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Effects of Temperature on the Fluoride Release and Recharging Ability of Glass Ionomers

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Clinical Relevance

The use of a higher temperature during topical fluoride applications may increase the fluoride recharging and re-release ability of glass ionomers, and a low oral environment temperature should be avoided during topical fluoride application.

SUMMARY

This study investigated the effects of environmental temperature on the fluoride release and recharging ability of glass ionomers. Five disk specimens (15 mm in diameter and 1 mm thick) were made of each of the following materials: a conventional luting glass ionomer, two high viscosity restorative glass ionomers and a restorative resin-modified glass ionomer. The fluoride release of each material was measured at 4°C, 37°C and 55°C. An additional three groups, which were made of the same materials, were stored in distilled and deionized water for 30 days and recharged in 250 ppm fluoride solution at 4°C, 37°C and 55°C for five minutes. The fluoride re-release was measured daily from two days prior to two days after the recharging process. At all temperatures, the luting glass ionomers showed the greatest fluoride release and recharging ability, followed by the resin-modified glass ionomer, then the high viscosity glass ionomers. For each material, the fluoride release increased with increasing temperature and all glass ionomers showed greater recharging ability at higher temperatures. An increase in environmental temperature increased both the fluoride release and recharging of the glass ionomers. This may be important in developing regimes for improving the delivery of topical fluoride products.

INTRODUCTION

The release of fluoride in the oral environment has been widely investigated[1] in relation to the therapeutic effects of fluoride in oral diseases.[2-3] Many devices[4-5] and materials[6] have been designed for the release of fluoride. Among them, glass ionomer cements (GICs) are

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the most successful, in addition to their having been widely used in clinical dentistry.[7]

Glass ionomer cements have undergone great improvements since their invention in 1969.[8] Recently, it has been shown that GICs are active during the life of a restoration, and they can react with the oral environment in a unique way so that GICs and related materials have a strong potential to be smart materials in dentistry.[9] Among all the properties of GICs, fluoride release has been considered particularly important. The process of fluoride release from a GIC can be affected quantitatively by several experimental[10] and intrinsic variables.[11] The pattern of fluoride release for glass ionomers follows a high initial release rate followed by a rapid reduction in the rate of release over time.[12]

Fluoride release from GICs may be improved by recharging from a fluoride source applied in the oral environment that produces an increased level of fluoride.[13] Fluoride's recharging ability and re-release after recharging of GICs are extremely important factors for the long-term efficacy of the material.

As broad temperature fluctuations occur in the oral environment, thermal circuits may frequently challenge the restorative materials placed in this environment. Until now, the relationship between fluoride release and recharging and the environmental temperature has been unclear. This study aimed to investigate the effects of temperature on the pattern of fluoride release and the fluoride recharging ability of GICs. The hypothesis tested was that the environmental temperature may affect the fluoride release and recharging ability of GICs.

METHODS AND MATERIALS

The restorative materials used in this study include: a conventional luting glass-ionomer, Ketac Cem Maxicap (3M ESPE, Oberbay, Germany) (KC); two high viscosity restorative glass ionomers, Ketac Molar Aplicap (3M ESPE) (KM) and Fuji IX GP Fast (GC Corp, Tokyo, Japan) (F) and a restorative resin-modified glass ionomer, Fuji II LC Capsule (GC Corp) (FLC) (Table 1). All materials are capsulated glass ionomers.

Fifteen disk specimens of each material were prepared. Each material was handled according to the manufacturer's instructions. Capsules were activated and mixed mechanically for 10 seconds using a Silamat device (Vivadent, Schaan, Liechtenstein). The material was then placed in a mold (inner diameter of 15 mm, depth of 1 mm) mounted on a polymethylmethacrylate plate covered with a Melinex film (Toray, Tokyo, Japan) and compressed using a second plate covered with film. The auto-cured glass ionomers (KC, KM and F) were allowed to set before their removal from the mold. While in the mold, the light-cured material (FLC) was cured with a 40-second exposure of each flat surface to a visible light source (Visilux 2, 3M ESPE).

After setting, all specimens were removed from the molds and stored in 100% relative humidity at 37°C for 24 hours, after which they were ground using dry 1200-grit silicone carbide paper. The dimensions (diameter and thickness) of each specimen were measured (Mitutoyo Digimatic, RS Components, Japan), and the weight was recorded by digital balance (Mettler-Toledo, Greifensee, Switzerland).

The disc specimens were stored in cylindrical containers lying horizontally, so that all surfaces of the specimens were immersed in storage water while maintaining minimal contact with the container walls. Each container was filled with 5 ml of distilled and deionized water (pH=7.0±0.3), which was changed daily in the first week and every three days thereafter. Each group (n=5)[14] was stored at 4°C, 37°C or 55°C. The solutions containing released fluoride were collected at 1, 2, 3, 5, 7, 14 and 28 days. The collected fluoride in the contained solution was measured as follows: 0.5 ml acetic buffer solution (TISAB III, Thermo Electron, Waltham, MA, USA) was added to 5 ml of the fluoride-containing solution and mixed. The fluoride concentration was determined using an ion selective electrode (ISE) (Orion Research, Lumberton, NJ, USA) calibrated by fluoride standard solutions, which were diluted to 0.1, 1, 10 and 100 ppm from a fluoride standard solution (Thermo Electron).

Another three groups of specimens of each material were prepared as above and stored at 37°C for 30 days.

Table 1: *Materials Used in This Study and Their Composition*

Product	Type of Materials	Composition (manufacturers information)	Batch #
Ketac Cem Maxicap (KC)	Conventional luting glass ionomer	Powder: fluoroaluminosilicate glass. Liquid: aqueous polycarboxylic acid.	151394
Ketac Molar Alicap (KM)	High viscosity restorative glass ionomer	Powder: fluoroaluminosilicate glass. Liquid: aqueous polycarboxylic acid.	126978
Fuji IX (F)	High viscosity restorative glass ionomer	Powder: strontium fluoroaluminosilicate glass. Liquid: aqueous polycarboxylic acid.	0312241
Fuji II LC (FLC)	Resin-modified restorative glass ionomer	Powder: fluoroaluminosilicate glass. Liquid: aqueous solution of polycarboxylic acid, TEGDMA and HEMA	0401165

HEMA=Hydroxyethylmethacrylate, TEGDMA=Triethyleneglycol demethacrylate

The distilled and deionized water were changed periodically (daily in the first week, then every three days). Next, three groups were recharged at 4°C, 37°C and 55°C in 250 ppm sodium fluoride solution for five minutes. Fluoride re-release was recorded two days before and two days after the recharging using an ion selective electrode as described above.

The cumulative fluoride release (28 days) of each material at each temperature was compared using one-way ANOVA and Tukey's test at a significance level of 0.05. For each material, the 24-hour fluoride re-release after recharging at different temperatures was compared using one-way ANOVA and Tukey's test at a significance level of 0.05.

RESULTS

All the materials showed a cumulative fluoride release, which increased with time. Figures 1, 2 and 3 show fluoride release at 4°C, 37°C and 55°C at each test period from the four materials.

The cumulative fluoride release of 28 days at 4°C was ranked from high to low as KC> FLC> F> KM (Figure 1). The luting cement showed the greatest fluoride release among all the materials ($p<0.05$). The fluoride release of resin-modified glass ionomer was significantly higher than that of high viscosity glass ionomers ($p<0.05$).

Figure 2 shows the cumulative fluoride release for 28 days at 37°C from the five materials. The amount of fluoride release for all materials was significantly greater than that at 4°C ($p<0.05$). The luting glass ionomer cements again showed the greatest fluoride release, and the difference in fluoride release in resin-modified glass ionomer and high viscosity glass ionomers was not significant ($p>0.05$).

For the four materials, Figure 3 shows the cumulative fluoride release at 28 days for 55°C; the cumulative amount of fluoride released for all materials was significantly higher than that at 4°C and 37°C ($p<0.05$). The cumulative fluoride release at 28 days for 55°C was ranked from high to low as KC> FLC> F> KM, which is the same as 4°C and 37°C.

The fluoride release for the 30 days aged specimens from high to low could be ranked as KC> FLC> F> KM. The amount of fluoride re-release after recharging at different temperatures, which is shown in Figures 4, 5 and 6, followed the same sequence in all materials. For each material, the 24-hour fluoride re-release after recharging at 55°C was significantly greater than for 4°C and 37°C ($p<0.05$), as shown in Figure 7. The 24-hour fluoride re-release after recharging at 37°C was greater than that for 4°C; however, a significant difference was only observed in the high viscosity glass ionomers and resin-modified glass ionomer, not in the luting glass ionomers ($p<0.05$).

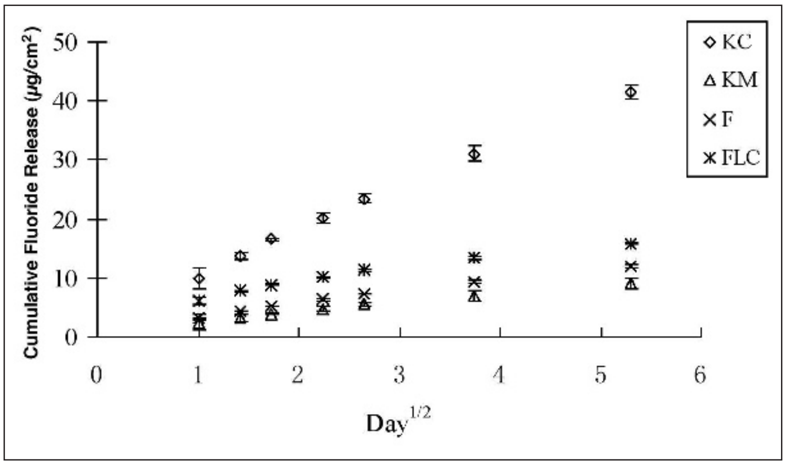

Figure 1: *The mean cumulative fluoride release (μg/cm²) (1-30 days) and its standard deviation at 4°C from all materials tested is plotted against the square root of time (days). The luting GIC (KC) showed the greatest fluoride release, while one of the high viscosity GICs (KM) showed the lowest fluoride release. The fluoride release from resin-modified GIC (FLC) was significantly greater than that of the high viscosity GICs. Note: Standard deviation bars are shown, but sometimes they are very small and bars often fall within the data symbols.*

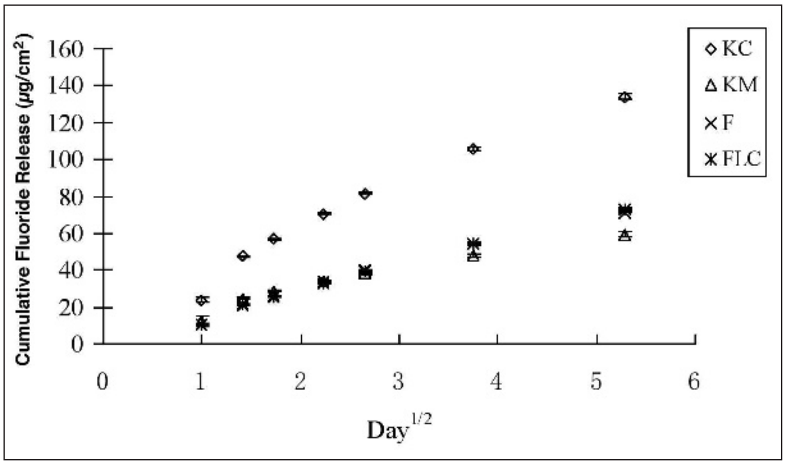

Figure 2: *The mean cumulative fluoride release (μg/cm²) (1- 30 days) and its standard deviation at 37°C from all materials tested is plotted against the square root of time (days). KC showed the greatest fluoride release and KM the lowest, as in Figure 1. Note: Standard deviation bars are shown, but sometimes they are very small and bars often fall within the data symbols.*

DISCUSSION

This study showed that the fluoride release and recharging of glass ionomers varies with storage temperatures. The amount of fluoride release was significantly affected by storing at 4°C, 37°C and 55°C ($p<0.05$). Central to the fluoride release of glass ionomers is their ability to support water diffusion.[15-16] Water diffusion through the material matrix drives fluoride ions to the surface, where they can be released into storage media. Therefore, the fluoride release of

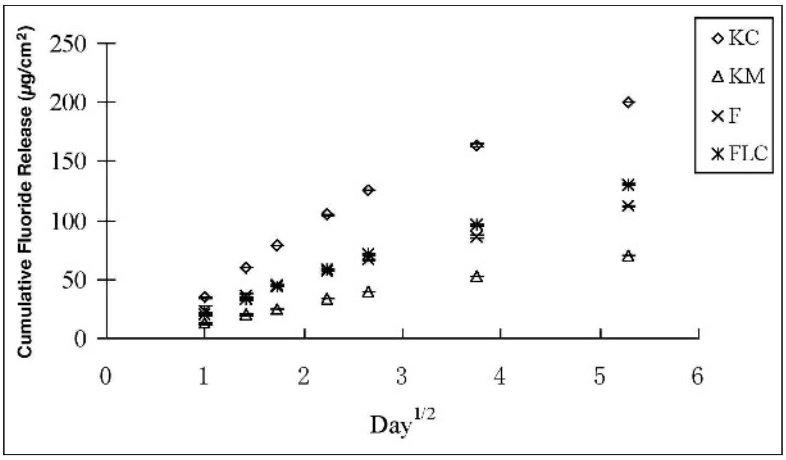

Figure 3: *The mean cumulative fluoride release (μg/cm²) (1- 30 days) and its standard deviation at 55°C from all materials tested is plotted against the square root of time (days). The amount of fluoride release was significantly higher than at 4°C and 37°C* (p<0.05)*, but the trends in ranking the materials were the same. Note: Standard deviation bars are shown but sometimes they are very small and bars often fall within the data symbols.*

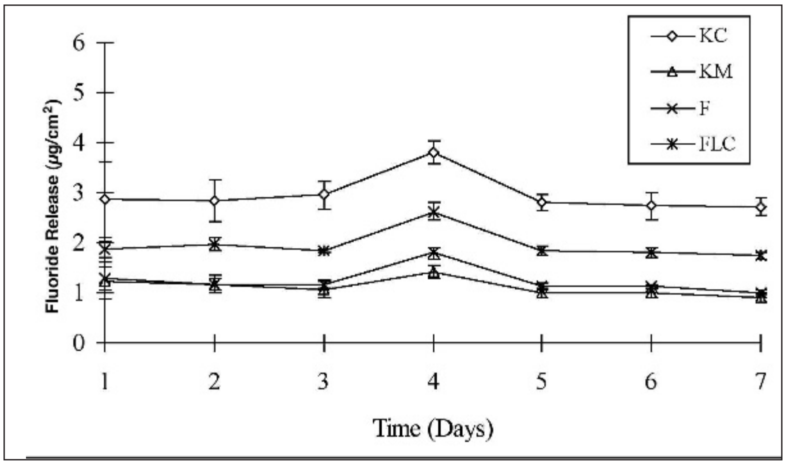

Figure 4: *Fluoride release (μg/cm²) before and after recharging at 4°C of all the materials after 30 days fluoride release. The amount of fluoride re-release after recharging followed the same trends in ranking as the fluoride release in Figure 1. Note: Standard deviation bars are shown, but sometimes they are very small and bars often fall within the data symbols.*

glass ionomers appears to be a water-diffusion controlled process, which has been reported by many authors; the fluoride release of glass ionomers was linearly proportional to $t^{1/2}$ within a relatively short time (a few days) after setting.[16]

In this study, the cumulative fluoride release of all glass ionomers and their related materials showed a good linear relationship, as it was plotted against the square root of time (days), which suggests that fluoride release under different temperatures is a diffusion controlled process. However, fluoride release significantly increased as the temperature increased.

The rate constants of most processes increase as the temperature is raised. This behavior is normally expressed mathematically by the Arrhenius equation:

$$\ln k = \ln A - \frac{E_a}{RT},$$

where k is the constant rate; A is the Arrhenius Factor; E_a is the activation energy and R is the universal gas constant.[17] This equation implies that the rate constant increases as the temperature is raised. Therefore, the diffusion behavior of the same material will change as the environmental temperature changes. As a water-diffusion controlled process, fluoride release from glass ionomers relies on the ionomers ability to support water diffusion through the matrix.

For the different materials tested under the same conditions, luting glass ionomers have always shown the highest fluoride release. The fluoride release of all glass ionomers aged after 30 days reached a relatively low level (1-3 μg/cm²) (see Figures 4, 5 and 6) before recharging. The daily fluoride release of different types of glass ionomers can be ranked as luting > resin-modified > high viscosity. After the application of high concentration sodium fluoride solution for five minutes, all specimens showed significantly greater 24-hour fluoride re-release ($p<0.05$). The amount of fluoride re-release after recharging at different temperatures for each material was still ranked in the same order. This may imply that glass ionomers with greater initial fluoride release have greater recharging ability. High concentration environmental fluoride solution may diffuse into the glass ionomers' matrix, and further ion exchange may occur between the cement matrix and the solutions diffused into that matrix.[18] When fluoride recharging is completed and the materials are exposed to a low fluoride concentration environment (distilled/deionized water), fluoride recharged into glass ionomers can be released by water diffusion. Therefore, fluoride release and re-release after recharging both may depend on water diffusion through the matrix.

In this study, 24-hour fluoride re-release of all specimens increased as the recharging temperature increased. The 24-hour fluoride re-release after recharging at 37°C was significantly greater in high viscosity glass ionomers and resin-modified glass ionomers than at 4°C ($p<0.05$). Twenty-four hour fluoride re-release after recharging at 55°C was significantly greater in all materials. Generally, glass ionomers released a greater amount of fluoride after they were recharged at higher temperature. The previous observation demonstrated that the fluoride release of glass ionomers was also greater at high temperatures, which confirms that diffusion coefficient increas-

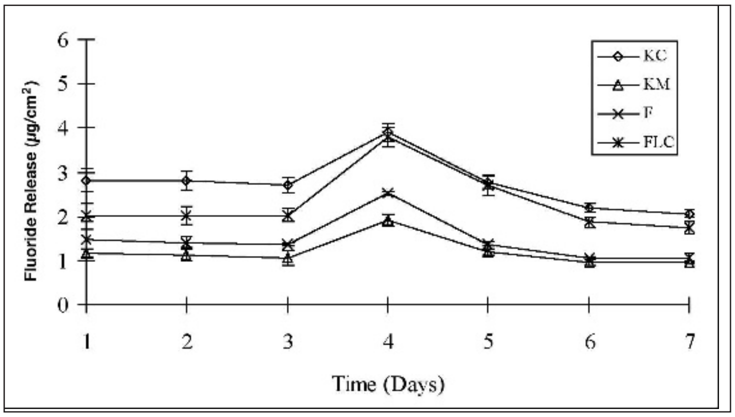

Figure 5: *Fluoride release (µg/cm²) before and after recharging at 37°C after 30 days fluoride release. Fluoride re-release followed the same trends as recharged at 4°C (Figure 1) but greater than that at 4°C. Note: Standard deviation bars are shown, but sometimes they are very small and bars often fall within the data symbols.*

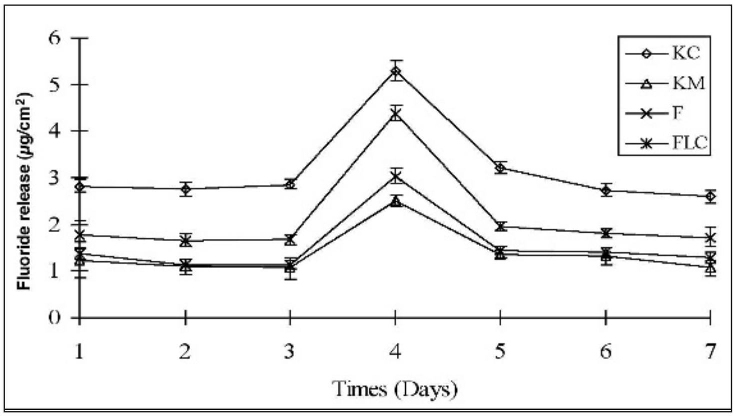

Figure 6: *Fluoride release (µg/cm²) before and after recharging at 55°C after 30 days fluoride release. For all materials, the 24 hours fluoride re-release after recharging at 55°C was significantly greater than at 4°C and 37°C. Note: Standard deviation bars are shown, but sometimes they are very small and bars often fall within the data symbols.*

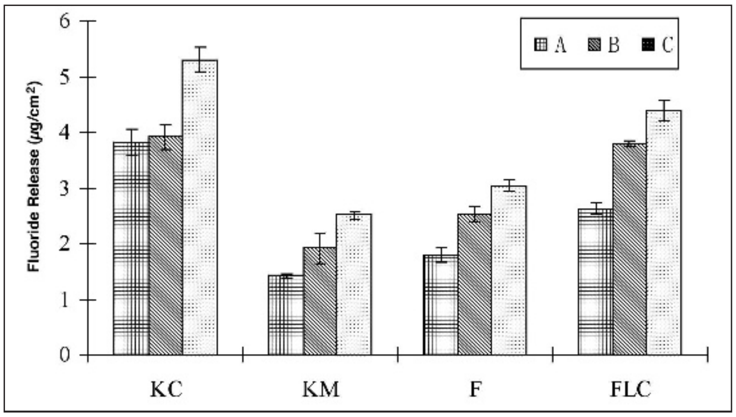

Figure 7: *Twenty-four hour fluoride re-release (µg/cm²) after recharging at 4°C (A), 37°C (B) and 55°C (C). For each material, the 24-hour fluoride re-release after recharging at 55°C was greater than at 4°C and 37°C but significant difference (p<0.05) was only observed in KM, F and FLC.*

es as the temperature is increased,[17] as is expected from the Arrhenius equation. In clinical practice, most topical fluoride treatment in patients with glass ionomers or related filling materials is achieved by daily tooth brushing and, occasionally, by mouth rinsing with fluoride solutions or by using gels.

CONCLUSIONS

Based upon results of this study, a higher temperature used during topical fluoride applications may increase fluoride recharging and re-release ability. A low oral environment temperature should be avoided during topical fluoride application.

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