INTRODUCTION

Resin-based composites, polyacid-modified resin composites, and glass-ionomer cement materials are well-established cosmetic restorative materials used extensively in modern-day preventive and conservative dentistry. However, these materials may absorb significant amounts of water when exposed to aqueous environments.\(^1,^2\)

Two different mechanisms occur when dental restorative materials are exposed to or stored in water: first, they gain weight from water uptake, and the second that they lose weight from dissolution in water.\(^3\) The property of sorption indicates a combination of adsorption and absorption. Adsorption involves adherence of liquid molecules to the surface of a solid material. Absorption involves penetration of liquid molecules into the structure of the solid material, mainly through the process of diffusion.\(^4\) Dental restorative materials are in continuous contact with fluids and saliva in the patient’s mouth. Consequently, the water sorption and solubility of these materials are of considerable importance. Fluid uptake into resin-based materials may have both beneficial and detrimental consequences.\(^5\) Hygroscopic expansion, which is caused by water uptake, may relieve the residual stresses of resin-based composite materials during polymerization shrinkage and may also reduce interfacial gap

ABSTRACT

The aim of the study was to evaluate the effects of immersion in artificial saliva and distilled water on the water sorption and solubility properties of different tooth-colored restoratives. Ten disks/material of (Z250, Z250XT, Z350XT, P90, SDR, F2000, N100) were fabricated. The specimens were placed in a desiccator at 37°C for 22 h and then transferred to another desiccator at 22°C ± 1°C for 2 h, weighed to a precision accuracy of ± 0.0001 g. This cycle was repeated to obtain constant mass \(m_1\), and then the specimens were divided into two groups (5/each), immersed in distilled water or artificial saliva at 37°C for 7 days. After storage, the specimens were re-weighted to obtain \(m_2\) and then reconditioned in the desiccator to obtain \(m_3\). Sorption and solubility were calculated. Data were statistically analyzed using one-way ANOVA and post hoc Tukey’s multiple comparison tests at \(\alpha = 0.05\). N100 showed the highest mean values for water sorption (113.19 and 136.55 \(\mu g / mm^3\)) in distilled water and artificial saliva, respectively. P90 had the lowest mean value for water sorption (18.06 \(\mu g / mm^3\)) in artificial saliva and no sorption in distilled water. N100 mean values for solubility in distilled water (9.66 \(\mu g / mm^3\)) and artificial saliva (8.55 \(\mu g / mm^3\)) were higher than the other materials. The solubility values for Z250XT, Z350XT, P90, SDR, and F2000 were significantly \((P < 0.05)\) higher in artificial saliva than in distilled water. Finally, dental restoratives with sorption and/or solubility values above the critical points (40 \(\mu g / mm^3\) and 7.5 \(\mu g / mm^3\) respectively) should not be used clinically.

Key words: Sorption, Solubility, Artificial saliva, Distilled water, Tooth-colored materials
width, with a corresponding reduction in microleakage. In contrast, fluid sorption may lead to discoloration and act as a plasticizer, leading to deterioration of physical/mechanical properties and decreasing the life of these materials, thus reducing their bond to tooth structures.

Glass-ionomer cements are also widely used in clinical dentistry. They are water-based materials that consist of special ion-leachable glass and water-soluble polymeric acids, and set by an acid-base reaction in the presence of water. Conventional and resin-modified glass-ionomer cements absorb water and may dissolve by surface wash-off, diffusion of water through pores and cracks in the cement, and diffusion from the bulk of the cement. Sorption and solubility of tooth-colored restorative materials can be affected by many factors, including material types, matrix composition variations, filler size and distribution, and efficiency of polymerization, as well as by the use of different immersion media.

However, the use of artificial saliva rather than water as an immersion medium would be more compatible with conditions in the oral environment and would provide more realistic information about these phenomena. Therefore, the aim of this in vitro study was to evaluate the effects of artificial saliva storage and immersion in distilled water on the water sorption and solubility properties of different novel direct tooth-colored restorative materials. The hypothesis tested was that, there will be no difference between the immersion in artificial saliva and distilled water on the water sorption and solubility properties of different direct tooth-colored restoratives.

METHODOLOGY

Seven direct tooth-colored restorative materials including microhybrid composite (Filtek Z250, 3M ESPE, St. Paul, MN, USA), nanohybrid composite (Filtek Z250XT, 3M ESPE), nanocomposite (Filtek Z350, 3M ESPE), silorane-based low-shrink composite (Filtek P90, 3M ESPE), flowable composite (SDR, Dentsply, Milford, DE, USA), compomer (F 2000, 3M ESPE), and a resin-modified glass-ionomer (N100, 3M ESPE) were tested, and their compositions are given in Table 1. The halogen curing device Elipar™ 2500 (3M ESPE) was used to polymerize the materials. Each specimen was polymerized for 40 seconds, and the output of the curing device was measured before the onset of the polymerization process and after a series of 10 specimens had been polymerized, with a radiometer (Optilux Model 100, SDS Kerr, Danbury, CT, USA), to verify that the density of the light power used was within the range of 480 to 520 mW/cm².

Specimen preparation and the water sorption and solubility tests were carried out according to ISO 4049-2009(E): Polymer-based restorative materials. From each material, 10 disks were fabricated in a stainless steel cylindrical mold (15.0 mm diameter × 1.0 mm thickness) at 22°C ± 1°C (room temperature). After removal from the molds, the specimens were transferred to a desiccator containing silica gel maintained at 37°C for 22 h and then transferred to another desiccator at 22°C ± 1°C for 2 hours. The specimens were weighed to a precision accuracy of ± 0.0001 g (Mettler-Toledo International Inc., Greifensee-Zürich, Switzerland). This cycle was repeated for 2 weeks until a constant mass (m₁) was obtained. We calculated the volume (V) of each specimen by measuring their diameters and thicknesses, using a digital micrometer (Sylvac Ultra-Cal Mark III, Fowler Tools and Instruments, Newton, MA, USA). The specimens of each material were divided randomly into two groups (5/each), immersed individually in glass tubes containing 15 ml distilled water or artificial saliva, and stored in an oven at 37°C for 7 days. Artificial saliva was comprised of sodium chloride (NaCl) 0.4 g, potassium chloride (KCl) 0.4 g, calcium chloride (CaCl₂. H₂O) 0.795 g, sodium-di-hydrogen phosphate (NaH₂PO₄.H₂O) 0.69 g, sodium sulfate (Na₂SO₄.9H₂O) 0.005 g, and distilled water 1000 ml, and the pH was adjusted to 7.17. Group 1 was immersed in distilled water at 37°C for 7 days. Group 2 was immersed in artificial saliva at 37°C for 7 days. After storage, the specimens were removed from the two solutions, blot-dried with absorbent paper, and air-dried for 15 seconds. The specimens were then re-weighed to obtain m₂ after which they were reconditioned to constant mass in the desiccator. This constant mass was recorded as m₃.

Sorption (Wₛ) and Solubility (Wₛ) were calculated as follows:

\[
\text{Sorption } W_s = \frac{(m_2 - m_1)}{V} \quad \text{Solubility } W_s = \frac{(m_1 - m_3)}{V}
\]

Statistical Analysis

The data were entered into Excel and analyzed with SPSS version 16.0 software (SPSS Inc., Chicago, IL, USA). Data were subjected to one-way ANOVA and post hoc Tukey’s multiple comparison tests, with the probability for statistical significance set at α = 0.05.

RESULTS

The mean values and standard deviations of sorption and solubility for the different materials tested in two different immersion media (distilled water and artificial saliva) were analyzed. The results indicated that, artificial saliva significantly increased water sorption and solubility compared to distilled water for all seven materials. No significant differences were found between the immersion in artificial saliva and distilled water on the water sorption and solubility properties of different direct tooth-colored restoratives.
Effect of immersion media on tooth-colored restoratives

<table>
<thead>
<tr>
<th>Material</th>
<th>Type</th>
<th>Composition</th>
<th>Manufacturer</th>
</tr>
</thead>
</table>
| Filtek™ Z250      | Micro-hybrid resin composite | **Matrix:** Bis-GMA, UDMA, and Bis-EMA.  
                              | **Filler:** zirconia/silica (0.01-3.5 μm).  
                              | **Filler by volume:** 60%  
                              | **Filler by weight:** 82% | 3M ESPE Dental Products, St. Paul, MN, USA. |
| Filtek™ Z250 XT   | Nano-hybrid resin composite  | **Matrix:** Bis-GMA, UDMA, Bis-EMA,  
                              | **Filler:** Surface-modified zirconia/silica (0.1-10 μm) and 20 nm surface-modified silica.  
                              | **Filler by volume:** 67.8%  
                              | **Filler by weight:** 81.8% | 3M ESPE Dental Products, St. Paul, MN, USA. |
| Filtek™ Z350 XT   | Nano-filled resin composite  | **Matrix:** Bis-GMA, UDMA, TEGDMA,  
                              | **Filler:** Combination of non-aggregated 20nm silica, non-aggregated 4-11nm zirconia, and aggregated zirconia/silica cluster filler.  
                              | **Filler by volume:** 63.3%  
                              | **Filler by weight:** 78.5% | 3M ESPE Dental Products, St. Paul, MN, USA. |
| Filtek™ P90       | Low-shrink resin composite   | **Matrix:** New ring-opening Silorane  
                              | **Filler:** Epoxy functional silane treated SiO₂ and ytterbium fluoride (0.1-2 μm)  
                              | **Filler volume:** 55%  
                              | **Filler by weight:** 76% | 3M ESPE Dental Products, Seefeld, Germany. |
| Ketac™ N100       | Light-curing nano-ionomer    | -De-ionized water  
                              | -HEMA  
                              | -Copolymer (a methacrylatemodified polyalkenoic acid)  
                              | **Filler:** Fluoroaluminosilicate glass nanomers, and nanoclusters (0.6-1.4μm)  
                              | **Filler by volume:** 59.5%  
                              | **Filler by weight:** 69% | 3M ESPE Dental Products, St. Paul, MN, USA. |
| SDR               | Posterior bulk fill flowable base | **Matrix:** MUDM, EBPADMA, TEGDMA  
                              | **Filler:** Barium and strontium alumino-fluoro-silicate glasses.  
                              | **Filler by volume:** 45%  
                              | **Filler by weight:** 68% | Dentsply Caulk, West Clarke Ave., Milford, DE, USA. |
| F2000             | Polyacid-modified resin composite | **Matrix:** CDMA oligomer, GDMA, hydrophilic polymer  
                              | **Filler:** Colloidal silica, and alumino-fluoro-silicate glasses (3-10 μm)  
                              | **Filler by volume:** 67%  
                              | **Filler by weight:** 84% | 3M ESPE Dental Products, St. Paul, MN, USA. |

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The highest mean values of water sorption were exhibited by N100 in the distilled water (113.19 μg/mm³) and artificial saliva (136.55 μg/mm³) and were significantly higher than those of the other materials (P<0.0001). P90 had the lowest mean value of water sorption (18.06 μg/mm³) in the artificial saliva and no sorption in distilled water.

The sorption values of Z250, P90, and N100 were significantly higher in the artificial saliva than in the distilled water (P < 0.0001), while the results were opposite in Z350XT and SDR (P<0.0001 and P= 0.02, respectively), with no significant difference in water sorption values in both media for Z250XT and F2000 (P = 0.789 and 0.519, respectively) [Figure 1].

The mean solubility values for N100 in the distilled water (9.66 μg/mm³) and artificial saliva (8.55 μg/mm³)
TABLE 2: MEAN VALUES (SDS) OF SORPTION AND SOLUBILITY FOR THE DIFFERENT MATERIALS THAT WERE TESTED IN TWO DIFFERENT IMMERSSION MEDIA (DISTILLED WATER AND ARTIFICIAL SALIVA)

<table>
<thead>
<tr>
<th>Property</th>
<th>Material</th>
<th>Distilled Water</th>
<th>Artificial Saliva</th>
<th>P Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sorption</td>
<td>Z250</td>
<td>18.94 (1.60)</td>
<td>28.69 (0.79)</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td>Mean (SD)</td>
<td>Z250XT</td>
<td>30.79 (0.85)</td>
<td>30.62 (1.09)</td>
<td>0.789</td>
</tr>
<tr>
<td></td>
<td>Z350XT</td>
<td>28.16 (0.74)</td>
<td>20.31 (1.02)</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td></td>
<td>P90</td>
<td>0</td>
<td>18.06 (0.93)</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td></td>
<td>N100</td>
<td>113.19 (0.93)</td>
<td>136.55 (1.89)</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td></td>
<td>SDR</td>
<td>41.09 (1.07)</td>
<td>39.13 (1.06)</td>
<td>0.020</td>
</tr>
<tr>
<td></td>
<td>F2000</td>
<td>40.16 (1.02)</td>
<td>40.59 (0.99)</td>
<td>0.519</td>
</tr>
<tr>
<td>Solubility</td>
<td>Z250</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Mean (SD)</td>
<td>Z250XT</td>
<td>0</td>
<td>5.04 (0.13)</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td></td>
<td>Z350XT</td>
<td>0</td>
<td>5.08 (0.13)</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td></td>
<td>P90</td>
<td>0</td>
<td>4.24 (0.29)</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td></td>
<td>N100</td>
<td>9.66 (0.49)</td>
<td>8.55 (0.62)</td>
<td>0.014</td>
</tr>
<tr>
<td></td>
<td>SDR</td>
<td>1.88 (0.26)</td>
<td>4.78 (0.17)</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td></td>
<td>F2000</td>
<td>5.89 (0.18)</td>
<td>6.18 (0.22)</td>
<td>0.054</td>
</tr>
</tbody>
</table>

were higher than those of the other materials (P< 0.0001) (Z250, Z250XT, Z350XT, and P90), which showed no solubility in the distilled water.

The solubility values for Z250XT, Z350XT, P90, SDR, and F2000 were significantly higher in the artificial saliva than in the distilled water (P< 0.0001 for all of them and P= 0.054 for F2000), while the opposite was true for N100 (P = 0.014) [Figure 2].

DISCUSSION

According to ISO standard 4049-2009, the maximum acceptable and critical values of sorption (Wsp) and solubility (Wsl) for polymer-based restorative materials are 40 μg/mm³ and 7.5 μg/mm³, respectively. Water sorption and solubility values above these critical points have an adverse effect on the physical and mechanical properties of these materials, such as strength, surface hardness, wear resistance, and color stability, and thus on their clinical behavior and biocompatibility.13, 16-20

Water sorption values of the resin-modified glass ionomer (N100) in distilled water and artificial saliva (113.19 and 136.55 μg/mm³, respectively) were the highest compared with those of the other materials, followed by posterior bulk fill flowable base (SDR) (41.09 and 39.13 μg/mm³, respectively) and then polyacid-modified resin composite (F2000) (40.16 and 40.59 μg/mm³ respectively).

The presence of hydrophilic constituents of HEMA or resin molecules that contain hydrophilic moieties and acid-base reaction as part of the photochemical polymerization process explains the high level of water sorption in N100, in addition to that the material may contain air bubbles due to the hand mixing of the two pastes, which may accelerate water sorption.3,21-23 Posterior bulk fill flowable base (SDR) contains the lowest filler load, with ethoxylated bisphenol A dimethacrylate (EBPADMA), modified urethane dimethacrylate (MUDM), and triethylene glycol dimethacrylate (TEGDMA) resins, and the last two are more hydrophilic than EBPADMA, so higher water sorption is expected from this material compared with other resin-based composite materials.3, 19

Polyacid-modified resin composite (F2000) had a higher water sorption value compared with the other resin-based composite materials tested in this study. This was not unexpected, due to the presence of the hydrophilic polymer glyceryldimethacrylate (GDMA); however, the value was slightly lower than that of the flowable resin composite (SDR), which might be due to high filler content.3, 24 In addition, the acid-base reaction is initially limited in this type of material because of its anhydrous structure, but once water is absorbed, a delayed acid-base reaction is likely.25

The findings of this study showed no water sorption in distilled water for the low-shrink resin composite (P90), and the water sorption values of the other three resin-based composites were lowest for Z250 followed...
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by Z350XT and Z250XT; which can be explained by the presence of hydrophobic siloxane backbone in P90 and the absence of TEGDMA in Z250, however water sorption of the polymers that contain BISGMA-co-TEGDMA were higher than BISGMA based polymers.26, 27

The immersion, in two different media (distilled water and artificial saliva), of specimens fabricated from different materials showed different results (Figure 1), and this finding was contrary to that of Zhang and Xu.28 This might be due to the difference in the components of the two immersion media.

The mean values of water solubility in both media (distilled water and artificial saliva) presented by all tested materials were lower than the maximum acceptable value (7.5 μg/mm³), except those for N100(9.66 and 8.55 μg/mm³, respectively), which exceeded the maximum acceptable value. This might be due to the more soluble matrix composition of N100 and the evaporation of water during drying.

Dental resin-based composites contain barium glass, strontium glass, zirconia, borosilicate glass, quartz, silica, ceramic, and prepolymerized resin as filler particles.29-31 Some studies have shown that barium and strontium glasses are more readily leached into water than silica particles, and that the resin-based composite with quartz filler is more stable than those with metallo-silica glasses.32, 33 In addition, the mass loss of silica-containing resin-based composites after water contact might be due to the dislodging of filler particles by the breaking of siloxane bonds, and to hydrolysis.34

This study showed that the resin-based composite materials (Z250XT, Z350XT, and P90) were not soluble in distilled water, and this has been supported by the absence of conclusive evidence of hydrolytic filler degradation.35 In contrast, resin-based composite materials (Z250XT, Z350, P90, and SDR) had more solubility in the artificial saliva than in the distilled water, and this was supported by evidence that the leaching process was faster in saliva than in distilled water.35

Only one material (Z250) showed no solubility in both media (distilled water and artificial saliva), and this was supported by the absence of evidence of degradation from this type of resin-based (micro-hybrid) composite. Solubility of F2000 in artificial saliva was higher than that in distilled water. This may be attributed to the pH of the medium, resulting in a more aggravated attack on the glass, and may increase fluoride release from the material.36

Due to the availability of a lot of dental restorative materials in the market, the data that relative to the mechanical and physical properties of dental materials should be included in manufacturers’ booklets to provide clinicians with the best scientific information about these materials. Furthermore, dental restorative materials with sorption and/or solubility values above the critical points (40 μg/mm³ and 7.5 μg/mm³, respectively) should not be used clinically.

CONCLUSIONS

Within the limitations of this in-vitro study, the following conclusions were drawn:

1. The hypothesis tested was rejected, and for that reason the testing of dental materials should be done in artificial saliva instead of distilled water, to mimic the oral environment.

2. Resin-modified glass ionomer (N100) showed the highest and non-acceptable mean values for water sorption in both media, followed by marginal values for compomer (F2000), and flowable composite (SDR).

3. Silorane-based low-shrink composite P90 had the lowest mean value for water sorption in artificial saliva and no sorption in distilled water.

4. Resin-modified glass ionomer (N100) showed the highest solubility and non-acceptable mean values in both media.

5. Microhybrid composite (Z250) showed no solubility in both media.

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REFERENCES


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