicate glass 70-80% Liquid: Polyacrylic acid 10-15% distilled water 10-15%
Ketac N100	Resin-modified GIC containing nano-fillers (nano-ionomer)	3M ESPE, Dental Products, St Paul, MN, USA	Paste A: Fluoro-alumino-silicate glass (FAS) 40-50%, silane-treated ZrO_2 silica 20-30%, silane-treated silica 5-15%, PEGDMA 5-15%, HEMA 1-10%, Bis-GMA <5%, TEGDMA <5% Paste B: Silane-treated ceramic 20-30%, silane-treated silica 20-30%, water 10-20%, HEMA 1-10%, acrylic / itaconic acid copolymer 20-30% Fillers: 69% w/w (42% nano-fillers, 27% FAS)
Dyract Extra	Polyacid-modified composite resin (compomer)	Dentsply DeTrey, GmbH, Konstanz, Germany	Glass: Strontium-alumino-sodium-fluoro-phosphor-silicate glass Monomers: Bis-GMA, UDMA, TEGDMA, TMPTMA, TCB Fillers: SrF_2, SiO_2 47% w/v, 73% w/w
Beautifil II	Composite resin containing S-PRG fillers (giomer)	Shofu Inc., Kyoto, Japan	Monomers: Bis-GMA 7.5%, TEGDMA <5% Fillers: S-PRG 68.6% w/v, 83.3% w/w
Wave	Flowable fluoride-containing composite resin	SDI Limited Bayswater, Victoria, Australia	Monomers: Multifunctional methacrylic ester, UDMA Fillers: SrF_2, silica fillers 40% w/v, 65% w/w
Filtek Z250	Non-fluoridated microhybrid composite resin (control)	3M ESPE, Dental Products, St. Paul, MN, USA	Monomers: Bis-GMA, Bis-EMA, UDMA Fillers: Zirconia/silica 60% w/v, 82% w/w

Table 2. The tested dental adhesive systems

Dental adhesive	Type	Manufacturer	Composition
Stae	Fluoride-releasing etch-and-rinse (2 steps) adhesive system	SDI Limited, Bayswater, Victoria, Australia	Proprietary hydrophilic / hydrophobic monomer HEMA, acetone/water solvent
Prime & Bond NT	Fluoride-releasing etch-and-rinse (2 steps) adhesive system	Dentsply DeTrey, GmbH, Konstanz, Germany	Nano-fillers SiO_2 Di-, tri-methacrylate resins, Bis-GMA, PENTA, TEGDMA, acetone solvent, cetylamine hydrofluoride
FL-Bond II	Giomer self-etch (2 steps) adhesive system	Shofu Inc, Kyoto, Japan	Primer: Carboxylic acid monomer, phosphoric acid monomer, water/ethanol solvent Bonding agent: S-PRG fillers, UDMA, 2-HEMA, TEGDMA
Adper Scotchbond 1 XT	Non-fluoridated etch-and-rinse (2 steps) adhesive system (control)	3M ESPE, Dental Products, St. Paul, MN, USA	Silica nano-fillers 10% Bis-GMA, HEMA, dimethacrylates water/ethanol solvent methacrylate copolymer of polyacrylic and polyitaconic acids

Eight disk-shaped specimens (7 mm in diameter with a thickness of 2 mm) for each material were prepared using cylindrical Teflon molds. Polyester strips were placed on both sides of the mold; glass plates were placed over the polyester strips and clamped to produce a smooth surface. The specimens of the resin-based materials were light-cured for 20 sec from both sides of the mold with a QTH light-curing unit (Elipar 2500, 3M ESPE, Dental Products, Seefeld, Germany) at 1300 mW/cm^2. The glass powder mixture of the GIC specimens was prepared in accordance with manufacturer’s instructions. Immediately after mixing, cement was inserted inside the mold and was covered by a polyester strip. Fuji IX GP specimens were allowed to set in the mold a total of 7 min, while Ketac N100 samples were light-cured for 20 sec from both sides of the mold. Excess material that extruded around the edge of the mold was carefully removed by using a surgical blade. The dental adhesives were dropped into the molds, covered with a polyester strip and a glass plate, and light-cured for 20 sec. The total surface of each specimen was 120.89 mm^2. The specimens were inspected under a magnifier from an optical microscope to ensure that the surface of the prepared specimens were free from air bubbles and cracks.

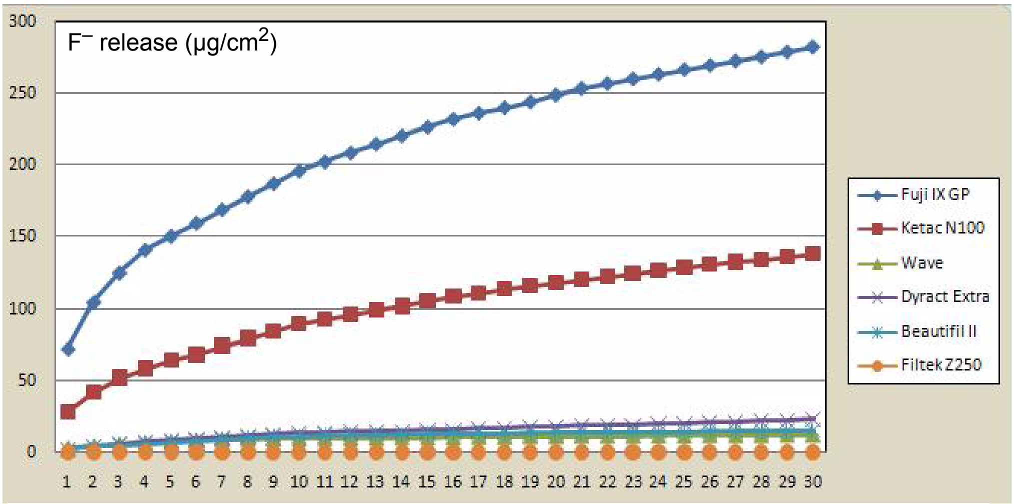
Each prepared specimen was suspended with non-fluoride dental floss in 4 mL deionized water in a plastic container, and incubated at a constant temperature of 37±0.5ºC during the whole experimental period. The first measurement of fluoride concentration of each solution was done 24 hr after preparation of the specimens. Each sample was rinsed with 1 mL of deionized water in the plastic container and then the specimen was transferred to a new plastic container with 4 mL deionized water. 0.5 mL of TISAB III (Thermo Fisher Scientific, Beverly, USA) was added at 10 vol% of determined solution. This created a constant background ionic strength for fluoride measurement. The fluoride concentration was then measured using a microanalytical technique with a fluoride ion-selective electrode (Orion 9609BNWP, Ionplus Sure-Flow Fluoride, Thermo Scientific, USA) with detection limit at ±0.001 ppm. Measurements performed at a constant room temperature of 23±1ºC and were converted to $\mu g/cm^2$ for statistical analysis. This regimen of specimen transfer and fluoride analysis of storage media was continued daily for 30 days. After day 30 until day 86 the specimens were transferred daily to new plastic containers with 4 mL deionized water.

Refluoridation of the test specimens was carried out as follows: on day 86 after measurement of fluoride release, the specimens were cleansed by rinsing them three times with 5 mL of deionized water, dried on paper for 2 min, and then immersed in 0.02 wt% NaF (90 ppm F^-) for 5 min. The specimens were then individually rinsed three times with 5 mL deionized water, air dried for 1 min and placed in individual plastic tubes containing 4 mL deionized water at 37ºC. Fluoride measurements were carried out at 24 hr intervals for 5 days. After 5 days the fluoride exposure procedure was repeated, and measurements were again recorded. In total, this refluoridation regime was carried out three times for a total fluoride release measurement period of 15 days—that is, three sets of 5-day measurement periods. After 15 days the same procedure was carried out using 0.04 wt% NaF (180 ppm F^-).

Data were statistically analyzed using one-way ANOVA. Differences in fluoride release amount between experimental groups were evaluated using Bonferroni *post hoc* test, at a level of significance $p<0.05$. Comparison of cumulative fluoride release for each experimental group was done using paired *t*-test and non-parametric Wilcoxon test at a level of significance $p<0.05$.

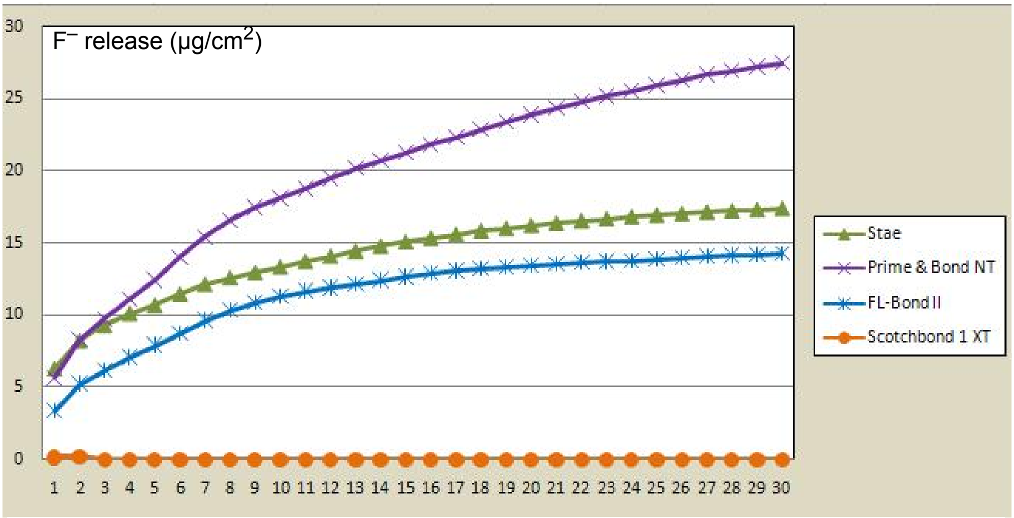
RESULTS

Figures 1a and 1b present graphically the cumulative fluoride release data of restoratives and adhesives during the 30-day experimental period.



Duration of exposure (days)

Figure 1a: Cumulative fluoride ion release (µg/cm^2) from the restorative materials during 30-day period.



Duration of exposure (days)

Figure 1b: Cumulative fluoride ion release (µg/cm^2) from the dental adhesives during 30-day period.

Cumulative fluoride release of restoratives and adhesives at 1st and 2nd 15-day period is presented in Table 3.

Table 3. Mean values and standard deviations (μg/cm^2) of cumulative fluoride release at 1st and 2nd 15-day periods before NaF treatment and at 15-day periods after refluoridations with 0.04% and 0.02% NaF for all dental materials tested*,†

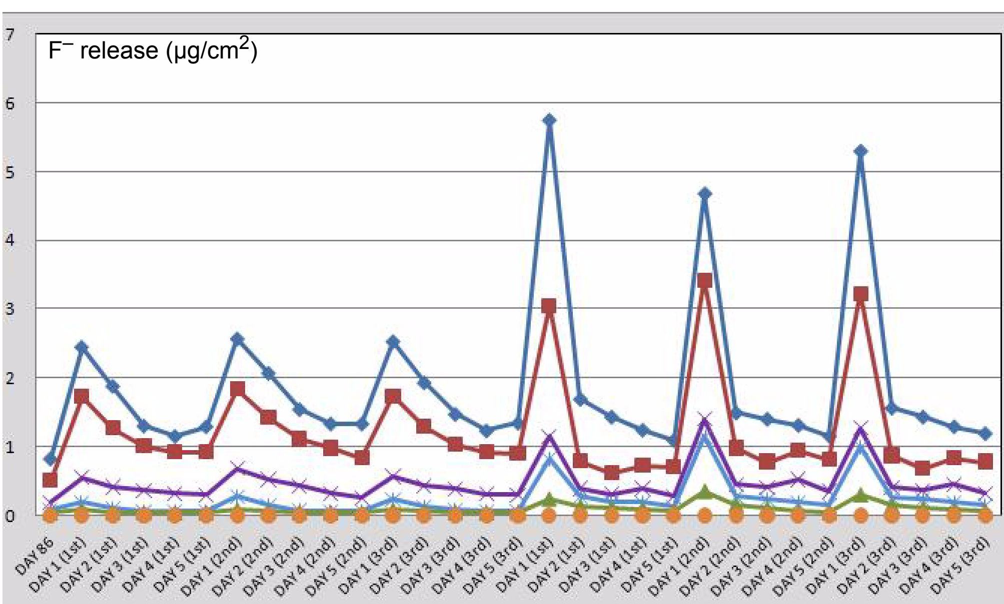
Dental material	Cumulative fluoride release (μg/cm^2)							
	Before NaF treatment			After refluoridation with NaF				
	Days 1–15	Days 15–30		0.04% NaF (Days 1–15)		0.02% NaF (Days 1–15)		
Restoratives								
Fuji IX GP	226.86 (32.40)[a]	55.80 (7.44)[a]	24.5%	32.00 (4.92)[a]	14.1%	25.41 (4.23)[a]	11.2%	
Ketac N100	105.37 (17.56)[b]	32.90 (4.11)[b]	31.2%	19.21 (2.74)[b]	18.2%	17.96 (2.76)[b]	17.0%	
Dyract Extra	15.54 (2.11)[c]	7.86 (0.72)[c]	50.5%	8.55 (0.89)[c]	55.0%	6.23 (0.77)[c]	40.0%	
Beautifil II	13.16 (2.63)[d]	2.40 (0.36)[d]	18.2%	5.38 (0.46)[d]	40.8%	1.62 (0.27)[d]	12.3%	
Wave	10.63 (2.36)[d]	1.75 (0.32)[d]	16.4%	2.05 (0.31)[e]	19.2%	0.79 (0.10)[e]	7.4%	
Filtek Z250	0.46 (0.00)[e]	0.00 (0.00)[e]	0.0%	0.00 (0.00)[f]	0.0%	0.00 (0.00)[f]	0.0%	
Adhesives								
Prime & Bond NT	21.28 (2.83)[a]	6.16 (0.88)[a]	28.9%	2.41 (0.38)[a]	11.3%	1.89 (0.31)[a]	8.8%	
Stae	15.07 (2.15)[b]	2.31 (0.39)[b]	15.3%	2.46 (0.41)[a]	16.3%	1.81 (0.28)[a]	12.0%	
Fluorobond II	12.65 (1.80)[b]	1.64 (0.27)[b]	12.9%	4.24 (0.63)[b]	33.5%	2.78 (0.49)[b]	21.9%	
Scotchbond 1XT	0.19 (0.00)[c]	0.00 (0.00)[c]	0.0%	0.00 (0.00)[c]	0.00%	0.00 (0.00)[c]	0.0%	

*Percentage (%) indicates the fractional amount of cumulative fluoride release when compared to the 1st 15-day period of fluoride release.

†Same superscripts indicate no significant differences ($p>0.05$) among the restorative materials or dental adhesives.

Statistical analysis indicated significant differences in fluoride release among the restoratives tested, as well as among the adhesives ($p<0.05$). For all the materials tested, fluoride release decreased with time. On day 1, all fluoride-containing restoratives and adhesives released their greatest amounts of fluoride as shown in Figure 1a. After day 1, fluoride release rapidly declined but stabilized at days 3–5, remaining relatively constant from then on until day 30. Among the restoratives, Fuji IX GP released the highest amount of fluoride ($p<0.05$). Comparison between GICs and resin-based materials showed that GICs released markedly higher amounts of fluoride than the resin-based restoratives ($p<0.05$). For the adhesives, they maintained a stable fluoride release in very low amounts from day 3 until day 30. Among the resin-based restoratives Dyract Extra exhibited the highest fluoride release during 30-day period ($p<0.05$), while Beautifil II and Wave did not show any significant difference ($p>0.05$). Among the adhesives, Prime & Bond NT released the highest amount of fluoride ions ($p<0.05$).

Table 3 shows the cumulative fluoride release data and Figures 2a and 2b present the fluoride release patterns of restoratives and adhesives, during three sets of 5-day periods after each refluoridation procedure with 0.02% and 0.04% NaF solutions.



Day during three sets of 5-day periods after each refluoridation procedure and before the first refluoridation (day 86)

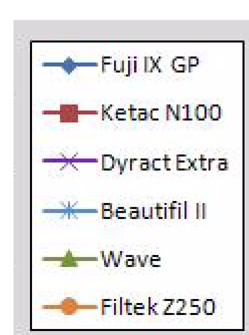
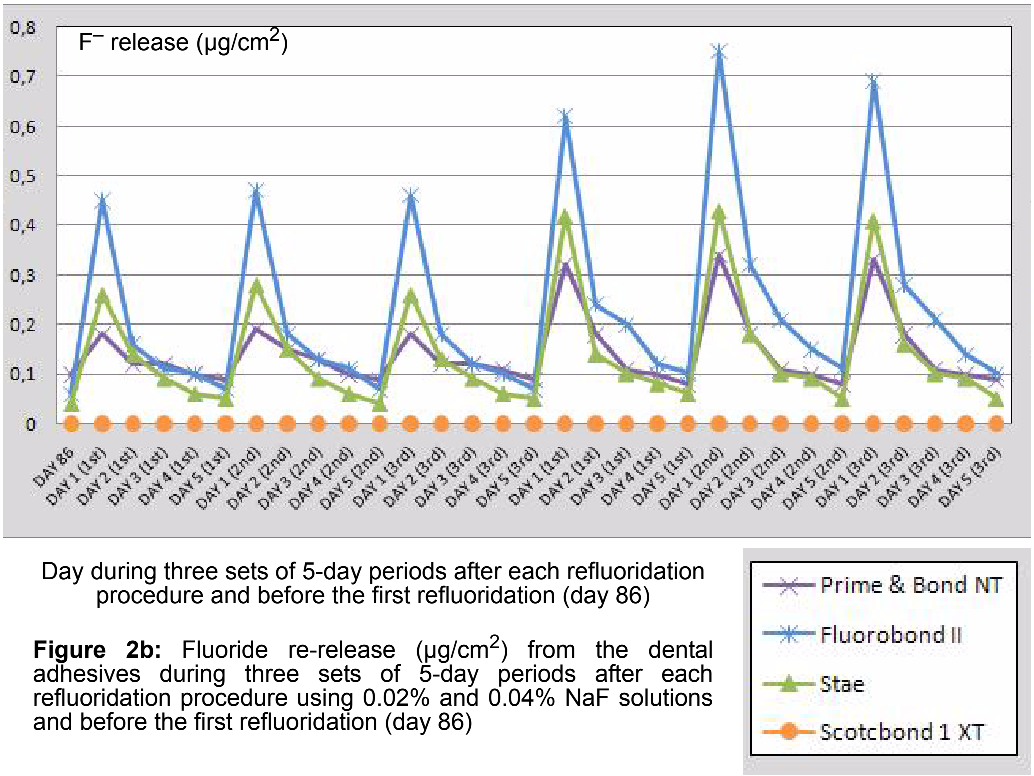


Figure 2a: Fluoride re-release (μg/cm^2) from the restorative materials during three sets of 5-day periods after each refluoridation procedure using 0.02% and 0.04% NaF solutions and before the first refluoridation (day 86)



Day during three sets of 5-day periods after each refluoridation procedure and before the first refluoridation (day 86)

Figure 2b: Fluoride re-release (μg/cm^2) from the dental adhesives during three sets of 5-day periods after each refluoridation procedure using 0.02% and 0.04% NaF solutions and before the first refluoridation (day 86)

For all the tested materials, fluoride re-release decreased with time. Exposure to 0.02% and 0.04% NaF solution for 5 min caused a significant, but variable, increase in fluoride re-release from all fluoride-containing materials ($p<0.05$). At day 1 after refluoridation, there was an increase in fluoride release from all fluoride-containing materials. After day 1, fluoride release rapidly declined. At day 5 after refluoridation, the amounts of fluoride released were almost the same as those before refluoridation (day 86). Among the restoratives, Fuji IX GP released the highest amount of fluoride during each 5-day period after refluoridation ($p<0.05$). Among the adhesives, the ranking was FL-Bond II followed by Stae and Prime & Bond NT ($p<0.05$). Table 3 also shows the comparison of initial cumulative fluoride release at 1st 15-day period against the cumulative fluoride release at 2nd 15-day period and after refluoridations (15-day period) with 0.02% and 0.04% NaF of the materials investigated. All the materials tested exhibited the highest cumulative fluoride release at 1st 15-day period. The specimens after refluoridation with 0.04% NaF re-released higher amounts of fluoride than after refluoridation with 0.02% NaF ($p<0.05$). Among the restoratives; Dyract Extra presented the highest percentage of fluoride re-release, while among the dental adhesives; FL-Bond II presented the highest percentage of fluoride re-release.

DISCUSSION

The fluoride-containing materials evaluated in this study released measurable quantities of fluoride during the 30-day experimental period. Additionally, at the 1st 15-day period the materials released much higher amounts of fluoride ions than the 2nd 15-day period. However, there were large variations in the amount of fluoride release among the materials. This observation is in agreement with the findings of many other authors.[9,10] Consequently, the results obtained from this study demand rejection of the first null hypothesis.

The release of fluoride ions from the materials is a complex process. It can be affected by several intrinsic factors, such as formulation of the organic matrix and fillers,[11] and the amount of inherent or added fluoride,[12] as well as the solubility and porosity of the materials.[13] It is also influenced by external variables such as pH and temperature of the environment, frequency of changing of the storage solution, and plaque and pellicle formation, as well as the type of storage media utilized.[14,15] Additionally, the powder/liquid ratio used in preparing the material, and the method of mixing,[16] curing time,[17] and exposed surface area[18] of the material may affect fluoride release.

Among restoratives, Fuji IX GP, which is a conventional GIC, released the highest amounts of fluoride. This finding has been previously reported.[9,10] Two mechanisms have been proposed by which fluoride may be released from GIC into an aqueous environment. The first mechanism is a short-term reaction, which involves rapid dissolution from outer surface into solution (process I), whereas the second is more gradual and results in a sustained diffusion of fluoride through the bulk cement (process II).[5] RMGICs were mostly found to have a potential for releasing fluoride in equivalent amounts to conventional GICs[19] However, in the current study, Fuji IX GP released a significantly higher amount of fluoride than Ketac N100. This potential may be affected not only by the formation of complex fluoride compounds and their interaction with polyalkenoate acid, but also by the type and amount of resin used for the photochemical polymerization reaction.[20]

In the present study the resin-based materials released much lower amounts of fluoride than the GICs. The results are in agreement with many previous studies.[19,21] This may be because they do not undergo an acid/base reaction, or it may also be a result of their low initial fluoride content.

Fluoride-releasing adhesives released considerable amounts of fluoride throughout the experimental period. The quantitative differences among the materials may be attributed to differences in the inherent fluoride content or in the amount of fluoride added by the manufacturer. The fluoride release of dental adhesives may also be influenced by the solubility and type of the active component, as well as by the phase (organic or inorganic) in which it is added.[22]

After refluoridation procedures, resin-based materials exhibited a significantly lower re-release property than GIC. Consequently, the second null hypothesis of this study is rejected. The precise nature of this mechanism is not fully understood, but it has been suggested that the recharging ability in the GIC is dependent on the glass component of the material and in particular upon the structure of the hydrogel layer around glass filler particles following reactions between the glass and polyacid component.[23] The increased fluoride release after exposure of resin-based materials to NaF solutions is most probably because of pores or surface-retained fluoride.

Among adhesives, FL-Bond II exhibited the greatest fluoride re-release, maybe due to PRG-fillers, which it contains.[24] *In vitro* studies exhibited inconsistent results concerning the capability of fluoride-releasing adhesives to influence formation of secondary caries[25] and there are no clinical studies supporting the effectiveness of these materials. Further investigation is needed to determine the impact of their role on secondary caries formation. Dijkman and Arends[26] found that a fluoride concentration between 5–80 ppm at the interface between restoration and tooth tissues, might be the optimal range to prevent caries formation. In the current study, the cumulative fluoride re-release of the dental adhesive systems after 15-day refluoridation period with 0.02% and 0.04% NaF was between 0.44–0.67 ppm and between 0.58–1.02 ppm, respectively.

The results of this study are in agreement with findings of other *in vitro* studies which suggested that fluoride release, after refluoridation of fluoride-containing restoratives, increased in the first 24 hr followed by a rapid return to near pre-exposure levels within several days.[10] This fluoride re-release is always lower than the initial fluoride release of the materials but is still significant[27].

The results of the current study showed that the fluoride re-release of the materials after refluoridation with 0.04% NaF was slightly higher than that with 0.02% NaF. This finding is in agreement with previous reports.[10,28] Nevertheless, the clinical implication of this difference in fluoride re-release around the dental tissues may not be significant. Jacobson et al.[29] showed that a concentration of fluoride ions around 3 ppm initiates the remineralization in enamel, while in lower concentrations there is no inhibition of demineralization in enamel. In the present study, fluoride re-release from the restoratives ranged from 0.02 ppm to 0.62 ppm after 0.02% NaF treatment and from 0.03 ppm to 1.39 ppm after 0.04% NaF treatment at 1st day of refluoridation procedures.

The clinical implication of fluoride re-release has not been estimated so far, but it may be more significant than the inherent fluoride release of the materials. This ability is very important due to the fact that a continuously increased level of fluoride release around restorations is necessary for the inhibition of secondary caries formation. The recharging ability of a restorative depends on the composition of the material, on the frequency of fluoride exposure, and on the kind and concentration of the fluoridating agent.[23]

CONCLUSIONS

Within the limitations of this *in vitro* study, it can be concluded that low-concentration fluoride solutions may be useful for patients with a high caries risk such as children. Currently, only a few clinical studies have looked into the demineralization behavior of tooth tissues adjacent to fluoride-releasing restorative materials. The results of these studies were contradictory, so the clinical relevance of fluoride-releasing restoratives is still debatable. As a matter of fact, further *in vivo* studies on secondary caries inhibition around restorations with recent fluoride-containing restorative materials and adhesive systems are needed to clarify the relationship between fluoride release, adhesion to tooth structure, and caries inhibition.

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