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Research article

WATER ADSORPTION CHARACTERISTICS OF NEW DENTAL COMPOSITES

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ABSTRACT

Water sorption of dental composites affects dimensional stability, mechanical properties and bonding strength with tooth structures. The diffusion coefficient of water through the resin should be identified. **Methods:** Ten new composites fillings (M1-M10) were prepared from new Fluoroaluminosilicate powder composition and BisGMA/TEGDMA together with the related compounds such as tri ethylene glycol dimethacrylate, N,N-Dimethyl amino ethyl methacrylate and Camphorquinone. Five disk shapes were prepared for each composite using a stainless steel mold 15 mm in inner diameter and 1 mm in thickness, according to ISO 4049, the curing of each composite disk for 40 sec. Each disk was immersed separately in water for 90 day all at (37 ±1). Water sorption and solubility were calculated by using these measurements, Diffusion coefficients were also measured with the solution of Fick's second law. **Results:** The water sorption (g/mm^3) after 90 day immersion ranged from 14.98 g/mm^3 (±0.90) for M10 to 36.81g/mm^3 (±0.46) for M6. The solubility ranged from 3.3 g/mm^3 (±0.90) for M6 to 8.55 g/mm^3 (±0.31) for M7, the equilibration time for water sorption was reached at 20 day. M6 had the highest diffusion coefficient 6.25 $\times10^{-9}$ cm^2/s (±3.46). **Conclusion:** This investigation revealed that M6 composite filling was the best one due to the lowest water solubility while the other investigated fillings showed moderate to high solubility values but all are in accordance with the International Standard ISO 4049.

Keywords: Water sorption, Solubility, Composites, Diffusion coefficient, Calcium Fluoroaluminosilicate.

INTRODUCTION

Materials left for long time in the oral environment will undergo an interaction with oral fluids**.** Visible light-curable polymeric composites are now routinely used as filling materials for dental restorations. These materials are based on polydimethacrylate matrix resins along with silane-coated inorganic fillers. They possess many advantages such as mechanical properties comparable to commercial dental amalgams and dental ceramics, excellent esthetic quality and the ability to bond to enamel surface. However, in aqueous environment they absorb water and release unreacted components.

There are two different mechanisms that occur when the previously mentioned dental restorative materials are exposed to or stored in water: the first is gaining weight from water uptake, and the second is losing weight from dissolution in water[1].

Water sorption has been studied in several glassy polymers used in dentistry. Composite resins[2,3], soft lining and poly(methyl methacrylate) denture bases[4] have all been shown to absorb water and, at the early stages, this sorption follows Fick's law of diffusion. Studies have mostly been focused on determining the water sorption characteristics of epoxy-based polymers [5-7]. However, data are scanty on the resins that are employed as adhesives for bonding to hydrated dentin.

The importance of composite-water interaction has been acknowledged in the ISO standard 4049 which states that the maximum values for water sorption and concurrent solubility for resin-based materials (composites and cements). In order to comply with this ISO standard, resin-based materials must have water sorption and solubility values equal or lower than 40 micrograms per cubic millimeter (sorption) and 7.5 micrograms per cubic millimeter (solubility) for specimens 15 mm in diameter and 1 mm thick[8].

Fluoride (F^-) releasing restoratives are frequently studied because the F^- ions could increase the dissolution resistance of the tooth structure, enhance remineralization and hinder demineralization[9,10]. Efforts have been made to develop a composite consist of an aluminosilicate glass matrix modified with other elements, and they contain large quantities of fluorine. Calcium Fluoroaluminosilicate glass powder is treated with a fluoride in an amount of from 0.01 to 5 parts by weight based on 100 parts by weight of the glass powder, The Calcium Fluoroaluminosilicate glass powder of the investigation is improved in not only physical properties such as crushing strength but also mixing workability without impairing the inherent characteristics thereof for the dental use[11]. So Calcium Fluoroaluminosilicate glass will be suitable as filler for resin-based dental composites because it is interact with the bone structure makes them useful materials for bone replacement in implants, naturally radiopaque and highly resistant to moisture.

The aim of the present study is to determine the water sorption characteristics of light-cured resins made from new composites of Calcium Fluoro aluminosilicate glasses filler with various weight ratios.

MARERIAL AND METHODS

The compositions of the 10 composite resins tested, The ethoxylated bisphenol A glycol dimethacrylate Bis_GMA was purchased from Sigma Aldrich (UK) and TEGDMA (triethylene glycol dimethacrylate) manufactured from Sigma Aldrich (UK), *N*,*N*-Dimethyl aminoethyl methacrylate (DMAEMA) and camphor Quinone (CQ) were purchased from Aldrich (UK), Those materials were used to prepare the monomer phase.

Calcium fluoroaluminosilicate glass was synthesized and sintered in our laboratory [12]. It was ball-milled and sieved to powder with a particle size < 25 μm. The particle size distribution was measured using a BET analysis (CHEMBET 3000 QUANTA CHROME). The average particle size was 2.64 μm. This Calcium fluoroaluminosilicate glasses was treated with γ -methacryloxypropyltrimethoxy-silane (γ-MPS) known as A-174 which was supplied by Sigma Aldrich (UK).

Preparation of composite: Ten types composites formulations,containing the resins BisGMA/TEGD MA in a w/w ratio of 70/30 as the base resins. Resins were activated for visible light polymerization by CQ (0.5 wt %) and DMPT (0.5 wt %). Matrix resins were loaded (76 wt% ~ 60% Vol) and were then silanized. This Calcium Fluoroaluminosilicate glasses hand mixing. The compositions of the studied dental composites are shown in Table 1. Water absorption was determined on disc specimens, 15 mm diameter and 1 mm thick, for up to 90 days using the method outlined in ISO 4049. The discs were prepared between glass plates and were cured by exposure to dental curing lamp for 40sec on each side. Samples were measured, weighed and placed in individual sealed containers of water at 37ºC. The specimens were removed from the storage water at regular intervals, blotted dry and re-weighed. After 90 days specimens were placed in a desiccator containing dry silica gel and re-weighed at regular intervals over a period of 2 weeks.

Table1: Composition (W%) of Calcium Fluoro-aluminosilicate Glass

Glass	SiO_2	Al_2O_3	CaF_2	Al_2PO_4	AlF_3	NaF
M1	22	18		22	15	23
M2	22	19	10	39	13	7
M3	29	16.6	34.2	9.9	5.3	5
M4	35	25	20	8	6	6
M5	39.52	23.6	13.65	3.62	9.7	9.91
M6	24.3	27.5	14.0	19.1	15.1	
M7	33.9	17.5	8	15	10	15.6
M8	56.5	33.5				10
M9	48.9	29.1	15			7
M10	36.3	22	12	9	14.3	6.4

Preparation of specimens: Water sorption and solubility tests were determined according to the specification standard for composite (ISO 4049: 2000). Specimen discs approximately 15±0.2 mm in diameter and 1±0.1 mm in thickness were fabricated in an aluminum mold between two glass slides they were irradiated for 40sec on each side using a quartz–tungsten–halogen light-curing unit (Optilux 500, Demetron Research Corporation, Danbury, CT, USA). The light-curing unit had an exit-window diameter of 8 mm and was operated at 400 mW/cm^2 with the curing tip placed 1 mm from the glass plate. Four specimen discs were prepared for each for the ten experimental resin formulations. The thickness of the samples was measured accurately at three points using a micrometer. Also their diameters were measured, and their volumes were then calculated in mm^3.

water sorption and solubility: All the specimens were placed in a desiccator and transferred in a preconditioning oven at 37ºC. After 24 hrs they were removed, stored in the desiccator for 1 hr and weighted to an accuracy of 0.0001 g using a KERN 770 Germany. This cycle was repeated until a constant mass (m_0) was obtained. Following, the discs were immersed in distilled water at 37ºC. At fixed time intervals they were removed, blotted dry to remove excess water, weighted and then back to the water. The time intervals were more during the first four day, preceding daily as the uptake slowed at more extended intervals. The uptake of water was recorded until there was no significant change in weight, i.e. equilibrium was attained. This took about 30-40 days.

Sorption, desorption, and material net loss percentages were calculated as follows:

$$WA = \frac{M_1 - M_2}{V} \quad (1)$$

$$WS = \frac{M_0 - M_2}{V} \quad (2)$$

Where M_1 is the weight of the sample after immersion and M_2 is the original weight of the sample before immersion.

Diffusion coefficients: According to the Fick's Law, the equation for diffusion in three dimension, when the diffusion coefficient D is constant, is expressed as

$$\frac{\partial c}{\partial t} = D\left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} + \frac{\partial^2 c}{\partial z^2}\right)$$

Here, x (m), y (m), z (m) is the coordinates, c (%) is the concentration where of the diffusing species at time t (s) is the time, and D (m/s) is the diffusion coefficient. For the one-dimensional model of linear flow of mass in the solid bounded by two parallel planes, the differential equation is expressed as follows:

$$\frac{\partial c}{\partial t} = D\left(\frac{\partial^2 c}{\partial x^2}\right) \quad (3)$$

The differential equation is solved for the region $-h<x<h$ with zero initial concentration of water and with surfaces $x=\pm h$ kept at constant concentration c_0 for $t > 0$: It should be noted here that the solution to the Fick's second law (Eq. (3)) might alternatively be expressed as

$$\frac{M_t}{M_\infty} = \frac{4}{l}\,(Dt)^{1/2}\left\{\pi^{-1/2} + 2\sum_{n=1}^{\infty}(-1)^n ierfc\left(\frac{n\,l}{2(Dt)^{1/2}}\right)\right\}$$

$$\frac{M_t}{M_e} = 2\left(\frac{D\,t}{\pi\,l^2}\right)^{1/2}, \quad (4)$$

where M_t was the mass uptake (g) at time t (s), M_e was the mass uptake (g) at equilibrium, l was the specimen thickness (cm) such that D is the diffusion coefficient (cm^{2}s^{-1}) calculated from the gradient of Mt/Me against $t1/2$. If the uptake M_t is measured at convenient intervals of time until equilibrium is reached, then a plot of M_t/M_∞ against $t^{1/2}$ should provide a straight line for the earlier stages with the slope, S

$$S = 2\left(\frac{D}{\pi l^2}\right)^{1/2}, \quad (4)$$

For which

$$D = S^2 \pi l^2/4$$

RESULTS

All of the studied composite resins increased in weight during immersion in water. The means of the percentage values for sorption, solubility and diffusion coefficient of the ten different types of Calcium Fluoroaluminosilicate materials were illustrated in Figure 1. During sorption, M9 and M10 showed significantly lower and no significant differences among them respectively, While M6, M5, M2 and M1 showed the highest sorption, which is lower than those required by ISO 4049 standard, 40µ g/mm^3. Water solubility also showed differences within the studied groups. Four main groups of fillings can be classified: M9, M10, showed moderate solubility while M4 showed less than the above two

M1, M3, M8 were the filling materials with the highest mean water solubility value .Finally M6 was found the best filling.

For all studied composites, the equilibration time for water uptake was of the order of 15-20 days depending on material and the volume of specimens. The rate of change in weight over the selected time intervals is presented in Figure 2 for all studied composites. Figure 3 the plots of *Mt/Me* versus square root of time, it can be seen that the plots are almost linear for M10 composites. The plots had linear increase in earlier stage and became balanced at the end of the process when the composites were completely saturated, that mean no more water can be absorbed or desorbed by the studied composites. The diffusion coefficients controlled process was confirmed by the linear part. Figure 4 shows the diffusion coefficients of the studied resins ranged between 0.87 and 6.2 $\times 10^{-10} cm^2/sec$ for M10 and M6 respectively.

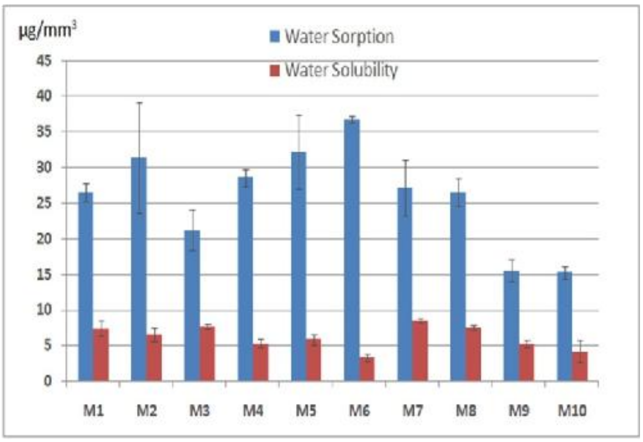

Fig. 1: Mean water sorption (µg/mm^3) and water solubility obtained for composite materials (teeth filling) tested.

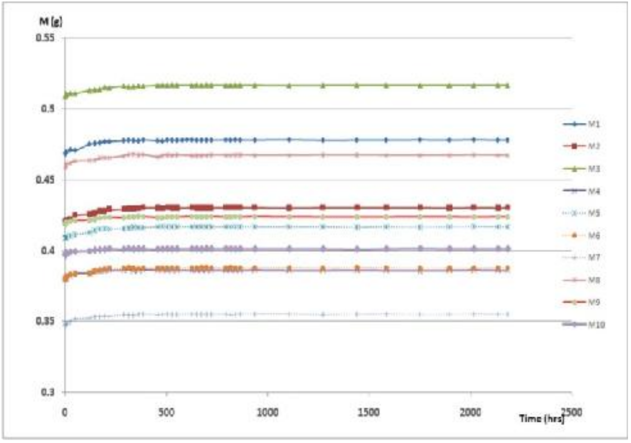

Fig. 2: Changes in weight over 90 days for composite materials.

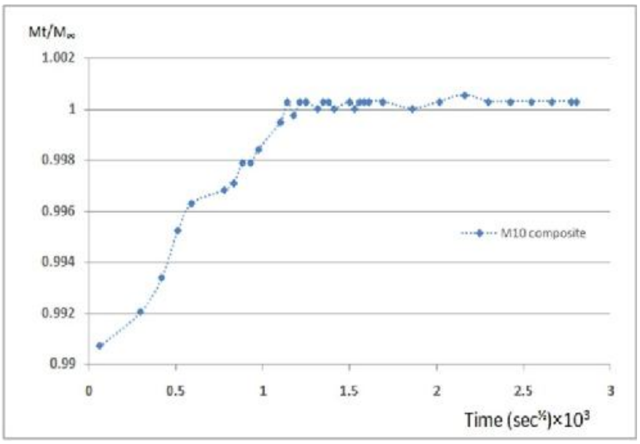

Fig. 3: M_t/M_∞ against $t^{1/2}$ (sorption) for M10 composites after immersion in water.

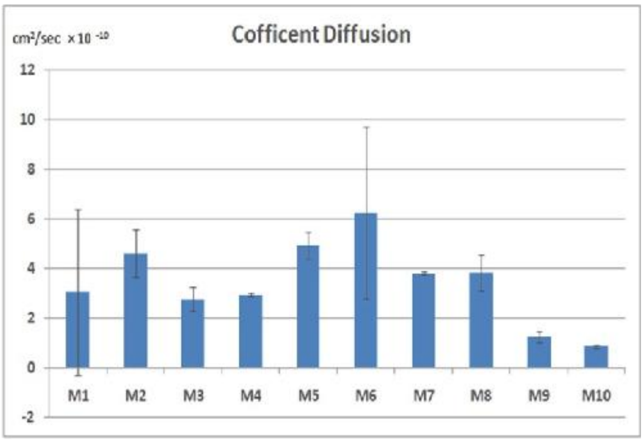

Fig.4:Mean diffusion coefficients values for composite materials tested.

DISCUSSION

The water sorption and solubility of dental restorative materials are of considerable clinical importance and cannot be neglected. Several data for water sorption and solubility for composite materials have been published, but it is difficult to correlate them as the results are often for different time periods and are expressed in different units. Moreover comparisons are difficult to make due to differences in reported specimen size, since different sizes of specimen will take different periods of time for water to completely infiltrate throughout the polymer matrix. Water molecules are able to diffuse through the inter-chain spaces of the resin matrix because of their smaller size of radius, which is less than 0.158 nm and smaller than the inter-chain spaces [13]. According to ISO 4049:2000 standard for dental restorative resins, a resin in order to be suitable for use as dental material must show water sorption lower than 40 µg/mm3 and solubility lower than 7.5 µg/mm^3. The values of water sorption for all studied composites, except that M6, are within the range of the ISO's standard. On the contrary, the values of solubility are within the range of the ISO's standard only for resin M7. Several factors, such as the polymeric matrix composition, filler particle type, mean particle sizes, and the degree of curing reached after the polymerization reaction can influence the solubility and sorption behavior of dental resin composites [14,15]. The studied resins used in this study have a great similarity in the filler particle content approximately 60% by volume. The Low water sorption values of resin are due to the method of preparation of Calcium Fluoroaluminosilicate glass, Filler particle size which was within 1 to 2 µm[12]. While the water solubility range was found between 2.9– 8.1 µg/mm^3 .These results were similar to those obtained by Oysaed and Ruyter (1.4– 9.0 µg/mm^3) [16].

Plots of Mt/M∞ against $t^{1/2}$ Figure 3 for M10 filling (similarly for all studied resin) were linear in the initial stages of absorption and desorption cycles, which shows that the uptake process for these composites is diffusion controlled. The diffusion coefficients which have been determined in this study is within the range of $0.7–6.2 \times 10^{-10}$ cm^2/sec figure 4, in good agreement with other works and comparable to those reported for composite resins soaked in water, despite the difference in the form of the water and hence in the driving force for sorption [3,17].

CONCLUSIONS

In this work, we conclude that the sorption and solubility values are in accordance with the ISO 4049:2000. The studied dental material (teeth filling) has shown that they have optimal physico-chemical properties for an adequate behavior in the oral aqueous environment, making it suitable for indirect composite restorations.

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Conflict of Interest: Nil

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