Long-term water sorption of three resin-based restorative materials

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Purpose: To evaluate water sorption of three resin-based restorative materials over 180 days of storage in de-ionized water.

Materials and Methods: Three disks (12.0 mm in diameter and 1.25 mm in thickness) were prepared for each light-cured resin-based material; a polyacid-modified resin composite (élan), an organically modified ceramic (Definite), and a hybrid resin composite (Prodigy). The disks were immersed in de-ionized water at 37°C for 180 days. The weight of the disks was measured at intervals of 1, 7, 14, 60, and 180 days. The results were analyzed using two–way ANOVA followed by the Bonferroni comparison test at 95% level of confidence.

Results: Significant differences (p<0.05) were found among the materials and the storage time. Over 180 days, élan showed earlier and higher water sorption compared with the other materials. Prodigy exhibited the least water sorption over the test period.

Conclusion: Water sorption of the resin-based restorative materials was material and storage time dependent. Prodigy resin composite was the most stable material with the lowest water sorption.

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Clinical Significance: The polyacid-modified resin composite (élan) showed earlier and higher water sorption behavior than the other materials. It was suggested that undesirable properties might appear subsequent to water sorption.

Key Words: polyacid-modified resin composite, resin composite, water sorption.

Introduction

Resin composite was introduced more than 40 years ago, and at that time its use was routinely for anterior teeth. Improvements in mechanical properties, biocompatibility, handling and color matching have lead to its wide acceptance for virtually all types of cavities. An attempt to combine the desirable properties of resin composite and those of glass ionomer cement (GIC) led to the development of polyacid modified resin composite (PAMRC). Resin-based materials demonstrate water sorption in the oral cavity, which is the amount of the water absorbed by the material on the surface and into the body while the restoration is in service.1 The most popular method for measurement of the water sorption of restorative materials is the gravimetric methodology that involves weighing at different periods.2-11

Water sorption affects the physical and mechanical properties of resin composite, such as dimensional change,12,15 decrease in surface hardness and wear resistance,14 filler leaching and change in color stability,15 reduction in elastic modulus,16 an increase in creep and a reduction in ultimate strength,2 fracture strength, fracture toughness, and flexural strength.17 Water sorption increases as the amount of resin matrix increases and filler content decreases, since the filler particles do not absorb water.2-4,12,16,18,19 Factors, such as type of resin composite material, chemical composition, storage time, type and pH of storage solution, and degree of polymerization, strongly affect the water sorption and leachability behavior of the material.5,7,20,21 Water
penetration into the material has been shown to be directly related to the degradation of the filler-matrix interface.\textsuperscript{3,22}

The purpose of this study was to evaluate the water sorption of three resin-based restorative materials over 180 days. The tested hypothesis was that the material type and storage time have an influence on water sorption.

Materials and Methods

The tested materials, their batch numbers, compositions and their manufacturers are listed in Table 1. Three types of resin-based restorative materials were used; a polyacid-modified resin-based composite (élan, Kerr Corp., Orange, CA, USA), an organically modified ceramic material (Definite, Degussa, Hanau, Germany), and a microhybrid composite (Prodigy, Kerr Corp.). The élan material has 76wt\% inorganic filler particles with an average particle size of 1.4 $\mu$m. Definite has 77wt\% inorganic filler particles with an average particle size of 1-1.5 $\mu$m. Prodigy has 79wt\% filler particles with an average particle size of 0.6 $\mu$m.

Table 1. Resin-based restorative materials used in the study.

<table>
<thead>
<tr>
<th>Material</th>
<th>Lot number</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>élan</td>
<td>807294</td>
<td>Polymerizable poly-carboxylic acid, ETMPTA, EBis-GMA, TEGDMA, Al-F-silicate, BPO</td>
</tr>
<tr>
<td>Definite</td>
<td>801705</td>
<td>Poly-DMA-polysiloxane, Various DMA, Glass polymerizable SiO$_2$, Bis-GMA, TEGDMA</td>
</tr>
<tr>
<td>Prodigy</td>
<td>25897</td>
<td>EBis-GMA, Barium aluminum borosilicate, SiO$_2$, TiO$_2$</td>
</tr>
</tbody>
</table>

ETMPTA, Ethoxylated trimethylolpropane triacrylate; EBis-GMA, Ethoxylated bisphenol-A diglycidyl methacrylate; TEGDMA, Triethyleneoxyglycol dimethacrylate; BPO, Benzoyl peroxide; DMA, Dimethacrylate; Bis-GMA, Bisphenol-A diglycidyl methacrylate.

Three specimens of each material were prepared following the manufacturer’s recommendations. A stainless-steel mold with a 12.0 mm in diameter and 1.25 mm in thickness was placed on a celluloid matrix strip on a glass slide, and one of the three materials was packed into the mold. A second strip with a glass slide was placed onto the resin-based material. Sufficient finger pressure was applied until the two strips contacted with the mold. The resin materials were light-cured with a halogen light-curing unit (DE 19963-0359, Dentsply/Caulk, Milford, DE, USA) from both top and bottom sides for a total of 120 s (Fig. 1).

The disks were dried in desiccators with calcium sulfate desiccant until a constant weight was achieved throughout daily weighing of the samples for 14 days. An electronic balance (PC 400, Mettler, Mettler Weagen GmbH, Switzerland) was used for weight determination to an accuracy of 0.1 mg. Each disk was stored in a
tightly capped polypropylene container with 10 mL of de-ionized water at 37°C over 180 days. The weights of the disks were measured at intervals of 1, 7, 14, 60, and 180 days. Water evaporation during the storage period was completed from a blank solution in order to keep the specimens continuously immersed in 10 mL of de-ionized water.

The specimens were dried with filter papers and weighed within one minute. The dry weight (the weight of the specimens after desiccation and before immersion in water) was subtracted from their wet weight (the weight of the specimens after their immersion in water) to determine water gain. The average water gain of the materials at each storage interval was recorded and divided by the total surface area of the disk to calculate water sorption.

A two-way analysis of variance (ANOVA) and Bonferroni test was used to observe differences in water sorption among the specimens as a function of time (paired comparison) and among the materials (non-paired comparison) as main effects at a 95% level of confidence.

Results

The amounts of water sorption of the three resin-based restorative materials accumulated over 180 days are summarized in Table 2 and Fig. 2. The amount of water sorption was influenced by the restorative material (F=59.949, p<0.001) and storage time (F=11.939, p<0.001).

The water sorption of the three materials was initially high during the first day of storage; however, élan absorbed approximately 56% of its total water sorption (over 180 days) in the first day of storage, while Definite and Prodigy absorbed approximately 47% and 40%, respectively.

Table 2. Accumulated water sorption (mg/cm²) of the resin-based restorative materials (n=3).

<table>
<thead>
<tr>
<th>Storage period</th>
<th>Material 1 day Mean SD</th>
<th>7 days Mean SD</th>
<th>14 days Mean SD</th>
<th>60 days Mean SD</th>
<th>180 days Mean SD</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>élan</td>
<td>0.58 0.25 a</td>
<td>1.31 0.28 A</td>
<td>1.26 0.27 A</td>
<td>1.26 0.25 A</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>A</td>
<td>A</td>
<td>A</td>
</tr>
<tr>
<td></td>
<td>Definite</td>
<td>0.33 0.00 a, b</td>
<td>0.59 0.05 B, C</td>
<td>0.79 0.15 C</td>
<td>0.72 0.10 B, C</td>
</tr>
<tr>
<td></td>
<td></td>
<td>a, b</td>
<td>b, C</td>
<td>d</td>
<td>c</td>
</tr>
<tr>
<td></td>
<td>Prodigy</td>
<td>0.19 0.04 b</td>
<td>0.44 0.20 c D</td>
<td>0.58 0.14 d D</td>
<td>0.39 0.10 e D</td>
</tr>
<tr>
<td></td>
<td></td>
<td>D</td>
<td>c</td>
<td>D</td>
<td>e</td>
</tr>
</tbody>
</table>

There were no significant differences within the horizontal columns and vertical rows labeled with the same large and small superscript letters, respectively (p>0.05).

The amount of accumulated water sorption of élan at the first day of storage was significantly lower than the amount at subsequent storage periods. However, Definite showed no difference in water sorption among the storage periods except between 1 day and 14 days, while Prodigy showed no difference in water sorption among the storage periods.

As expected, over 180 days of the storage, the water sorption of élan was significantly higher than that of Definite and Prodigy except for 1 and 180 days, in which there were no significant differences between élan and Definite. Meanwhile, no significant difference was recorded between Definite and Prodigy through the storage periods.
Discussion

Nowadays, hybrid resin composite is recommended for use in a wide range of clinical situations. It contains fine and microfine filler particles that occupy around 80 wt% of the resin material. This formula provides mechanical properties superior to other classes of resin composites. Prodigy is a light-cured micro-hybrid composite for anterior and posterior applications. Its average particle size of 0.6 µm ensures an excellent and lasting finish together with ease of handling and placement.

Definite is a new class of highly filled hybrid resin composite with a different filler distribution that changes their consistency to being less sticky and stiffer in comparison with a conventional hybrid. Definite is based on Ormocer technology characterized by a novel inorganic-organic copolymer in its formulation instead of a traditional monomer system. A multifunctional urethane and thioether methacrylate aloxysilanes have been developed for synthesis of an inorganic-organic copolymer.

The élan material is a polyacid-modified resin composite, so-called “compomer” which has two main constitutes; a dimethacrylate monomer with two carboxylic groups and filler similar to ion-leachable glass present in glass-ionomer cement (GIC). The ion-leachable glasses are mostly calcium-aluminum fluorosilicate to ensure direct bonding with the matrix through partial silanization. New bifunctional monomers react simultaneously with methacrylates by a process of polymerization and with the glass particles in the presence of water to initiate an acid-base neutralization reaction. The élan material was selected for evaluation because this resinous material contains elements of GIC, that have hydrophilic properties and would be anticipated to show more water sorption as is case with resin-modified GIC when compared to the composites.

The three resin-based materials exhibited high water sorption at the initial time of storage. Under all time conditions, the equilibrium water content of élan was greater than those for Definite and Prodigy. These results agreed with the results of previous studies.

The early and high water sorption of élan might be attributed to less resistant to crack propagation compared
with the resin composites, which might be a factor for acceleration of water sorption. Also, polyacid-modified resin composite has a high fluoride content with no water in its composition. The initial setting of the material is due to photo-polymerization and subsequently an acid-base reaction after exposure of the material to water as a second stage mechanism. The polyacid-modified resin composite contains bifunctional monomers with hydrophilic functional groups and two double bonds. Water uptake would take place preferentially at the hydrophilic site of the resin matrix after polymerization.

Definite composite showed a significant difference in water sorption between 1 day and 14 days intervals. Diaz-Arnold et al. measured the water sorption of resin composite adhesives with near-infrared spectroscopy. They also reported that the most rapid water sorption occurred within the first 14 days of storage in the following 370 days water storage period. The minimal amount of water sorption was obtained in Prodigy through the whole time frame, which may be due to the highly filled resin with a hydrophobic monomer of Bis-GMA.

Polishing was not carried out during specimen preparation in this study. Therefore, the surface layer of the specimen was resin rich, and totally different from the inside of the specimen. If the resin rich surfaces of the specimens were removed before, the amount of water sorption is expected to be lower. Water sorption of a resin composite is a diffusion-controlled process. Long-term water penetration into 2.5 mm block of dental composites was evaluated using silver nitrate staining. The staining material continued to penetrate up to three years with a proportional behavior. Therefore, specimen size is one of the critical factors affecting the time required to be saturated. A large portion of the water was reversibly absorbed by the resin composite materials. However, Ferracane and Condon (1990) have described the inability to remove 100% of absorbed water by short-term desiccation.

It was reported that high water sorption at the first day was responsible for the large hygroscopic expansion, which continued up to 180 days. In contrast, other researchers reported that hygroscopic expansion following water sorption equilibrates after 7 days. Since direct composite restorations are widely accepted, further characterization of the effects of water on the resin composite materials is warranted.

**Conclusion**

The resin-based materials investigated in this study demonstrated different levels of water sorption. The polyacid-modified resin composite showed higher water sorption than composite resin. Prodigy (hybrid composite) was a stable material with the least water sorption. The water sorption of élan and Definite tended to increase up to 7th day and 14th day, respectively.

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**References**


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