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Hydrolytic degradation of UDMA-based resin composite sealants

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The purpose of this study was to measure water sorption and solubility of some experimental UDMA-based resin composite sealants. The experimental resin composites consisted of a UDMA resin system and inorganic filler based on quartz and sodium fluoride. Samples of resin composites were stored in bidistilled water over a 21-day period. The results obtained for the proposed resin composite sealants showed that the presence of sodium fluoride influence water sorption and solubility. Water sorption and solubility increased in time and increase with amount of NaF from composites. Resin composite sealants with 20% by weight of NaF had the highest in water sorption and solubility values. The surface of composites investigated by SEM showed slow differences.

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Keywords: Dental composites, Resin composite sealants, Water sorption, Solubility

1. Introduction

Resin-based dental composites were developed as alternative to dental amalgam. These materials are based on polymer matrix (dimethacrylate resin and monomer of dilution) reinforced with a silane coated inorganic filler. Dental resin composites showed in time good mechanical and esthetic properties comparable to dental amalgam or dental ceramic.

Materials which are placed for long periods in oral environment will undergo an interaction with oral fluids. This interaction may involve dissolution or degradation of surface layers whilst in others the interaction may involve a leaching out of unbound or loosely bound components or an uptake of fluids into the structure of materials [1]. Water sorption in composite materials is a diffusion-controlled process, and the water uptake occurs largely in the resin matrix [2]. The water absorbed in resin composites could influence the phenomenon of plasticity in polymer matrix and can deliver fluoride and other ions outside from the bulk of the composites [3]. Water sorption could have both negative and benefical effects. In time the mechanical properties [4], porosity [5], color and translucence [6] could be influenced negatively of water sorption and solubility. Incorporation of fillers into a resin-material improves the mechanical properties [7, 8] and decreases the degradation in time [7].

P-aminobenzoic acid/cyclohexan on formaldelyde resins have been recently demonstrated as good hardners for epoxy resins [9].

One positive effect of water sorption is the compensation of polymerization shrinkage and the relaxion stress [10]. The addition of fluoride compounds in dental cements is known as an important factor for the fluoride release and the inhibition of dental caries. Fluoride release from dental restorative materials may prevent development of secondary carious lesions at the restoration tooth interface and can develop remineralization of enamel or decalcified dentin [11, 12]. Fluoride release inhibits bacterial metabolism from *Streptococcus mutans* [13] and *Streptococcus sobrinus*. *Streptococcus mutans* and *Streptococcus sobrinus* were associated with the initiation of human caries, while lactobacilli are associated with progression of the established lesion [14].

Fluoride release from dental composites is possible by solubility of filler based on fluoride salt [3] or by solubility fluoride ions from polymer matrix [15]. In all cases fluoride release is possible by a degradation of dental composite. There are some limits [16] for water sorption and solubility that can be tolerated and it lead to acceptable mechanical properties, porosity, color and translucence.

Dental sealants are thin plastic coatings (filled or unfilled) painted on the chewing surfaces of the molars and premolars to prevent dental decay (caries). Made of clear or shaded plastic, sealants are applied to the teeth to help keep them cavity-free.

The aim of this study was to analyze the influence of NaF on the water sorption and solubility of UDMA-based resin composite sealants. The surface of resin composites were investigated by SEM.

2. Experimental procedure

2.1. Materials

Five types of experimental light-curing resin composites were obtained by using a polymer matrix (Fig. 1) based on UDMA (urethane dimethacrylate resin)

(Aldrich Chemical, Milwaukee, WI, USA) and TEGDMA (triethyleneglycol dimethacrylate) (Sigma Chemical, St. Louis, MO, USA.) and inorganic filler based on quartz with variable quantities of sodium fluoride (0%, 5%, 10%, 15% and 20% by weight from filler) (Table 1).

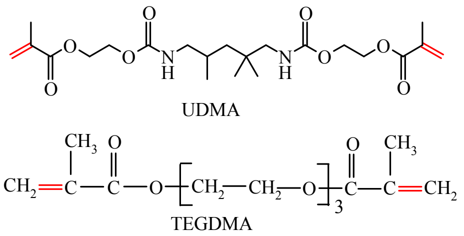

Fig. 1. Chemical structures of the UDMA and TEGDMA monomers.

Table 1. Composition of experimental light-curing resin composites.

Samples	Organic Composition (% by weight)	Inorganic Composition (% by weight)
C 0	UDMA– 55% TEGDMA – 45% DMAEMA – 1% CQ – 0,5%	Quartz – 100%
C 5	UDMA– 55% TEGDMA – 45% DMAEMA – 1% CQ – 0,5%	Quartz – 95% NaF– 5%
C 10	UDMA– 55% TEGDMA – 45% DMAEMA – 1% CQ – 0,5%	Quartz – 90% NaF– 10%
C 15	UDMA– 55% TEGDMA – 45% DMAEMA – 1% CQ – 0,5%	Quartz – 85% NaF– 15%
C20	UDMA– 55% TEGDMA – 45% DMAEMA – 1% CQ – 0,5%	Quartz – 80% NaF– 20%

Experimental resin composite sealants were prepared by incorporating the powder system in monomers liquid at the P/L ratio 1:1 w/w. The powder used was a mixture of silanized quartz with sodium fluoride. Camphorquinone (CQ) and N,N-dimethylaminoethyl methacrylate (DMAEMA) constituted the photoinitiator system and were supplied by Merck–Schuchardt and, respectively, by Aldrich Chemical, Milwaukee, WI, USA.

The macrofiller, consisting of Romanian quartz, was supplied by "Uricani" Factory and it was purified and grounded in our laboratory at a specific surfaces area of 9.7 m^2/g. The macrofiller (quartz) was mixed with sodium fluoride powder ("Târnăveni" ICC, RO). The grain size distribution of filler was determined by laser granulometry (model 715, Cilas Alcatel) and showed a particle diameter for quartz of 2.8 µm and 14.6 µm for NaF. Macro filer (quartz) was treated with a coupling agent γ-methacryloyloxypropyl-1-trimethoxysilane (silane A-174; Fig. 2) [3].

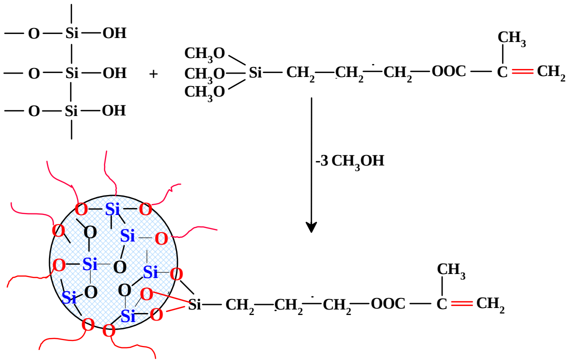

Fig. 2. Schematic diagram of surface treatment of inorganic silica filler with γ-methacryloyloxypropyl-1-trimethoxysilane (A-174).

2.2. Methods

2.2.1. Specimen preparation

Samples (n = 5) for water sorption and solubility test 15 mm diameter by 1 mm height were cured by XL3000 photocuring source (3M Dental Products, St Paul, MN, USA). This source consisted of a 35-W tungsten halogen lamp. Specimens were prepared by overfilling the mold with the tested resin composites and by covering it with a transparent sheet of polyester film and a glass slip and pressed to remove any excess. The glass plate was removed and the specimens were irradiated for 60s on each side. Immediately after polymerization, the samples were removed from the mold and any specimen with visible voids was disicarded. Every sample was weighed to an accuracy ±0.0001 g and suspended in 20 ml bidistilled water.

2.2.2. Water sorption and solubility

After 1 week the specimens were removed, washed with bidistilled water, dried by blotting with absorbent paper, and waving in air for 15 s. The specimens were transferred to a vacuum desiccator at 37 ºC (±2) and dried over silicagel. After 24 h the samples were removed and weighed to an accuracy ±0.0001 g. Water sorption (W_{SP}) and solubility (W_{SL}) were calculated according to the ISO standard [15] as described below:

Solubility

$$W_{SL} = m_1 - m_3 / V \quad \mu g/mm^3$$

m_1—is the mass of the specimen in micrograms, before immersion in water.

m_3—is the mass of the reconditioned specimen, in micrograms.

V—is the volume of the specimen, in cubic millimeters.

Water sorption

$$W_{SP} = m_2 - m_3/V \quad \mu g/mm^3$$

m_2—is the mass of the specimen, in micrograms, after immersion in water for 7 days.

m_3—is the mass of the reconditioned specimen, in micrograms.

V—is the volume of the specimen, in cubic millimeters.

Resin composite samples were tested in bidistiled water at days 7, 14 and 21.

2.2.3. Examination of surface

The surfaces composite's were investigated by SEM (Hitachi S-2600N) before and after one week of immersion

2.2.4. Statistical analyses

Data were analyzed by one-way analysis of variance (ANOVA) with the level of significance set at 0.05. There was no statistically significant difference.

3. Results and discussion

The literature data for water sorption and solubility of dental composites have showed very different values. Some of the factors than could influence the degradation of dental composites are:

1. Filler size, the volume of filler from dental composites [17], granulometry of powder [18], surface treatment of the fillers [2, 18, 19] and type of powder [1, 21]. GIC (glass ionomer cement) and RMGIC (resin-modified glass ionomer cement) have showed higher water sorption and solubility values than resin composites [21].

2. Polarity of polymer matrix (hydrophilic or hydrophobic) [22].

3. Temperature from medium of storage [19, 23], time of storage [23, 24], degree of conversion [23, 25], bubble of air from matrix, pH of the storage medium [26].

Time and amount of NaF from composites influenced in a significant way solubility values (Fig. 3), especially in the case of composites C15 and C20. The water sorption results (Fig. 4) showed not significant differences after 7, 14 and 21 days of storage for composites with 0 %, 5 % and 10 % by weight of NaF.

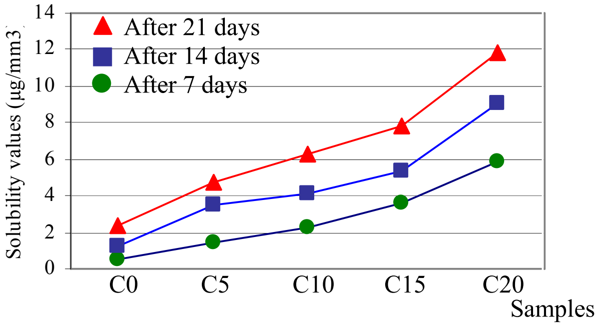

Fig. 3. Mean values of solubility after 7, 14 and 21 days.

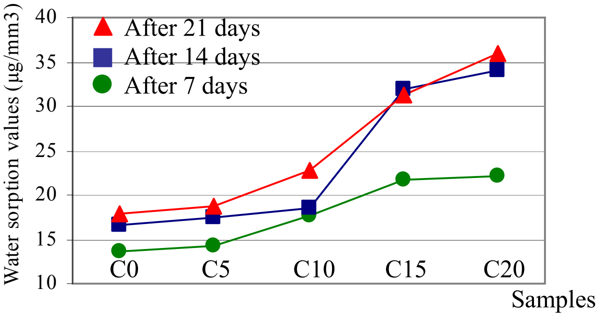

Fig. 4. Mean values of water sorption after 7, 14 and 21 days.

The increasing of water sorption and solubility values for composites with highest NaF in composition could be explained mainly by on NaF solubility in water. Fluoride release from dental composites is possible by solubility of fluoride compounds from dental composites and diffusion fluoride ions in water and transport of water outside from dental composite matrix [1, 3]. The highest values for the water sorption were recorded for composites with 15% and 20% by weight of NaF. On the first day the solubility at the composite surface is the most important process. In time the process of water sorption is combined with solubility of composites and the degradation increase in time.

The lowest water sorption and solubility for composites C0 (without NaF in composition) could be explained by an insolubility of quartz filler. The values registered for C0 during days of storage could rather be explained by monomer elution [26, 27] or by a degradation of filler-polymer interface [23]. The UDMA polymer matrix used in this study showed a higher hydrofobicity [22] than composites based on BIS-GMA [3].

The degradation at the surface of resin composites was confirmed also by SEM photograph from the surface of composites (Fig. 5). After 1 week of storage the SEM photograph of composite surface showed not significant difference for composites C0, C5, and C10. These results were confirmed also of water sorption and solubility results. A slow degradation of the surface of composites was observed for composites C10 (15% NaF) and C20

(20% NaF). The presence of the voids in SEM photograph of composite surface C10 and C20 after 7 days could be explained by a release of NaF powder or an elution of quartz filler polymer matrix. Elution of quartz filler from composites could be explained by a breaking of chemical bond at the interface filler-polymer matrix and from the composite [19].

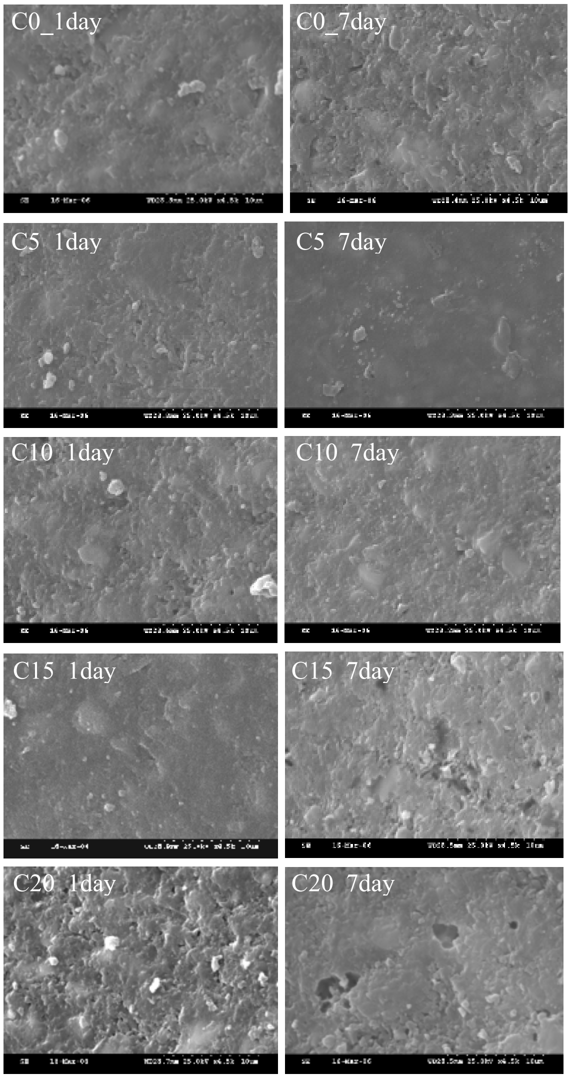

Fig. 5. SEM photograph of surface composites: C0, C5, C10, C15 and C20 before immersion and after 7 days from immersion in water.

4. Conclusions

The sorption and solubility behavior for resin composite sealants were influenced by time of storage.

The increase of NaF amount in resin composites increase water sorption and solubility values. The highest values for water sorption and solubility were recorded for composites C20 (with 20% by weight of NaF). The higher degradation for samples composites C20 was confirmed also from SEM photograph of surface composites.

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