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Effect of a neutral citrate solution on the fluoride release of conventional restorative glass ionomer cements

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Summary *Objectives*. This study investigates the effect of a neutral citrate solution on the fluoride release of 10 acid–base setting glass ionomer cements during 140 days at 37 °C.

Methods. Five disks of 10 acid–base setting restorative glass ionomers were prepared according to the manufacturer's instructions. These specimens were immersed individually in 25 ml of a 0.01 mol/l citrate solution with pH = 7. Over 140 days, the solutions were regularly renewed and the fluoride concentration eluted during each period was determined with a combined fluoride ion selective electrode.

Results. The cumulative fluoride release was the result of an initial high release that ceased after some time and a long-term of low fluoride release. The long-term fluoride release was higher in neutral citrate solution than in water. For some formulations the short-term fluoride release also was higher in neutral citrate solution than in water suggesting that the polysalt matrix composition could be important in this respect.

Significance. The fluoride release process is due not only to a loss of relatively loosely bound fluoride in the cement matrix, but also to the release of strongly bounded fluoride inducing a long-term fluoride release. The effect of citrate on the fluoride release process may increase depending on the acid (polyacrylic acid versus copolymers of polyacrylic acid) used for the polysalt formation in the hardening cement. Depending on the competition between the polyacrylate anion and the citrate anion for the metal cation extraction the fluoride release process may be retarded.

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Introduction

Glass ionomer cements (GIC) are considered as adhesive bioactive materials with therapeutic action due to a continuous fluoride release over extended periods of time [1]. It is generally accepted that glass ionomers inhibit secondary caries in vitro, though this property apparently is not corroborated convincingly by in vivo findings [2,3]. A caries prophylactic effect of GIC could be ascribed to an incorporation of fluoride ions in enamel and dentin, and it is also known that the fluoride ions can inhibit the adherence of bacteria or bacterial growth [4-6].

Several studies [7-13] have shown that the fluoride release is enhanced in acidic solutions. Solutions of organic acids such as acetic, lactic and citric acid have been used to determine the effect of acids on the in vitro fluoride release. The low pH of such solutions results in an increase of the GIC fluoride release [8,12,13]. Moreover, it was suggested that the complexing ability of the anion of these organic acids also contributes to the enhanced GIC fluoride release [8,12,14]. This would imply that in neutral citrate solutions complex formation could result in an enhanced fluoride release of GIC. Such effect has been demonstrated for resin-modified glass ionomers and polyacid-modified composites [15], but not extensively on conventionally setting restorative GICs studied.

Therefore, the purpose of the present study is to investigate the effect of a neutral citrate solution on the fluoride release of conventionally setting GIC. Citrate is most suited for such study in view of its great complexing ability with metal cations such as Al^{3+} and Ca^{2+} compared to e.g. acetate [16-18].

Materials and methods

In the present study, the GIC formulations used in a previous investigation were investigated [19]. The composition of these restorative glass ionomer cements are summarized in Table 1. The glass ionomers that were supplied in powder/liquid were hand-mixed and handled according to the prescriptions of the manufacturer (filling consistency). The encapsulated glass ionomers were mechanically mixed for 10 s using a Silamat (Vivadent, Schaan, Liechtenstein).

Five disks (6 mm in diameter and 3 mm thick) of each glass ionomer formulation were made at different times using a fresh mix for each sample. The cement paste was then transferred or injected (capsules) into a split stainless-steel mould. After being filled, the mould was closed (separation with celluloid strips), excess of material removed and the material was allowed to set under pressure at room temperature for 15 min. Subsequently, each glass ionomer disk was equilibrated into a polyethylene flask containing 25 ml of an 0.01 mol/l citrate solution adjusted at pH 7 with a KOH solution. The flasks were shaken in a water bath at 37.0($\pm$0.1) °C at a rate sufficient for a good homogenization of the solutions without causing turbulence.

After 0.25, 1, 2, 3, 4, 7, 14, 21, 28, 56, 84, 112 and 140 days from the start of the equilibration, the disks were taken out of the respective solutions, quickly blotted dry with filter paper and immediately immersed in another 25 ml of freshly-made testing medium. The concentration of fluoride in the leaching solutions was determined after being diluted (1:1) with Total Ionic strength Adjustment Buffer (TISAB), and using a reference and fluoride-ion selective

Table 1 Glass ionomer formulations used in the present study.

Product	Powder	Liquid	P/L	Batch no.
Fuji II (hand-mixed)	CAFS-glass + PAA	PAA + TA + water	2.7:1	P: 090582, L: 090582
Fuji Cap II (capsule)	CAFS-glass + PAA	PAA + TA + water	2.8:1	Cap: 261281
Miracle Mix (hand-mixed)	CAFS-glass + PAA + AA-powder	PAA + TA + water	5.9:1	P: 180360, L: 230260, AA: 030888
Chemfil II (hand-mixed)	CAFS-glass + PAA + TA	water	6.8:1	P: 880522, L: water
Chemfil Cap II (capsule)	CAFS-glass + PAA + TA	PAA + water	3.3:1	Cap: 891030
Chelon-Fil (hand-mixed)	CAFS-glass (Ca:Na:F:P:Al: Si: = 11:2:13:2:16:56)	PAMA + TA + water	3.2:1	P: T 032, L: T 009
Ketac-Fil (capsule)		PAMA + TA + water	2.95:1	Cap: S 293
Chelon-Silver (hand-mixed)	CAFS-glass (Ca:Na:F:Al: Si: = 11:2:13:2:16:56) +	PAMA + TA + water	3.2: 1	P: T 113, L: T 113
Ketac-Silver (capsule)	sintered Ag (0.92:1)	PAMA + TA + water	3.72: 1	Cap: T 052
Shofu II (hand-mixed)	CAFS-glass	PACA + TA + water	2.5: 0.8	P: 068807, L: 788000

CAFS-glass: calcium alumino fluoro silicate glass; PAA: poly(acrylic acid); TA: tartaric acid; PAMA: copolymer of acrylic and maleic acid; PACA: copolymer of acrylic and tricarboxylic acid.