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Enhancement of fluoride release from glass ionomer cement following a coating of silver fluoride

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Abstract

Background: This study investigated the extent to which a coating of 10% silver fluoride (AgF) on discs of glass ionomer cements (GICs) would enhance the release of fluoride ion into eluting solutions at varying pH.

Materials and methods: Forty discs each of Fuji IX, Fuji VII and of Vitrebond were prepared in a plastic mould. Twenty discs of each material were coated for 30 seconds with a 10% solution of AgF. Five discs each of coated and uncoated material were placed individually in 4ml of differing eluant solutions. The eluant solutions comprised deionized distilled water (DDW) and three separate acetate buffered solutions at pH 7, pH 5 and pH 3. After 30 minutes the discs were removed and placed in five vials containing 4ml of the various solutions for a further 30 minutes. This was repeated for further intervals of time up to 216 hours, and all eluant solutions were stored. Fluoride concentrations in the eluant solutions were estimated using a fluoride specific electrode, with TISAB IV as a metal ion complexing and ionic concentration adjustment agent. Cumulative fluoride release patterns were determined from the incremental data.

Results: The coating of AgF greatly enhanced the level of fluoride ion release from all materials tested. Of the uncoated samples, Vitrebond released the greater concentrations of fluoride ion, followed by Fuji VII. However, cumulative levels of fluoride released from coated samples of the GICs almost matched those from coated Vitrebond.

Conclusions: It was concluded that a coating of 10% AgF on GICs and a resin modified GIC greatly enhanced the concentration of fluoride released from these materials. This finding might be applied to improving protection against recurrent caries, particularly in high caries risk patients, and in the atraumatic restorative technique (ART) of restoration placement.

Key words: Silver fluoride, glass ionomer cement, fluoride release, acidic saliva.

Abbreviations and acronyms: AgF = silver fluoride; ART = atraumatic restorative treatment; DDW = deionized distilled water; GIC = glass ionomer cement; HEMA = hydroxyethyl methacrylate; KI = potassium iodide; SnF₂ = stannous fluoride; RMGIC = resin modified glass ionomer cement; TISAB = total ionic strength adjustment buffer.

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INTRODUCTION

The use of GICs as a lining or restorative material has been shown to provide an advantage over other restorative materials in reducing the development of secondary or residual caries.^{1,4} This has been claimed to occur largely as a result of the release of fluoride ion from these materials into the adjacent tooth structure or external tooth environment.^{3,5,6} However, analysis of these reports indicates that the rate of fluoride ion release varies considerably amongst glass ionomer cements (GICs). There is also evidence that GICs can take up and store fluoride ions from sources entering the environment of the teeth, in particular concentrated topical fluorides.⁷

One possible avenue to enhance fluoride ion uptake and storage in GIC is the application of silver fluoride solution (AgF). In the late 1970s, paediatric dentistry staff in the Faculty of Dentistry at The University of Sydney, applied 40% AgF solution to the teeth of children with large cavities to slow down the progression of caries and prevent the exposure of the dental pulp at a time when there were insufficient staff to carry out more comprehensive treatment. Studies by Craig *et al.*⁸ showed that there was a significant reduction in the progression of caries when the teeth were treated with 40% AgF and 10% stannous fluoride (SnF₂). However, Gotjamanos⁹ has expressed concern at the concentration of fluoride ion in a 40% solution and its potential fluoritic effect on young children. Also, the advent of GICs and atraumatic restorative treatment (ART) now make the use of these materials and methods, even in remote communities, an

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alternative approach to caries control. Even so, there may still be an advantage in combining both GIC and AgF treatments to achieve an even greater degree of success in caries management, particularly in remote communities, but at a reduced concentration of AgF.

One of the disadvantages of the use of AgF was the dark staining of oral tissues that resulted. This can now be prevented from occurring by the subsequent immediate application of a concentrated potassium iodide (KI) solution (Craig, personal communication; Knight *et al.*¹⁰).

The objective of this experiment was to determine the potential to increase release of fluoride ion from Fuji IX, Fuji VII (GC Corp., Japan) and Vitrebond (3M ESPE, USA) into deionized distilled water (DDW) and a variety of acetate buffered solutions at differing pH levels following exposure of these materials to a 10% solution of AgF. These solutions were intended to simulate saliva in terms of its calcium and phosphate ion content, and the pH to which the teeth and restorations could be periodically exposed.

MATERIALS AND METHODS

Preparation of GIC discs

Forty discs each of Fuji IX, Fuji VII and Vitrebond were prepared to a size of 4mm diameter x 6mm depth using a plastic mould. Vitrebond, a resin modified GIC (RMGIC), was included as it has been found in previous studies to have the highest levels of fluoride ion release of all GICs.¹¹ All discs were allowed to set for 24 hours in a humid environment before use. Twenty discs of each material were immersed in 10% AgF solution for one minute and dried on blotting paper.

Preparation of eluant solutions

Eluting solutions comprised DDW (pH 6.5) and acetate buffers based on the artificial caries demineralization solution of ten Cate and Duijster.¹² These contain 0.5M glacial acetic acid and 2.2mM CaHPO₄. The pH of separate samples of the solutions was adjusted using 5% NaOH to pH 7, pH 5 and pH 3. Two hundred and fifty vials containing 4ml of each eluant solution were prepared. This allowed for five eluant samples to be collected from each of the six categories of GIC, at each change over period, together with control eluant samples.

Method

The six categories of GIC discs, Fuji IX, Fuji VII and Vitrebond, both with AgF coating and without, were placed in vials of eluant solutions. After 30 minutes, all discs were removed and placed in fresh solutions for a further 30 minutes. This change of solutions occurred at the following intervals of time to 216 hours (nine days): 0.5, 1, 2, 4, 8, 16, 32, 64 and 128 hours. At 216 hours the experiment was terminated with the discs being stored in a moist environment for future

Table 1. i, ii, iii, iv: Mean incremental fluoride release from Fuji IX, Fuji VII and Vitrebond into eluants at different pH levels (ppm or µg/ml)

i. DDW						
Hours	Fuji IX	Fuji VII	Vitrebond	Fuji IX + AgF	Fuji VII + AgF	Vitrebond + AgF
0.5	0.07	0.32	0.74	23.0	24.0	10.0
1	0.039	0.24	0.5	2.7	5.2	4.0
2	0.034	0.38	0.9	1.6	6.0	4.1
4	0.058	1.0	1.6	1.0	7.0	3.9
8	0.09	0.8	3.0	0.9	6.0	6.2
24	0.11	0.9	4.0	0.5	2.5	5.0
72	0.36	3.0	12.0	0.72	3.4	9.8
216	0.36	3.0	12.0	0.92	3.4	16.0

ii. pH 7						
Hours	Fuji IX	Fuji VII	Vitrebond	Fuji IX + AgF	Fuji VII + AgF	Vitrebond + AgF
0.5	0.044	0.018	0.17	11.0	26.0	2.9
1	0.025	0.018	0.036	0.37	1.0	0.7
2	0.018	0.018	0.062	0.38	0.7	0.74
4	0.018	0.02	0.13	0.37	0.6	1.7
8	0.018	0.02	0.47	0.38	0.81	2.2
24	0.016	0.028	0.6	0.25	0.62	1.6
72	0.016	0.31	2.0	0.5	1.2	3.1
216	0.016	0.32	2.3	0.8	1.3	6.0

iii. pH 5						
Hours	Fuji IX	Fuji VII	Vitrebond	Fuji IX + AgF	Fuji VII + AgF	Vitrebond + AgF
0.5	0.058	0.086	1.1	24.0	16.0	5.0
1	0.058	0.086	0.4	1.9	2.9	1.3
2	0.058	0.11	0.8	2.0	4.0	1.4
4	0.07	0.24	1.5	1.7	4.0	1.1
8	0.11	0.37	2.0	1.7	3.1	3.5
24	0.18	0.61	3.2	1.1	3.1	3.3
72	0.45	2.1	5.1	1.6	5.0	5.0
216	0.51	2.6	6.4	1.6	4.9	11.0

iv. pH 3						
Hours	Fuji IX	Fuji VII	Vitrebond	Fuji IX + AgF	Fuji VII + AgF	Vitrebond + AgF
0.5	1.0	3.8	4.4	11.5	14.0	9.6
1	1.2	3.7	5.4	3.6	10.0	8.4
2	2.8	6.8	8.3	2.3	11.0	10.0
4	3.7	7.2	11.0	2.0	9.2	12.0
8	4.0	8.2	11.0	1.8	7.2	14.0
24	4.0	9.2	17.0	1.5	5.0	17.0
72	5.8	14.0	23.0	2.1	6.2	24.0
216	4.8	15.0	31.0	2.7	8.4	26.0

reference. All 960 eluant samples, together with control eluant samples, were stored at 4°C until assessed for fluoride ion content.

Readings from each of the five samples were determined for 10 separate categories of test material, and the differences between readings for each category were found to be less than one per cent. For this reason, as there were greater than 1000 samples to be read, it was decided to pool the five eluant samples from each category.

Assessment of fluoride ion content in each eluant sample

Incremental fluoride release during the exposed time periods was measured using a fluoride specific ion electrode (Orion, USA). Total ionic strength adjustment

Table 2. i, ii, iii, iv: Accumulative amounts of fluoride released from Fuji IX, Fuji VII and Vitrebond into eluants at different pH levels (ppm or µg/ml)

i. DDW						
Hours	Fuji IX	Fuji VII	Vitrebond	Fuji IX + AgF	Fuji VII + AgF	Vitrebond + AgF
0.5	0.07	0.32	0.74	23.0	24.0	10.0
1	0.109	0.56	1.24	25.7	29.2	14.0
2	0.143	0.94	2.14	27.3	35.2	18.1
4	0.201	1.94	3.74	28.3	42.2	22.0
8	0.291	2.74	6.74	29.2	48.2	28.2
24	0.401	3.64	10.74	29.7	50.7	33.2
72	0.761	6.64	22.74	30.42	54.1	43.0
216	1.121	9.64	34.74	31.34	57.5	59.0

ii. Acetate buffer, pH 7						
Hours	Fuji IX	Fuji VII	Vitrebond	Fuji IX + AgF	Fuji VII + AgF	Vitrebond + AgF
0.5	0.044	0.018	0.17	11.0	26.0	2.9
1	0.069	0.036	0.206	11.37	27.0	3.6
2	0.087	0.054	0.268	11.75	27.7	4.34
4	0.105	0.074	0.398	12.12	28.3	6.04
8	0.123	0.094	0.868	12.50	29.11	8.24
24	0.139	0.122	1.468	12.75	29.73	9.84
72	0.155	0.432	3.468	13.25	30.93	12.94
216	0.171	0.752	5.768	14.05	32.23	18.94

iii. Acetate buffer, pH 5						
Hours	Fuji IX	Fuji VII	Vitrebond	Fuji IX + AgF	Fuji VII + AgF	Vitrebond + AgF
0.5	0.058	0.086	1.1	24.0	16.0	5.0
1	0.116	0.172	1.5	25.9	18.9	6.3
2	0.174	0.282	2.3	27.9	22.9	7.7
4	0.244	0.522	3.8	29.6	26.9	8.8
8	0.354	0.892	5.8	31.3	30.0	12.3
24	0.534	1.502	9.0	32.4	33.1	15.6
72	0.984	3.602	14.1	34.0	38.1	20.6
216	1.494	6.202	20.5	35.6	43.0	31.6

iv. Acetate buffer, pH 3						
Hours	Fuji IX	Fuji VII	Vitrebond	Fuji IX + AgF	Fuji VII + AgF	Vitrebond + AgF
0.5	1.0	3.8	4.4	11.5	14.0	9.6
1	2.2	7.5	9.8	15.1	24.0	18.0
2	5.0	14.3	18.1	17.4	35.0	28.0
4	8.7	21.5	29.1	19.4	44.2	40.0
8	12.7	29.7	40.1	21.2	51.4	54.0
24	16.7	38.9	57.1	22.7	56.4	71.0
72	22.5	52.9	80.1	24.8	62.6	95.0
216	27.3	67.9	111.1	27.5	71.0	121.0

buffer IV (TISAB IV; Orion, USA) was used to de-complex contaminating metal ions presumed to also be leached from the GICs, and to provide a more constant background ionic strength. One millilitre of TISAB IV buffer was added to 1ml of each eluant sample. Standard fluoride solutions were prepared by mixing equal volumes of fluoride standards at concentrations of 0.01ppm, 0.1ppm, 1.0ppm, 10ppm, 100ppm in DDW with equal volumes of TISAB IV. Millivolt readings of standard fluoride/TISAB IV solutions were measured using the fluoride specific ion electrode with a digital pH/mV meter (Orion, USA).

Three samples of each eluate were tested for each time and eluate category, and the mean of the three separate readings calculated. The results were used to establish the standard reference curve, which showed

the relationship between the logarithm of fluoride concentration in the solution and the millivolts measured with the electrode. The readings in millivolts from all the samples of unknown fluoride concentration were converted to ppm F using the reference graph. The cumulative pattern of fluoride release for each sample was later calculated from the incremental data and expressed in µg/ml.

RESULTS

Table 1 shows the incremental concentrations of fluoride ion released into the different eluant solutions over the various time periods, in ppm (µg/ml). These results are shown in cumulative levels at the various collection times in Table 2.

Of the uncoated materials, Vitrebond showed the highest cumulative fluoride release followed by Fuji VII and Fuji IX under all conditions tested. Fluoride ion was still being released by the ninth day (216 hours). The highest cumulative fluoride release was at pH 3 followed by DDW (Vitrebond) and pH 5 (Fuji IX and Fuji VII). The lowest cumulative release was at pH 7 in all the samples tested.

The GICs and the RMGIC all showed increased fluoride release when coated with 10% AgF. However, the pattern of uptake varied considerably between the GICs and the RMGIC. The initial fluoride release patterns from Vitrebond + AgF were considerably lower than for Fuji VII and IX + AgF, though in most cases the cumulative amount released from Vitrebond increased to almost the same final amount as for the GICs.

DISCUSSION

The results indicated that surface application of a 10% solution of AgF to GICs will enhance the amount of fluoride released into the surrounding environment, irrespective of which materials already demonstrate a high rate of fluoride release in their own right. However, as stated earlier, this application must be followed immediately by an application of KI to prevent dark staining of the GIC and adjacent enamel occurring. A prior coating of AgF/KI to demineralized dentine immediately prior to the placement of a Fuji IX restoration did not significantly alter the uptake of fluoride ion into the dentine compared with when AgF alone was applied.¹³ Even though this is a different model system, it suggests that a similar result might apply when GIC coated with AgF/KI is exposed to the wider oral environment.

The baseline result from all materials was in accordance with the information provided by the manufacturers. Vitrebond by itself provides the highest accumulation of fluoride ion released compared with both conventional GICs tested. The composition and structure of Vitrebond is different compared with conventional glass ionomer cement, it being an RMGIC in which several components were added such as

hydroxyethyl methacrylate (HEMA) and photoinitiator, besides the other common compositional elements of conventional GICs. However, it is currently not known why its fluoride release is so high in comparison with that from the conventional GICs.

These findings are of significance to clinical dentistry. Even though AgF might be applied externally to GIC restorations, or coat the base of a cavity prior to placement of GIC, its effect in enhancement of the fluoride release pattern from such restorations would still substantially increase the concentration of fluoride available at these locations. The majority of research into the role of fluoride released from GICs supports the claim that fluoride released from GICs has an anti-cariogenic effect and enhances the remineralization process on the tooth surface.^{5-7,14,15}

These results are consistent with those demonstrated in previous studies.¹⁶⁻¹⁸ Silver fluoride treated GICs released more fluoride compared with untreated cements.¹⁸⁻²⁰ These results also show that acidic conditions will increase the release of fluoride; the lower the pH, the higher the cumulative release of fluoride, as found in previous studies.^{17,21} The greatest accumulation of fluoride released was at pH 3, followed by pH 5 (Fuji IX and Fuji VII) and DDW (Vitrebond). Results of fluoride release were lowest at pH 7 in all the samples tested.

Thus these data also demonstrated the combined action of acidity and relative ionic concentration of the eluant solutions in contributing to the F release pattern. GIC discs release more fluoride ion in DDW compared with that in pH 7 eluant solution. There were few ions present in the DDW solution, and the less tightly bound fluoride ions were released, resulting in a higher, cumulative release of fluoride than at pH 7 with equivalent tonicity to the remaining solutions. Previous studies showed that glass ionomer cements release more fluoride in deionized water compared with artificial saliva.^{21,22} On the other hand, GIC discs in pH 5 and pH 3 solutions showed higher fluoride release compared with that at pH 7 due to the higher level of acidity. The question arises as to whether this occurs as a result of a dissolution process or an acid-activated, equilibrium-driven process of diffusion. It possibly involved both. There was evidence that with sustained acidic pH challenge, loss of surface material occurs from the traditional GICs.²³ However, chemically, the conditions indicated that a diffusion process must be a major driving force in the fluoride release process.

Another factor was the specific differences in F release profiles between the different GIC based materials. Vitrebond gave the greatest accumulation of fluoride release at pH 3 followed by DDW. However, it also provided the lowest fluoride release, up to 30 minutes from when it was coated with 10% AgF, followed by a relatively greater increase in fluoride release with time.

Unfortunately, the method for assessment of fluoride release did not allow for discrimination between originally bound fluoride ion and more freely available.

It was not possible to make these measurements with a fluoride electrode without the use of TISAB as the electrode would be seriously contaminated by the metal ions in the solution. Alternative methods such as prior separation of the fluoride by acid hydrolysis did not assist in this regard.

The study by el Mallakh²⁴ concluded that the *in vitro* release of fluoride from glass ionomer cements does not represent the actual release in the oral environment. Nevertheless, the addition of the AgF substantially increases the amount of fluoride release *in vitro*, and it might be expected that this additional release will occur *in vivo*. As such, as well as contributing to inhibition of demineralization and enhancement of remineralization, this additional concentration might reach that at which it could exert a bacteriostatic effect.

A study by Marsh and Bradshaw²⁵ showed that 1mmol/L fluoride (18.87ppm (µg/ml) F) combined with a moderated low pH can prevent *Streptococcus mutans* growth. The results of fluoride release on normal GIC discs (Fuji IX and Fuji VII) at 30 minutes ranged between 0.018ppm (µg/ml) F to 3.8ppm (µg/ml) F (9×10^4 mmol/L to 0.20mmol/L). The results of fluoride release at 30 minutes after GIC discs (Fuji IX and Fuji VII) were coated with 10% AgF ranged between 11ppm F to 26ppm F (0.58mmol/L to 1.378mmol/L). The higher concentration of fluoride release after coating with 10% AgF might be sufficient to exert some inhibition of bacterial growth. However, it would exert a marginal effect compared with the effect of the low pH at which such concentrations of fluoride are released and the additive effect of the heavy metal ion also present.

CONCLUSION

GICs and RMGICs can have their fluoride concentrations enhanced by a coating of silver fluoride and release more fluoride ion into the oral environment compared with untreated materials. It would be essential to follow this coating immediately with KI to ensure no staining resulted. There is evidence to suggest that the addition of KI would not significantly affect the concentration of fluoride released.

The greatest accumulation of fluoride released was at pH 3, followed by pH 5 (Fuji VII and Fuji IX) and DDW (Vitrebond). This method of enhancement of fluoride release could be very beneficial for patients with high caries risk and could be used in conjunction with the ART technique. However, additional clinical trials are needed to evaluate the effect of silver fluoride/potassium iodide in conjunction with GIC on dental caries *in vivo*.

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