FLUORIDE RELEASE AND RECHARGE POTENTIAL OF REMINERALIZING ORTHODONTIC ADHESIVE SYSTEMS

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ABSTRACT: The aim of this study was to assess the fluoride (F) release potential of F-containing adhesives Transbond Plus (TB+), Light Bond (LBF), and Geristore-Tenure (GS), and a control adhesive without F content, Transbond XT (TBXT), in relation to microhardness (µH) and degree of conversion (DC). Ten specimens of each adhesive system were illuminated at 1100 mW/cm^2 for 10 and 20 sec, totalling 80 samples. Fourier transform infrared spectroscopy was used for the assessment of DC. µH was assessed before and after four-weeks of immersion in artificial saliva (pH 4.8) at 37°C. The amount of F release was monitored by a F ion-selective electrode. F release was related to adhesive system type and curing time ($p<0.05$) and decreased in the following order, with the subscript indicating the illumination time in sec: $TB+_{10}>TB+_{20}>GS_{10}>GS_{20}>LBF_{10}>LBF_{20}$. The decrease of µH after four weeks of immersion in artificial saliva is related to increased release of F ($r=0.651$; $p<0.001$). TB+ had significantly superior white spot lesions preventive potential due to twice higher F ions release, but its high µH changes raise concerns of bond strength properties. The tested materials showed low F recharge potential.

Keywords: Cariostatic agents; Degree of conversion; Hardness; Orthodontic adhesives.

INTRODUCTION

In an attempt to reduce the incidence of caries in the form of white spot lesions (WSL), great efforts have been made in introducing reservoirs for prolonged intraoral release of fluoride (F), and also in introducing new bioactive remineralization agents.[1-5] A systematic review has shown that strong evidence on the power of F in prevention of WSL during orthodontic treatment and on the best prevention protocols is lacking,[6] and recent research has confirmed that F-containing orthodontic adhesives do not offer WSL-preventive advantages over conventional ones.[7,8]

The research of the mechanical properties of F-releasing orthodontic adhesives has been limited.[9] Microhardness (µH) is an important physical property of adhesives[10,11] that could be an indicator of chemical degradation[12] as well as of bond strength,[13] although some have reported differently.[14] The aim of the present study was to investigate the F release profile of biointeractive orthodontic adhesive systems of various chemical composition and degree of conversion (DC), as well as changes in mechanical properties due to exposure to acidic saliva, in a simulation of intraoral conditions in the presence of acidogenic pH profile of plaque.

MATERIALS AND METHODS

Four types of commercially available adhesive systems of different formulations were tested (Table 1); F-containing adhesives: light-cure Transbond Plus with the Transbond XT primer (TB+) (3M Unitek, USA), Light Bond Paste With Fluoride combined with the Light Bond Sealant With Fluoride primer (LBF) (Reliance

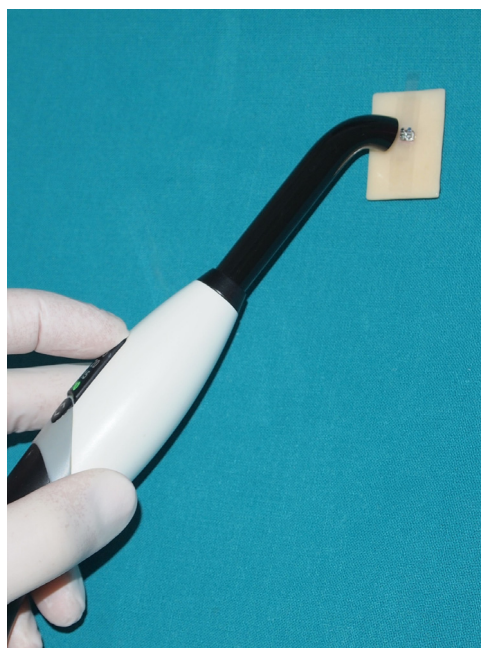[a]Department of Orthodontics, University of Rijeka, Faculty of Medicine, 51000 Rijeka, Croatia; [b]Division of Analytical Chemistry, Department of Chemistry, Faculty of Science, University of Zagreb, 10000 Zagreb, Croatia; [c]Department of Pharmacology, School of Dental Medicine, University of Zagreb, 10000 Zagreb, Croatia; [d]Department of Medical Sciences, University of Trieste, 34129 Trieste, Italy. For correspondence: M Trinajstic Zrinski, Department of Orthodontics, University of Rijeka, Faculty of Medicine, 51000 Rijeka, Croatia; E-mail: magda.zrinski@medri.uniri.hr

Orthodontic Products Inc., USA), and the dual-cure Geristore® adhesive with Tenure® primer (GS) (DenMat, USA), and a control non F-containing adhesive Transbond XT combined with the Transbond XT primer (TBXT) (3M Unitek, USA).

Table 1. Composition of adhesive samples

Orthodontic adhesive system	Monomers	Fluoride
TB XT	bisphenol A glycidyl dimethacrylate (Bis-GMA) (adhesive); Bis-GMA and triethylene glycol dimethacrylate (TEGDMA) (primer)	–
TB +	polyethylene glycol dimethacrylate, citric acid dimethacrylate oligomer, and Bis-GMA (adhesive); Bis-GMA and TEGDMA (primer)	70–90% F-releasing silica filler CAS (Chemical Abstracts Service) 100402-78-6
LBF	TEGDMA (adhesive); Bis-GMA, TEGDMA and (urethane dimethacrylate) UDMA (primer)	<1% NaF
GS	ethoxylated bisphenol A dimethacrylate (EBPADMA) (Geristore® adhesive); EBPADMA and TEGDMA (Tenure® primer)	1–5% F-releasing silica filler CAS (Chemical Abstracts Service) 68611-44-9

A cellulose strip was placed on a metal-ceramic pad imitating the surface of the tooth and a thin layer of primer applied followed by adhesive. This was then covered with another cellulose strip and firmly pressed with a metal premolar bracket. The Bluephase curing unit (Ivoclar Vivadent, Liechtenstein) was used for illuminating the specimens at 1100 mW/cm^2, (10 or 20 sec) (Figure 1). The illumination times chosen were available in the curing unit settings and were in accordance with the manufacturers' recommendations which vary from 6 to 20 sec. In assessing the DC, Fourier transform infrared spectroscopy in conjunction with attenuated total reflectance was used on the EQUINOX 55 interferometer (Bruker Corporation, USA).[15] Each adhesive system sample was immersed in 1 mL of artificial saliva[16] in 1.5 mL tubes. Immersion lasted for 28 days in a thermal

Figure 1. The adhesive was placed between two cellulose strips under a metal bracket and illuminated from the mesial and distal sides.

chamber at 37ºC as a simulation of intraoral condition, and the pH of 4.8 was a simulation of the acidogenic pH profile of one- and two-day-old dental plaque.[17]

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Microhardness was assessed before and after immersion by the Vickers method at Leica VMHT MOT (Walter Uhl, Germany). On the 28th day each specimen was washed with distilled water (dH_2O) and recharged by exposure to 1 mL of 750 ppm neutral NaF solution for 5 minutes. Thereafter, they were washed with dH_2O, immersed in new sealed containers with 1 mL of artificial saliva and stored in an incubator for 24 hours. The amount of F release was measured with a F-ion selective electrode on the Expandable Ion Analyzer EA 940 (Orion Research, USA) after addition of 3.5 mL of re-distilled water and 0.5 mL of the TISAB III solution (Thermo Fisher Scientific, USA).[18] One- and two-way analyses of variances with Student-Newman-Keuls post-hoc test were used to assess the difference in DC, μH, and F release between adhesive types and illumination time. Pearson correlation was applied to explore the relationship between F release and changes in mechanical properties of adhesives. Statistical software IBM SPSS 22 (IBM Corp, Armonk, USA) was used.

RESULTS

Differences between the tested materials are presented in Figures 2 and 3.
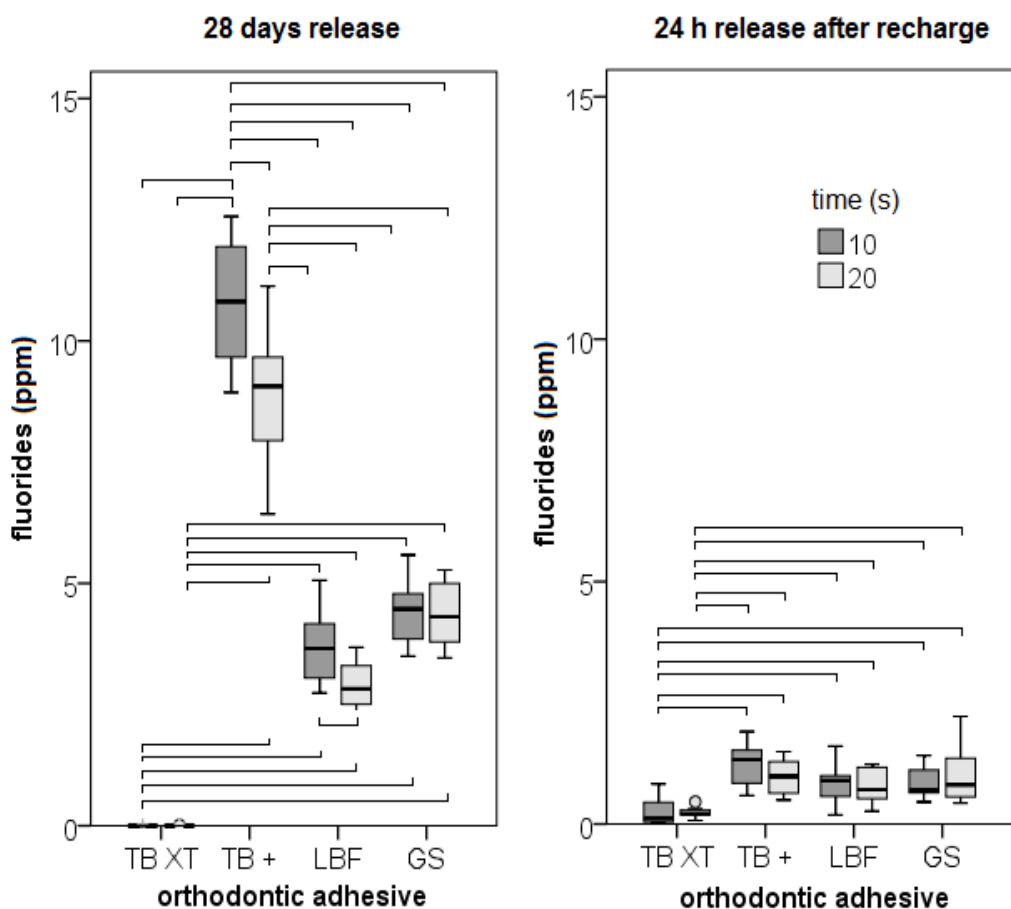
Figure 2. F release in adhesive-illumination groups. The line in the box represents the median, the box represents the interquartile range, whiskers represent the minimum and maximum values, and the point (°) stands for outlier. The lines denote groups between which the difference is significant at p<0.05.
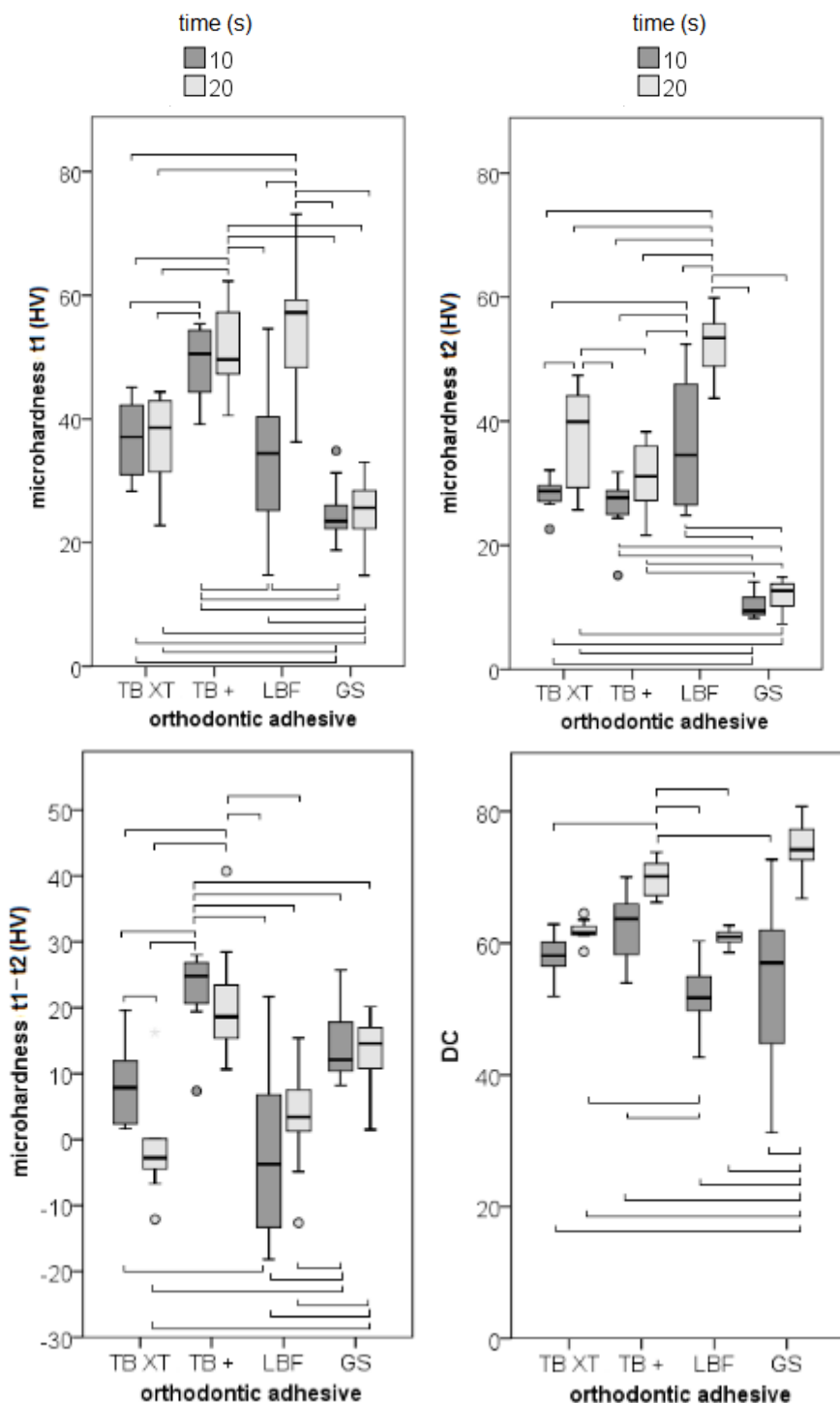
Figure 3. Mechanical properties in adhesive-illumination groups. The line in the box represents the median, the box represents the interquartile range, whiskers represent the minimum and maximum values, and the point (°) stands for outlier. The lines denote groups between which the difference is significant at p<0.05.

F release was related to adhesive system type, curing time, and combination of adhesive and time ($p < 0.05$) and decreased in the following order: $TB+_{10} > TB+_{20} > GS_{10} > GS_{20} > LBF_{10} > LBF_{20}$ (Figure 1). The subscript indicates illumination time. Significantly higher release was found in LBF and TB+ after 10 sec of illumination than in their 20 sec pairs ($p < 0.05$). F release 24 hours after recharge was similar in all F-containing adhesive systems, but much higher than in the control TBXT ($p < 0.001$). TB+ tended to have superior and LBF inferior ion release potential after recharge. Curing time had no influence on F release after recharge. The decrease of μH after four weeks of immersion in artificial saliva is related to increased release of F ($r = 0.651$; $p < 0.001$; Table 2).

Table 2. Correlation between change of microhardness ($\Delta\mu H$), curing time, and F release (without non-biointeractive adhesive TBXT)

		$\Delta\mu H$	Time	F release 28 days	F release 24 hours after recharge
$\Delta\mu H$	r	1	0.040	0.651*	0.209
	p		0.764	<0.001	0.119
Curing time	r	0.040	1	-0.176	−0.039
	p	0.764		0.186	0.773
F release 28 days	r	0.651*	−0.176	1	0.432*
	p	<0.001	0.186		0.001
F release 24 hours after recharge	r	0.209	−0.039	0.432*	1
	p	0.119	0.773	0.001	

* denotes significant correlations

The time of illumination of the adhesive system was not related to the change of μH, nor to the quantity of released F after 28 days, or after recharge ($r = 0.432$; $p = 0.001$). DC showed significant correlation with the time of illumination ($r = 0.697$; $p < 0.001$), but, however, not with the initial μH, change of μH, nor ion release after 24 hours, or after recharge.

DISCUSSION

The present study demonstrates that the release of F after orthodontic bracket bonding is related to the chemical composition, illumination time, and mechanical properties of F-containing orthodontic bonding materials. The amount of released ions after recharge is similar irrespective of chemical composition and curing time of the F-containing materials. Still, the initial F release tends to be proportionally related to F release after recharge, although accounting for only a small portion of variability of 19%. Since it has been proven that the highest amount of F release occurs in the first 24 hours,[19] it is evident that F-containing adhesives have low long-term caries-preventive potential. Although TB+ initially demonstrated superior F release, the comparison of tested materials highlights that it has the lowest potential of storage and release of ions after recharge (11% of initial capacity), and GS and LBF show a higher potential (21 and 25%, respectively). Previous research has shown limited F recharge capacity of orthodontic adhesives, as well.[20] By comparing the difference in the release of F after recharge between TBXT and TB+, it appears that the surface accumulation of F could be around 2.5%. The present study demonstrated that F release is linearly related to change of µH and that a higher quantity of released ions leads to a decrease in µH, which raises concerns about bond strength properties. Although a strong linear relationship between the DC and µH of composite materials has been reported,[21] it was not the case in this study. The results of our study are in concordance with Tassery et al.[22] Moreover, change in µH due to exposure to saliva appears not to be directly related to DC. Probably both DC and µH are influenced by the chemical composition of materials.[13] DC is related to the time of illumination with higher curing time inducing higher DC, probably not linearly but rather presenting a logarithmic trend line, since in shorter time (3 sec) DC appears to be much lower, while in longer curing times (6 and 8.5 sec) the difference is small.[13] DC in the present research is not related to ion release after 24 hours or after recharge, although there is evidence of higher F release in materials displaying higher DC.[23] The advantage of the present research is that it simulates real geometry and amount of adhesive as well as a real DC of orthodontic adhesive systems, since both components of the adhesive system were used. Our results indicate that the long-term reservoir potential for F ions in these materials is low. In our opinion, the use of F releasing orthodontic adhesives is not justified.

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