

Fluoride release from different tooth colored restorative materials: An in vitro study

Abdul-Haq A SULIMAN*
Manal A AL-TAEE**

ABSTRACT

This in vitro study evaluated the amount of fluoride released from fluoride-containing materials over a period of (30) days. Twenty eight disk samples (2.5 mm depth x 10 mm diameter) were prepared and divided into four groups: Group I Vitremer glass ionomer, group II resin composite Tetric, group III resin composite Definite, and group IV Compoglass.

The samples were stored in an incubator at 37° C for (24) hours. Each disk was suspended in an individual plastic tube containing (3) ml of deionized water. The water was changed every (24) hours, fluoride release was determined at each day for (30) days, after buffering the solution with equal volume of TISAB. The fluoride release was measured with a fluoride-ion selective specific electrode previously calibrated from (0.05) to (100) ppm. Fluoride release was expressed in part per million (ppm).

ANOVA test was used to evaluate the data. The results revealed that Vitremer glass ionomer released significantly more fluoride (41 ppm) for the first day than all the other products. This was followed by Compoglass which exhibits more fluoride release than the other two types (32 ppm), the Tetric composite resin was the third material in the amount of fluoride released (3.75 ppm), and finally the resin composite Definite released fluoride in about (0.75 ppm). All fluoride release was decreased from day (1) to (30) observation period.

Key Words: Fluoride release, resin composite, glass ionomer.

*Abdul-Haq Abdul-Majeed SULIMAN; BDS, MS, MS, PhD : Prof. Department of Conservative Dentistry, College of Dentistry, University of Mosul, Mosul, IRAQ.

**Manal Abdul-Jabar AL-TAEE; BDS, MSc : Assistant Lecturer. Department of Pedodontics, Orthodontics, & Preventive Dentistry, College of Dentistry, University of Mosul, Mosul, IRAQ.

الخلاصة

أجريت هذه الدراسة خارج الجسم لتقييم كمية الفلورايد المتحررة من مواد حاوية على الفلورايد خلال فترة (٣٠) يوماً. تم تحضير عينة من (٢٨) قرص (٢,٥ ملم عرض x ١٠ ملم قطر)؛ وقسمت إلى أربع مجاميع: المجموعة الأولى (Vetrimer glass ionomer)، المجموعة الثانية (Resin composite Tetric)، المجموعة الثالثة (Resin composite Definite)، والمجموعة الرابعة (Compoglass). خزنت العينة في حاضنة بدرجة حرارة (٣٧°) مئوية لمدة (٢٤) ساعة وتم تعليق الأقراص في أنبوب بلاستيكي يحتوي على ماء منزوع الأيونات يتم تغييره كل (٢٤) ساعة. تم قياس كمية الفلورايد المتحررة يومياً ولمدة (٣٠) يوماً (بعد معادلتها بكمية مساوية من مادة TISAB) باستخدام قطب خاص بأيون الفلورايد تم تعبيره مسبقاً من (٠,٠٥) إلى (١٠٠) جزء من المليون. إن كمية الفلورايد المتحررة قيسبت بوحدة جزء من المليون.

تم استخدام اختبار (ANOVA) لتقييم البيانات احصائياً. أثبتت النتائج أن مادة (Vetrimer glass ionomer) تفرر الكمية الأكبر من الفلورايد (١ جزء من المليون) في اليوم الأول أكثر من المواد الأخرى، تليها مادة (Compoglass) والتي تفرر كمية (٣٢) جزء من المليون. جاءت مادة (Resin composite Tetric) بالمرتبة الثالثة (٣,٧٥ جزء من المليون)؛ وأخيراً مادة (Resin composite Definite) (٠,٧٥ جزء من المليون). إن كمية الفلورايد المتحررة تناقصت من اليوم الأول إلى اليوم الثلاثون خلال فترة البحث.

INTRODUCTION

Microleakage has been found to be responsible for recurrent caries. Clinically there is no dental material that can produce perfect enamel and dentin seal. Theoretically the continual release of fluoride ions from a restorative material could reduce or eliminate recurrent caries⁽¹⁾. Recurrent caries is one of the primary causes of failure of dental restorations, which lead to replacement of the dental restorations⁽²⁾. Therefore, the addition of fluoride to conventional dental material such as amalgam alloy, resin composite, and various cements has generally been attempted. Specific material containing fluoride has been recommended for the restoration of root surface caries that are more likely to show recurrent caries⁽³⁾. Although silver amalgam has been successfully used to restore carious root, resin composite, glass ionomer and resin modified glass ionomer are also used to restore root surfaces since they are esthetically more acceptable and bond to tooth structure. Fluoride release from these materials inhibits recurrent

caries in the restored surfaces. The dynamic movement of fluoride into and out of restorative materials and the role of fluoride releasing materials in the inhibition of recurrent caries in vivo and in vitro have been studied ⁽⁴⁾.

In this concept, glass ionomer has been found to be the best material for fluoride conduction into tooth substance ⁽⁵⁾. Glass ionomer cements (GIC) have been shown to be a very useful adjunct to restorative dentistry. Their major advantages including the ion exchange, adhesion to both enamel and dentin and continuing fluoride release through out the life of the restoration have been appreciated ⁽⁶⁾. Also the uptake of fluoride by dentin and enamel walls in contact with glass ionomer restorations has been demonstrated ^(7,8).

The anticariogenicity of glass ionomer seems to be due to fluoride flux centered into the component of the glass powder that leaks from this material and reacts with the surrounding tooth structure to decreasing its solubility ⁽⁹⁾.

Also the fluoride releasing glass ionomer has been reported to be bactericidal in vitro. This may be beneficial to modern dentistry, which is directed to the preservation of tooth tissue during restorative treatment, but little is known about its in vivo behavior ⁽¹⁰⁾.

The purpose of this study is to determine and evaluate the amount of the fluoride released from different tooth colored restorative materials.

MATERIALS AND METHODS

The materials used in this study are shown in table (1).

Table (1): Materials used in the study

No.	Material	Manufactures	Batch No.
1	Vitremer core build up restorative system	3M, Dental product, ST. Paul, MN 55144 USA	3303L
2	Compoglass F	Vivadent, Schan-Liechtenstein FL-9494	B10384
3	Tetric Ceram	Vivadent, Schan-Liechtenstein FL-9494	B36278
4	Definite	Degussa-Hulls AH	039817

Twenty-eight disk samples were prepared and divided into four groups, (7) disks for each group. Group I Vitremer glass ionomer, group II resin composite Tetric ceram, group III resin composite Definite and group IV Compoglass F. The samples were prepared by pouring the materials into a stainless steel mold with the 2.5mm depth and 10 mm diameter.

Then a two glass slides used to compress the materials, during this a stainless steel wire was incorporated into each disk sample to be served as an attachment. All materials were then light cured by a curing gun (Dentsply Equipment Division Dentsply International Inc USA). The curing was applied in four perpendicular points for each surface, and for (40) seconds each side.

After curing, the disk samples were removed from their molds, and suspended into a polyethylene test tubes which contain 3() ml deionized water.

Sample were placed in an incubator at (37°) for the first (24) hour, and after this (24) hour, the samples were removed from the incubator, and each sample was removed from it's tube, and placed into a new tube contain (3) ml of deionized water and returned to the incubator.

The previous tubes which contain the deionized water into which fluoride was released were buffered with similar amount of TISAB (3 ml), then a fluoride -ion- specific electrode F1052f Radiometer A/S (Emdrupvej 72 DK 2400 Copenhagen NV Denmark) was used to measure the released fluoride. This procedure was repeated every day and for (30) days.

The measuring unit is by Milivolts (Mv). Then the reading from the device was pointed to the standard solution curve according to the calibration curve method⁽¹¹⁾ the reading was determined by the use of the non liner equation (Exponential e^x) to obtain the concentration of the fluoride in part per million (ppm) .

Statistical Analysis

The means have been tested for their significant by using (ANOVA) test, Duncan Multiple Range Test to compare the amount of fluoride released between each type of the restorative materials. The differences were considered to be significant when the probability was equal or less than 5% level $p \leq 0.05$.

RESULTS

The amount of fluoride ion released from the tested materials in ppm for the observation period is shown in table (2). The amount of fluoride released from the glass ionomer materials is shown graphically in figures (1) and (2) shows that released from resin composite materials.

Table (2): Amount of fluoride released in part per million (ppm)

Days	Definite	Vitremer	Compoglass	Tetric Ceram
1	0.75	41	32	3.75
2	0.375	30	29	3.5
3	0.475	18	34	0.8
4	0.375	15	28	0.75
5	0.225	18	25	0.2
6	0.225	18	17.5	0.4
7	0.225	15	8	0.7
8	0.225	15	7.5	0.7
9	0.46	15	9.9	0.7
10	0.225	7.75	9.9	0.4
11	0.15	7.75	9	0.23
12	0.1	7.75	9	0.099
13	0.1	7.75	5	0.23
14	0.1	7.75	5	0.23
15	0.1	8	4.9	0.195
16	0.1	8	3	0.1
17	0.1	8	3	0.1
18	0.1	8	3	0.37
19	0.1	14	5	0.37
20	0.1	14	5	0.37
21	0.1	8	4.5	0.225
22	0.1	8	3.5	0.09
23	0.08	7.4	0.4	0.09
24	0.08	7.4	0.7	0.08
25	0.08	7.4	0.4	0.08
26	0.08	7.4	0.4	0.08
27	0.08	7.4	0.38	0.025
28	0.08	7.4	0.38	0.025
29	0.08	7.4	0.38	0.025
30	0.08	7.4	0.38	0.025

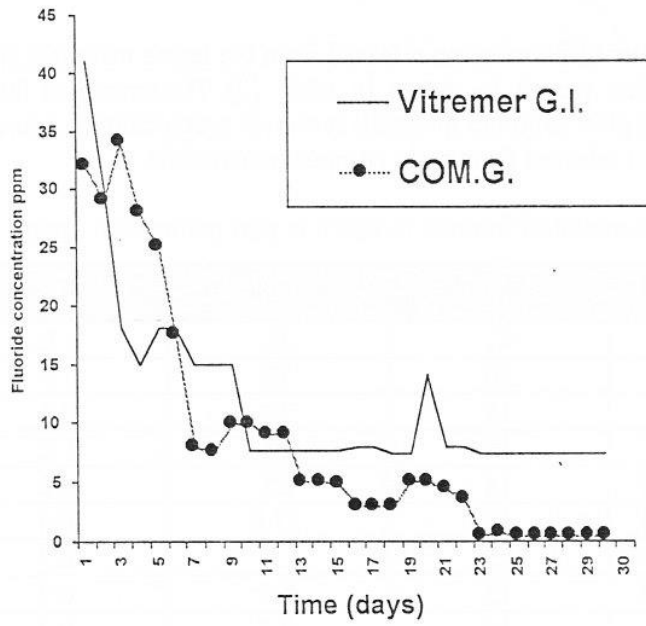


Figure (1): Fluoride release from Vitremer core build up system (3M) and Compoglass (Vivadent) the time in days

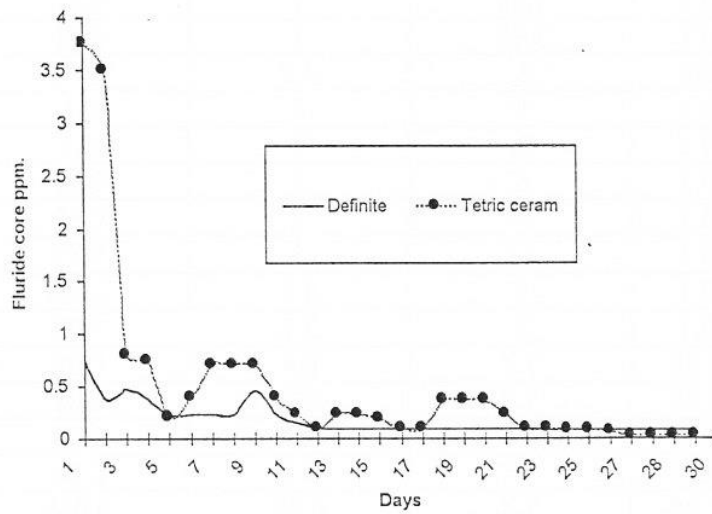


Figure (2): Fluoride release for resin composite Definite and Tetric Ceram (the time in days)

For the glass ionomer material the maximum fluoride release was observed in early days and decreased gradually to reached a constant level. The greatest amount of fluoride released was found with Vitremer glass ionomer followed by Compoglass F.

For Vitremer glass ionomer the amount of the fluoride release in the first day was very high (41 ppm). Gradually the amount of fluoride release was diminished until it reaches (7.4) in day (23) and remain constant at that level till the end of the experiment.

For Compoglass the amount of fluoride released in the first day was (32 ppm), in the next day was (29 ppm). While in the third day increased to (34 ppm). It decreased sharply to reach (8 ppm) in the seventh day, then gradually the amount of fluoride released was diminished slowly until reached (3.5 ppm) in day (22), then it reached (0.38 ppm) till the end of the experiment.

The fluoride released from the composites is presented in figure (2). For both composites the maximum fluoride released was observed on the first day the amount of fluoride released decreased sharply during the following two days, and then gradually reached a constant level. The greatest amount of fluoride released was found with Tetric ceram followed by Definite which showed the least amount of fluoride released during all time of experiment.

Fluoride released from resin composite Definite was very low as compared to glass ionomer material used. The amount of fluoride released in the first day was (0.75 ppm). In the second day there is a sharp decrease (0.375 ppm) while in the seventh day it reached (0.22 ppm), then gradually diminished until it reached (0.08 ppm) in day (23) and remain constant till the end of the experiment. The resin composite Definite released fluoride lesser than all other materials throughout the period of this experiment.

Fluoride release from Tetric ceram in the first day was (3.75 ppm). In the next day (3.5 ppm), while in the seventh day decreased to reach (0.7 ppm), in the twentieth day (0.37 ppm) then it decreased until it reached (0.025 ppm) in day twenty-seventh and remain constant till the end of the experiment.

Table (3) shows the mean of fluoride released per week for each materials during the experimental period and also determine the significant difference between the same material at different weeks, and between the different materials at the same week using Duncan Multiple Range test.

Table (3): The weekly mean of fluoride release (ppm) for all types of materials

Types of Material	Time in Week			
	1 st Week	2 nd Week	3 rd Week	4 th Week
Compo-glass F	24.78 (a.A)	7.525 (b.B)	3.857 (b.B)	0.4275 (b.C)
Vitremer G.I	22.142 (a.A)	9.5938 (a.B)	9.5434 (a.B)	7.4 (a.B)
Tetric ceram	1.525 (b.A)	0.2977 (c.B)	0.1244 (c.B)	0.04 (c.C)
Definite	0.3706 (b.A)	0.1219 (c.A)	0.01(c.B)	0.08 (c.B)

*Capital letter compare between the same material of different weeks.

Small letter compare between different materials at the same week.

Means with the same letters are statistically not significant.

The study revealed that the mean fluoride released by Compoglass F and Vitremer in the first three weeks was significantly higher than Tetric ceram and Definite, while in the fourth week the Vitremer material released significantly higher amount of fluoride compared with the other three materials.

The finding also show that the amount of fluoride release by each materials in the first week was significantly higher than the rest of the time intervals. The Compoglass F material reported high significant fluoride released in the second week compared with fourth week, while the other three materials their were decreased in amount of fluoride released but there were no significant differences in the mean of the fluoride released per week between second, third and fourth weeks.

The minimum and maximum amount of fluoride released for different materials, and the mean and standard deviation of amount of fluoride released during the experimental period is shown in table (4)

Table (4): Minimum, maximum, means and standard deviation of fluoride released (ppm) for materials tested during (30) days

Type of Materials	No.	Min.	Max.	Mean	SD
Vitremer G.I.	30	7.4	41	11.925	7.617
Compoglass F	30	0.38	34.00	8.8040	10.2961
Tetric ceram	30	0.025	3.72	0.5060	0.8812
Definite	30	0.8	0.75	0.1811	0.1584

The Vitremer G.I reported the high mean of amount of fluoride released (11.29 ppm) followed by Compoglass (8.80 ppm), while the Tetric ceram and Definite reported low amount (0.50 ppm and 0.18 ppm) respectively.

The ANOVA and Duncan's Test was used to determine the differences in the mean amount of fluoride released from the different restorative materials for the period of one month (Tables 5 and 6).

Table (5): ANOVA test result for the mean fluoride release for the four materials during (30) days

	Sum of Squares	Df	Mean Square	F cal.	F tab.
Between Groups	3159.916	3			
Within Groups	4780.254	116	1053.305	25.5	8.5494
Total	7940.170	119			

Table (6): A comparison of mean fluoride release between materials tested for the period of one month

Types of Materials	Duncan's Test		
	Time (days)	Mean Difference	Letter of Groups
Compo-glass	30	8.8040	a
Vitremer G.I	30	11.9250	a
Tetric ceram	30	0.5000	b
Definite	30	0.1810	b

*Groups with the same letter are not significantly different.

From these statistical analysis it was observed that there were highly significant different between the materials. The G.I materials released high amount of fluoride compared with resin composite and these difference were highly significant, while there was no significant difference between G.I materials (Compoglass F and Vitremer G.I) and there was no significant difference between resin composite materials (Definite and Tetric ceram).

DISCUSSION

In this study, four types of commercial restorative materials had been tested to evaluate the amount of fluoride release in short term (30 days) and to compare between the ability of each material to release fluoride in artificial environment (in vitro) through the use of deionized water as an incubation media with incubation temperature of (37°C).

The materials used in this study were four types of restorative materials in which fluoride is incorporated with their composition two of which were glass-ionomer, (Vitremer and Compoglass) and the other two where a resin composite (Tetric ceram and Definite).

All materials chosen for the study were light cured in order to eliminate the effect of weight and volume change as in self-curing glass ionomer materials. Such effect which was mentioned by others^(12,13).

The result of study revealed that all measurements of fluoride in the four types of the materials were higher in the 1st and 2nd days, then it was decreased spontaneously till the last week of experiment and remain constant. The initial release by all materials was highest during the first (24) hours and decreased sharply this is in agreement with other studies^(14,15). Vitremer material produced a significantly higher fluoride than the other materials in the first day (41 ppm), followed by Compoglass (32 ppm), while the Tetric ceram resin composite and the Definite resin composite have a very low amount of fluoride release (3.75) ppm and (0.75) ppm respectively. Within a few days their were slightly decreased in fluoride released and then there were sharp decreased in fluoride released for each materials reached a fairly constant level, that mean the pattern of fluoride release was similar for all materials. However, the amount of fluoride release was significantly greater for G.I.C materials at each measurement intervals.

This results was in agreement with that of Kawai *et al.*⁽³⁾, Swift⁽⁵⁾, and Aurjo *et al.*⁽¹⁵⁾, in which the pattern of fluoride release was similar for all materials studied, being greatest at the beginning of the experiment and gradually diminishing to a much lower and fairly constant level.

During the measurement of fluoride release from the Compoglass materials, a period of increasing in the fluoride released was obtained following obvious decreased measurements as shown in figure (1). This is attributed to the effect of resinous material incorporated in the material that may affect the release of fluoride ions by delaying the dissociation and release of these ions. The increase and decrease of fluoride ion released from the glass ionomer may be due to the composition of the glass ionomer, which consists of different salts of fluoride which may differ in their solubility and cause these changes in releasing of fluoride every day along the time of the experiment^(16, 17). This pattern of fluoride release from glass ionomer in this study is also in agreement with Tay and Brawen⁽¹⁸⁾, it was observed from the result of the study that the GIC (Vitremer) has very high fluoride released at the last day of experiment (7.4 ppm) than the other material used and this amount was even very high than the initial amount of fluoride released in the first day of composite materials (Tetric ceram and Definite). Also Compoglass F reported released high amount of fluoride at the last day (0.38 ppm) compared with the composite materials (Tetric ceram and Definite, 0.025 ppm and 0.08 ppm respectively).

The study reported the mean fluoride released per week for each material during the experimental period (table 3). The findings show that the mean fluoride release by Compoglass F (24.78 ppm) and Vitremer (22.14 ppm) in the first week was very significantly higher than of Tetric ceram (1.53 ppm) and Definite (0.37 ppm). In the first week there were no significant difference between Compoglass and Vitremer, while in the second, third, and fourth weeks the Vitremer released a higher amount of fluoride than Compoglass F.

The composite materials reported no significant difference between them (Tetric ceram and Definite) in the (4) weeks of experiment, but released significantly low amount of fluoride compared with (G.I.C) materials.

The amount of fluoride released for each material at different week interval table (3). The study revealed that the amount of fluoride released by each material in the 1st is significantly higher than the rest of the time intervals. The Compoglass F material recorded high significant fluoride released in the second week compared with fourth week, while the other three materials there was decreased in amount of fluoride released. However there were no significant differences in the mean of fluoride released per week between second, third and fourth week intervals.

The mean amount of fluoride released by each material for (30) days (experimental period) is shown in table (6). The study indicate that the (G.I.C) material released fluoride (20-50) times more than the composite

materials, this finding is in agreement with the study carried by Arneds and Robin ⁽¹⁹⁾ and Arends *et al.* ⁽²⁰⁾. They show that approximately (10-50) times more fluoride released by (G.I.C) than by fluoride releasing composite.

The study show that the Vitremer release high amount of fluoride. (11.92 ppm) compared with the other materials, followed by Compoglass F (8.8 ppm). However, there were no significant differences between Vitremer and Compoglass in the mean of fluoride released for (30) days, while these materials were very high significant difference than composite materials.

The composite materials released very small amount of fluoride and there were a quick diminish in the amount of F. released this was in accordance with other studies ^(3, 18-21).

This result also in agreement with study carried out by Dijkman and Arend ⁽²²⁾ who explained this sharp decreased in fluoride released from the composite and polyacid modified resin occurred possibly because their were two process involved: a rapid surface elution followed by a slower continues bulk diffusion of fluoride ion. Recently, investigation have been made to incorporate a micro encapsulated fluoride salts into the composite material in order to maintain a gradual and continuous slowly release of fluoride ions through a continuous process of fluoride ion diffusion from these very slowly dissolute salts of fluoride including yttrium fluoride (YF₃) and ytterbium fluoride (YbF₃) these salts have been found to be very slowly effected with water and very slowly dissolute ^(23, 24).

CONCLUSIONS

1. From this study it was concluded that the amount of fluoride release was significantly high for the glass ionomer (Vetremer glass ionomer and Compoglass) than the composite resin (Definite and Tetric ceram).
2. Greater amounts of fluoride were released during the 1st and 2nd days for all the types of the materials used, than the rest of the observation time (30 days).
3. The fluoride was released from all the materials till the last day of the experiment (30th day).
4. The irregularities in the amount of the fluoride released (an increase in the level of fluoride following a decrease) was explained by the content of each type of the materials used which is slowly dissociated to released fluoride indifferent levels.

REFERENCES

1. Trimpeneers LM, Dermaut LR. A clinical evaluation of the effectiveness of a fluoride-releasing visible light-activated bonding system to reduce demineralization around orthodontic brackets. *Am J Orthod Dentofacial Orthop.* 1996; 22: 110-118.
2. Donly KJ, Darke DR, Wefel JS. Effects of aged fluoride -containing restorative materials on recurrent caries. *J Dent Res.* 1998; 77: 418-425.
3. Kawai K, Tantbiroj D, Kamalawat AS, Hasegawat, Refiel DH. In vitro enamel and cementum fluoride uptake from three fluoride-containing composites. *Caries Res.* 1998; 32: 463-469.
4. Burgess JO. Dental materials for the restoration of root surface caries. *J Dent Res.* 1995; 74: 342-351.
5. Swift EJ. Caries-inhibitory properties in vitro of a silver glass ionomer. Ph.D Thesis submitted to the University of Iowa. USA. 1989.
6. Mount GJ. Clinical performance of glass -ionomers. *Compend Contin Educ Dent.* 1998; 10: 981-986.
7. Mesenberg JH, Hals E. Influence of glass ionomer /Silver cermet restorations on interproximal bacterial growth. *J Dent Res.* 1980; 60: 670-680.
8. Hicks MJ, Flait ZCM, Silverstone LM. Secondary caries formation in vitro around glass ionomer restorations. *Quintessence Int.* 1986; 17: 527-532.
9. Swartz ML, Phillips RW, Clarke HE. Long-term fluoride release from glass-ionomer cements. *J Dent Res.* 1984; 63: 158-160.
10. Kreulen CM, De-Soet JJ, Weerheijm KL, Van-Amerongen WE. In vivo cariostatic effect of resin modified glass ionomer cement and amalgam on dentine, *ASDC J Dent Child.* 1997; 64: 131-135.
11. Martin S, Frants AF, James WRJ. Use of total ionic strength adjustment buffer determination of fluoride in water supplies. *J Anal Chem.* 1968; 7: 1169-1172.
12. Wandera A, Spencer P, Bohaty B. In vitro comparative fluoride release, and weight and volume change in light-curing and self-curing glass ionomer materials. *Pediatr Dent.* 1996; 18: 210-214.
13. Miller BH, Komatsuh H, Nakajima H, Okabe T. Effect of glass ionomer manipulation on early fluoride release. *J Am Dent Assoc.* 1995; 8: 182-186.
14. Arnold AM, Holmes DC, Wistrom DW, Swift EJ. Short-term fluoride release/uptake of glass ionomer restoratives. *J Dent Mater.* 1995; 11: 96-101.
15. Araujo BF, Gracia-Gody GF, Cury AJ. Fluoride releasing from fluoride -containing materials. *Oper Dent.* 1996; 21: 185-190.

16. Mount GJ. Glass ionomers: A review of their current status. *Oper Dent.* 1999; 24: 115-124.
17. Tate W H, Porter KH, Dosch RG. Successful photo curing dental restoration. *Oper Dent.* 1999; 24: 109-114.
18. Tay WM, Brawen M. Fluoride ion diffusion from poly alkenoate (glass ionomer) cement. *Biomaterials.* 1988; 9: 454-456.
19. Arends J, Robin J. Fluoride release from a composite resin. *Quintessence Int.* 1988; 19: 513-514.
20. Arends J, Dijkman GE, Dijkman AG. Review of fluoride release and secondary caries production by fluoridating composites. *Adv Dent Res.* 1995; 9: 367-376.
21. Ten Cate JM, Van Duinen RNB. Hypermineralization of dentinal lesions adjacent to glass ionomer cement restorations. *J Dent Res.* 1995; 74: 1266-1271.
22. Dijkman GEHM, Arends J. Fluoride release from composites a correlation with in situ data. *J Dent Res.* 1994; 74: 950-971.
23. Chan DCN, Swift EJ, Bishara SE. In vitro evaluation of fluoride-releasing orthodontic resin. *J Dent Res.* 1990; 69: 1576-1579.
24. Donly KJ, Gomez C. In vitro demineralization of enamel caries at restoration margins utilizing fluoride releasing composite resin. *Quintessence Int.* 1994; 25: 355-358.