Discolouration of Glass-Ionomer Cement at Different Fluoride Concentration Levels

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Abstract

Background: Although application of professionally applied Acidulated Phosphate fluoride (APF) gel is effective in prevention of dental caries, APF-induced discolouration and/or surface degradation of dental restorations has been reported. However, no publication has reported a dose-response effect of APF gel on Glass Ionomer Cement (GIC) restorations *in vivo* and/or human teeth. Therefore, this study aimed to examine the discolouration effect of APF gel at various concentration levels on GIC restorations of human teeth *in vitro*.

Materials and Methods: Seventy extracted human teeth were used. Each was restored with GIC and then topically applied with a different dilution of APF gel. Change of shade and/or colour of restorations was assessed by a trained examiner.

Results: GIC discolouration was detected on 4 (5.71%) and 23 specimens (32.86%) after the first and the second APF applications, respectively. The discolouration rate increased with APF gel concentration on the second application (χ^2=38.314, df=1, p<0.001) but not the first (χ^2=2.352, df=1, p=0.125). Discolouration of GIC restorations was more likely to form under application of a higher concentration of APF gel (OR=1.099, 95%CI=1.052, 1.148), a higher concentration of fluoride (OR=1.001, 95%CI=1.000, 1.001) and/or a lower pH value (OR=0.002, 95%CI=0.000, 0.039).

Conclusions: Discolouration of GIC was associated with concentration and frequency of APF gel application in extracted natural human teeth *in vitro*. Increasing discolouration rates were related to increased fluoride concentration and increasing acidity of APF gel. Further investigation is indicated.

Key words: Fluoride, Glass ionomer cement, Discolouration, pH

Introduction

Changes in surface morphology of aesthetic restorative materials, particularly composite resins, following the application of professionally applied Acidulated Phosphate Fluoride (APF) gel *in vitro* have been reported in the literature [1-10]. Studies have shown that composite resins are susceptible to surface degradation, evident as increased surface roughness [1-5] and weight loss [2,6]. Loss of the filler particles in composite resins as a result of acid dissolution has been attributed as a possible cause of surface degradation [1,2,4-7]. Previous studies have reported that APF gels similarly result in surface degradation of Glass Ionomer Cement (GIC). Cehreli *et al.* [4], Neuman and Garcia-Godoy [8], and Yip *et al.* [9] have shown that the application of APF gel increases the chemical erosion of certain GICs by acid-etching the surface. These studies suggested that the roughened surface contributes to plaque accumulation and produces surface staining. However, no literature could be found that tested the effect of APF gel on GIC restorations *in vivo* or extracted natural human teeth.

The application of APF gel has been shown to cause colour instability in indirect composites [10]. Nevertheless, there is a lack of literature reporting the aesthetic changes to GIC following professional APF gel application. This is particularly pertinent as professionally applied topical fluorides are used to recharge fluoride-releasing restorative materials [11]. Literature reporting the effect of diluting APF gel on discolouration, recharge or surface degradation of GICs is also scarce. Therefore, this study aimed to examine the discolouration effect of APF gel at various concentration levels on GIC restorations *in vitro*.

Materials and Methods

Human teeth were used in this study. The minimal sample size required was calculated with Epi Info (Version 7.0.9.7, Centers for Disease Control and Prevention, Atlanta, GA, USA). We estimated that 10 human teeth in each group was the minimal number of subjects for reporting 95% confidence limits for the discolouration rate with an explanatory power at 80%. This calculation assumed the discolouration rates equal to 60% in the high fluoride group and 0.01% in the control group, with the 1:1 ratio of high fluoride subjects to control subjects. Since six different concentrations of fluoride were compared with the control group, a total of seventy extracted human teeth specimens collected from the Oral Surgery Department of the Oral Health Centre of Western Australia and from the Morley Dental Clinic were used in this study. The selection criteria included permanent molars with a minimum requirement of at least the buccal or lingual surface intact. The specimens were stored in 5000 ppm sodium hypochlorite solution for over 24 hours and were rinsed thoroughly for 30 seconds in tap water prior to preparation. A box preparation of 2 mm in all dimensions was prepared for each specimen located to the intact buccal or lingual surface and the adjacent occlusal surface. Each specimen was restored with shade A2 Fuji IX

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Table 1. *The APF gel dilution for each group and the respective fluoride concentration and pH value as calculated.*

Group	APF dilution (%)	[F] ppm (%)	pH
A (control: tap water)	-	1 ± 0.2 (0.0001)[a]	7.0 ± 0.5
B	4.07	500 ± 40 (0.05)	4.9 ± 0.5
C	8.13	1000 ± 80 (0.10)	4.6 ± 0.5
D	16.26	2000 ± 160 (0.20)	4.3 ± 0.5
E	32.52	4000 ± 330 (0.40)	4.0 ± 0.5
F	65.04	8000 ± 650 (0.80)	3.7 ± 0.5
G	100	12300 ± 1000 (1.23)	3.5 ± 0.5

[a]Water fluoridation was implemented in Perth, Australia, at the time of the research.

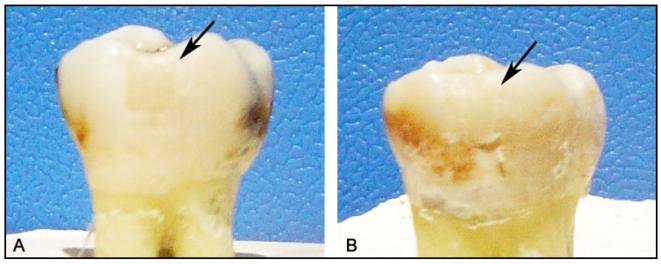

Figure 1. *Specimens displaying the typical appearance of the absence of discolouration. Arrows indicated the margin of restorations.*

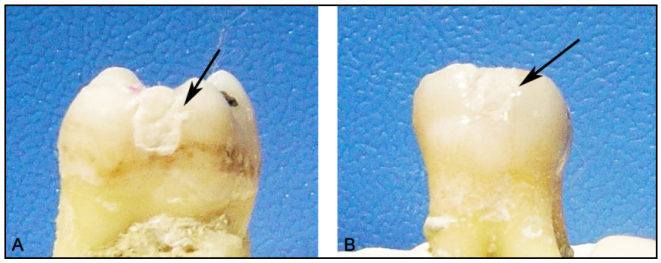

Figure 2. *Specimens displaying the typical appearance of the presence of discolouration. Arrows indicated the margin of restorations.*

GIC (GC America), mixed and prepared according to the manufacturer's specifications, smoothed over with a No. 6 plastic instrument and allowed to set over 24 hours.

The specimens were divided into seven groups, each group consisting of ten teeth. For each group, a different dilution of Vivoral topical APF gel (Ivoclar) was applied as shown in *Table 1*. The dilution was applied with a cotton bud for 4 minutes then wiped off with absorbent paper to simulate the manufacturer's directions for use. The dilutions were prepared by adding calculated amounts of tap water to the APF gel then shaken and mixed thoroughly. The specimens were stored at room temperature (25°C) for one hour then each specimen was thoroughly rinsed for 30 seconds with tap water to simulate the effect of the patient not eating or drinking as per the directions for use of the APF gel. Before and after the preparation, the human teeth were separately preserved in tap water to avoid dehydration. This was due to a recent study reporting a difference in shades between dehydrated and rehydrated teeth [12].

After 24 hours, comparisons of the specimens were made with the manufacturer's shade guide by a trained examiner, using a green background to re-calibrate the examiner's eyes between each comparison. Results were recorded as yes/no for discolouration, this being defined as a change in shade. Every fifth specimen was re-examined for intra-examiner reliability and produced 100% reliability. The respective concentrations were applied a second time to each specimen, and comparisons were made following the same protocol as outlined above. The results collated were subjected to statistical analysis using the IBM SPSS Statistics (version 20.0, IBM Corporation, Somers, NY, USA). One-way Analysis of Variance (ANOVA) comparison of means was used to determine the difference between the groups for each application. Linear-by-linear association chi square test was used to analyse the relationship between APF gel concentration and discolouration rates for both the first and second application. For the second application, logistic regression analysis was used to determine the quantitative effect of increasing APF gel concentration, fluoride concentration and pH values on discolouration rate.

Table 2. *Discolouration rates following 1st and 2nd application of APF gel.*

Group	1st Application Discolouration %	2nd Application Discolouration %
A	0	0
B	0	0
C	10	10
D	0	10
E	10	20
F	0	90
G	20	100

Results

Of the 70 specimens in total, discolouration of the GIC occurred in 4 of the specimens (5.71%) after the first application and 23 specimens (32.86%) after the second application. A comparison of the typical appearance of the absence and presence of discolouration of GIC can be viewed in *Figure 1 and 2*. Discolouration rates for each group after the first and second application are presented in *Table 2*.

One-way ANOVA comparison of means of the first application was unable to demonstrate a significant difference between any two given groups ($F=1.147$, $df=6$, $p=0.346$) but was able to demonstrate a significant difference between any two given groups for the second application ($F=27.209$, $df=6$, $p<0.001$). Using the linear-by-linear association chi square model, it was confirmed that there was differences between any two given groups on the second application and demonstrated that discolouration rates increased significantly with APF gel concentration on the second application ($\chi^2=38.314$, $df=1$, $p<0.001$) but not the first ($\chi^2=2.352$, $df=1$, $p=0.125$).

Following the second application, analysis of the relationship between APF concentration and discolouration rate by logistic regression displayed a chi square model of $\chi^2=57.011$ ($df=1$, $p<0.001$), a correct percentage of 92.9%, a Hosmer and Lemeshow goodness of fit at 2.040 ($df=5$,

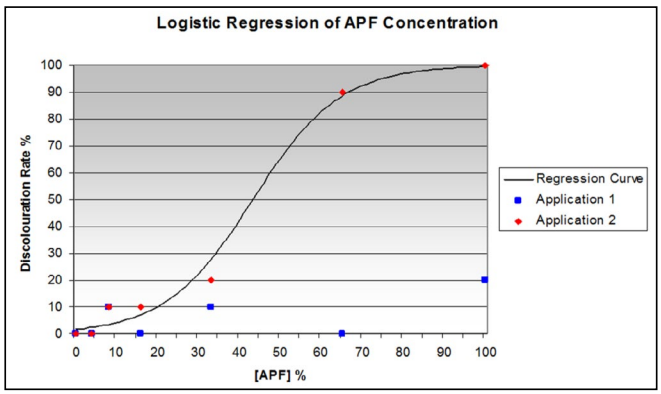

Figure 3. *Logistic regression curve of APF concentration for the second application and actual discolouration rates for each application.*

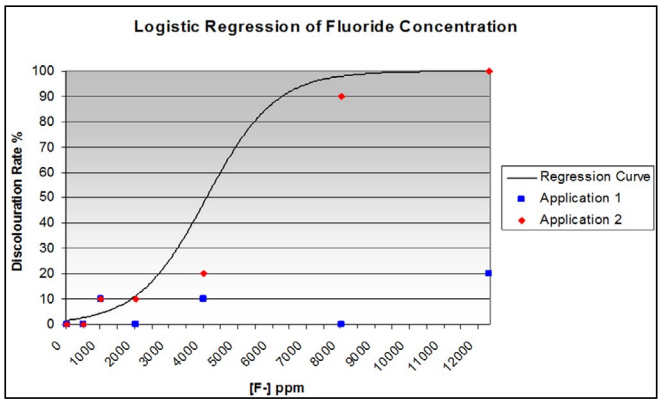

Figure 4. *Logistic regression curve of fluoride concentration for the second application and actual discolouration rates for each application.*

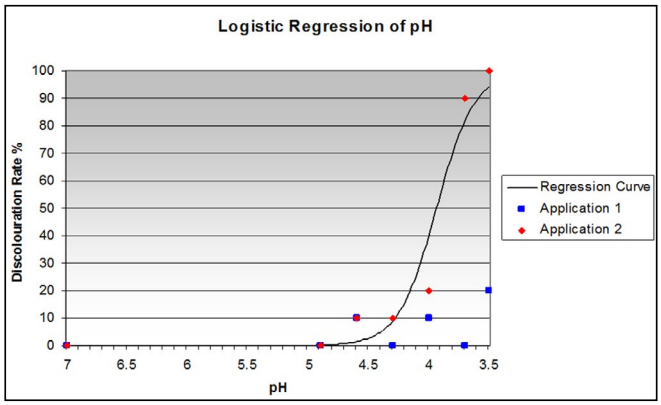

Figure 5. *Logistic regression of pH values for the second application and actual discolouration rates for each application.*

p=0.844). This confirmed a significant trend of increasing discolouration rates upon increasing APF gel concentration with an odds ratio of 1.099 (95%CI=1.052, 1.148). The logistic regression curve of APF concentration is displayed in *Figure 3*.

Similarly, analysis of the relationship between fluoride concentration and discolouration rate by logistic regression displayed a chi square model of χ^2=57.011 (df=1, p<0.001), a correct percentage of 92.9%, a Hosmer and Lemeshow goodness of fit at 2.040 (df=5, p=0.844). This demonstrated a significant trend of increasing discolouration rates upon increasing fluoride concentration with an odds ratio of 1.001 (95%CI=1.000, 1.001). The logistic regression curve of fluoride concentration is displayed in *Figure 4*.

Analysis of the relationship between pH value and discolouration rate by logistic regression displayed a chi square model of χ^2=53.208 (df=1, p<0.001), a correct percentage of 92.9%, a Hosmer and Lemeshow goodness of fit at 7.676 (df=5, p=0.175). This demonstrated a significant trend of increasing discolouration rates upon decreasing pH level with an odds ratio of 0.002 (95%CI=0.000, 0.039). The logistic regression curve is displayed in *Figure 5*.

Discussion

This study demonstrated that significant discolouration rates of GIC were only achieved, following two applications of APF gel. This suggests the possibility that a threshold exposure time in the form of prolonged and/or frequent contact to an acidic solution exists for discolouration of GIC to occur. Concurrently, increasing the concentration of the solution resulted in increased discolouration rates. This suggests that a similar threshold level may exist with regard to the pH value. Previous studies have attributed discolouration to increased surface roughness by chemical erosion and acid etching following the application of APF gel [4,8,9]. If acid attack is the sole cause of surface degradation which in turn is the sole cause of the discolouration, then this problem can be overcome by reducing the acidity of APF gel.

Alternatively, a threshold concentration and exposure time for fluoride uptake may exist for discolouration of GIC to occur. Billington *et al.* [13] and De Witte *et al.* [14] reported that neutral sodium fluoride solution similarly causes surface degradation to GIC. De Witte *et al.* [14] suggested that contact with NaF increases the concentration of fluoride within the cement allowing for greater numbers of fluoride ions available to compete with carboxylate groups to form complexes with Al3+ ions. The formation of these fluoride complexes $[Al(H_2O)_{6-n}F_n]^{3-n}$ with $n \geq 2$ results in reduced numbers of ionic cross-links and site-bounded aluminium. This causes a gradual disintegration of the polysalt matrix to the extent of which depends on the concentration, time and frequency of application of the fluoride solution. In theory, this hypothesis can be applied to acidic solutions such as APF as neither phosphate nor sodium has a role in the fluoride complex formation. If the fluoride concentration has an effect on the discolouration of GIC whether in tandem with acid concentration or not, then discolouration can be avoided by diluting APF gel. Studies on neutral sodium fluoride were unable to demonstrate a dose-response relationship [15,16]. Varying the fluoride concentration from 2% to 0.2% to 0.05% [15] or from 2.3% to 1.1% [16] showed no significantly different cariostatic effect. Further studies need to be carried out using a chroma meter to compare the extent of discolouration between a fluoride free acidic solution and APF gel in order to delineate the effects of fluoride and acid on GIC.

It is also interesting to note that discolouration appeared as areas of opacity that were localised to the margins of the restoration (*Figure 2*). All of the studies reviewed were neither *in vivo* nor in restored extracted teeth but carried out on lone standing cylinders, disks or blocks of the restorative material. There are two possible suggestions for the localisation of discolouration. Firstly, increased surface area to volume ratio

of the restoration allows greater exposure to fluoride and/or acid at the line angles and vertices. If this were true, we would expect to see discolouration occurring at the occlusal buccal line angle of the restoration as well, which we did not. Secondly, the tooth/restoration interface may have a causal role in discolouration. A discrepancy in the tooth/restoration interface may allow greater retention of APF gel at the margins and thus increased exposure time in these areas.

This study used an observational method to assess the discolouration. This was a research limitation of the study. Application of techniques such as scanning electron microscopy [2-4,6-9], photomicrograph [5] and spectroscopy [3] would benefit the methodology of this study. Future investigation is indicated.

Conclusions

Discolouration of GIC was associated with concentration and frequency of APF gel application in extracted natural human teeth *in vitro*. Increasing discolouration rates were related to an increased fluoride concentration and increasing acidity of APF gel. Further research with a larger sample size and comparisons to fluoride free acidic solution and neutral fluoride solution is indicated.

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