ABSTRACT

Water contributes to the setting reaction of self-adhesive luting cements, however, it can also accelerate their degradation. Objectives: The aim of this study was to compare a self-adhesive resin luting cement to other resin-based and glass-ionomer luting materials with regards to water sorption (WS) and solubility (WSB). The tested null hypothesis was that there is no difference in respect to these properties among the materials. Material and methods: Eight specimens from each group (15 mm x 0.5 mm) were prepared from self-adhesive luting cements Biscem (BC) and Enforce (E), chemical-cure cements C&B (CB) and Cement Post (CP) and a glass-ionomer luting cement Meron C (M) as the control group. The dual-cure products were light-cured beneath an IPS Empress Esthetic ceramic disk (20 mm x 1.5 mm) and for the chemical reaction materials, a 15 min-interval was respected for removal from the mould. The WS and WSB were respectively calculated as (m2-m3/V) and (m1-m3/V). Mass values of m1, m2 and m3 were determined by cycles of desiccation, water-immersion and a new desiccation. For each property, the data was analyzed by one-criteria ANOVA and Tukey tests (p < 0.05). Results: The glass-ionomer cement presented the highest WS, followed by the self-adhesive luting cements. Other resin cements were less susceptible to WS. No materials differed from each other when the WSB was considered, except for the M, which presented the lowest WSB. Conclusions: Self-adhesive luting cements were more prone to WS since water is essential to their setting reaction. However, their WSB was similar to the other resin-based cements.

Keywords

Glass-ionomer luting cement; Resin cements; Self-adhesive luting cements; Solubility; Water sorption.
**INTRODUCTION**

A remarkable trend to simplify dental materials is observed in bonding restorative dentistry, which in fact, is greatly represented by simplified dentin bonding systems [1-3]. Many investigations are proposed to analyze their performance, which implies in reduced clinical steps but do not properly represent the best results [2,4]. These products are based on a concept of modification of tooth structures with their acidic content, expecting less sensitive techniques than total etching systems [5]. In the same direction, resin cements have been following the same modifications and a new category of self-etching adhesive resin cements was made available. According to the manufacturers, these materials do not require pretreatment of the tooth and are simple to use [6].

In order to clarify the applicability of these materials, different comparisons of distinct properties are necessary to avoid complications on the selection of a luting cement for clinical service [5-7].

The resin-based luting cements provide high bond strength to the materials and to hard dental tissues, less solubility, great stability and bio-compatibility [8]. However, since the oral environment consists of saliva with organic and inorganic substances, it likely accelerates the degradation over time [9].

Especially in this moist environment, the hydrophilic nature of the polymers determines the level of capacity to absorb water [10]. This feature is due to the chemical constitution of monomers with carbon and oxygen backbones, associated to hydrolytically susceptible ester groups [4].

Moisture diffuses into the polymers at different degrees, depending on a number of molecular and microstructural arrangements, mainly polarity of the molecular structure, presence of chemical groups capable of forming hydrogen bonds with water, degree of cross-linking, presence of residual monomers and/or other water-attracting species, i.e glass surfaces and crystallinity of the polymer (well-defined crystallites are inaccessible to water). As a result among these factors, the mechanism of water diffusion can be summarized in two main theories: (1) the free volume theory, according to which water diffuses through voids within the polymer and (2) the interaction theory, according to which water binds to specific ionic groups of the polymer chain. In this case, water diffusion occurs according to the water-affinity of these groups [2,4,9,11].

It is expected that the absorption of water into polymer materials allow a volume expansion that potentially compensates for the initial setting shrinkage and, consequently, decreases the interfacial gap width and the resultant micro-leakage [12,13].

On the other hand, excessive moisture can adversely affect the hybrid layer durability due to the degradation of either collagen fibrils or resin material [2,14-18].

It is known that the conversion of monomers into a polymer network is never complete for resin-based materials [11]. Clinically, as these materials are frequently light-activated through surfaces such as the dental structure or restorative materials, they act attenuating the necessary energy to polymerize them. As a result, a trend is observed that deteriorates the adhesive interface and decreases the mechanical properties. Significant amounts of unreacted monomers and other additives could be released, compromising the bio-compatibility of the dental restoration products [18-22]. These molecules are potentially hazardous to the pulp tissue [20,21].

Regarding the considerations mentioned above, the predictability in the long term of a resin based luting cement is strongly related to its susceptibility to water sorption and solubility [19]. Since the acid content of self-etching adhesive luting is significant, this property attributed to simplified dentin bonding systems can exacerbate their hydrophilic potential. Moreover, it is expected that water plays an important role in the setting reaction as it occurs to glass-ionomer based materials, which would justify its need.

Thus, this study aimed to compare a self-adhesive resin luting cement to other resin-based and glass-ionomer luting materials regarding water sorption and solubility. The tested null hypothesis was that there is no difference in respect to these properties among the materials.

**METHODS AND MATERIALS**

This in vitro experimental design involved one factor (luting cement) in eight levels. The tested materials are presented in Table 1.
Table 1 – Characteristics of Luting Materials Under Investigation

<table>
<thead>
<tr>
<th>Category</th>
<th>Materials</th>
<th>Manufacturer</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Self-adhesive luting cements</td>
<td>Rely X Unicem</td>
<td>3M ESPE, St Paul, MN, USA</td>
<td>Powder: Glass particles, initiators, silica, substituted pyrimidine, calcium hydroxide, peroxide composite and pigment. Liquid: Methacrylate phosphoric acid ester, dimethacrylate, acetate, stabilizer and initiator.</td>
</tr>
<tr>
<td>Biscem</td>
<td>Bisco Inc., Schaumburg, IL, USA</td>
<td>Base: BisGMA, TEGDMA, EDMAB, dihydroxyethyl-p-toluidine, CQ, MEHQ, BHT, dental glass, silica. Catalyst: Bis (Hydroxyethyl Methacrylate) di phosphate, TEGDMA, benzoyl peroxide, BHT, dental glass, silica.</td>
<td></td>
</tr>
<tr>
<td>Dual-cure resin cements</td>
<td>Allcem</td>
<td>FGM, Joinville, SC, Brazil</td>
<td>Methacrylic monomers, such as TEGDMA and Bis-EMA, inorganic load, photo-initiator, co-initiator, catalysts, and pigments.</td>
</tr>
<tr>
<td>Enforce</td>
<td>Denstply, Rio de Janeiro, RJ, Brazil</td>
<td>Base: TEGDMA, BDMA, boron aluminium glass silicate, pyrolytic silanized silica, canforoquinone, EDAB, BHT, mineral pigments, DHEPT. Catalyst: Titanium Dioxide, pyrolytic silanized silica, mineral pigments, Bis-GMA, EDAB, BHT, TEGDMA, Benzoyl Peroxide.</td>
<td></td>
</tr>
<tr>
<td>Bifix</td>
<td>Voco, Cuxhaven, Germany</td>
<td>Composition information was not supplied by manufacturer.</td>
<td></td>
</tr>
<tr>
<td>Chemical-cure Cements</td>
<td>C &amp; B</td>
<td>Bisco Inc., Schaumburg, IL, USA</td>
<td>Base: BisGMA, Ethoxylated Bisphenol A’ Dimethacrylate, TEGDMA, dihydroxyethyl-p-toluidine, MEHQ, dental glass, silica, sodium fluorode, pigments. Catalyst: BisGMA, TEGDMA, benzoyl peroxide, BHT, silica.</td>
</tr>
<tr>
<td>Cemment Post</td>
<td>Angelus, Londrina, PR, Brazil</td>
<td>Base: Barium Glass Ceramic, silica pyrogenic Bis-GMA, TEDMA, BHT, catalysts, pigments. Catalyst: Barium glass ceramic, pyrogenic silica, Bis-GMA, TEDMA, benzoyl peroxide, stabilizer.</td>
<td></td>
</tr>
</tbody>
</table>


A clearer shade of each material was selected for this study, which does not necessarily mean the same as each manufacturer determines it. This study was performed in compliance with ISO 4049:1988 standard specifications [22], except for the photo-polymerization, as will be described.

For each material, eight specimens (15 mm x 0.5 mm) were prepared, which were directly dispensed into a stainless steel matrix until filled completely, and some excess was extruded. Air bubbles were eliminated and a polyester strip was placed over the luting cement and covered with a glass slide. The dual cure products were light-cured beneath an IPS Empress Esthetic (Ivoclav–Vivadent, Schaan, Liechtenstein) ceramic disk (20 mm x 1.5 mm) and for the chemical cure materials, a 15 min-interval was allowed before its removal from the mold. The polymerized specimens were removed from the matrix and the excesses were eliminated with a scalpel blade.

The specimens were stored in desiccators at 37 °C.
containing silica gel. The discs were weighed daily in an analytical balance (Tel Mark; Bel Quimis, São Paulo, SP, Brazil) accurate to 0.01 mg, constituting a weighing cycle every 24 h. The complete cycle was repeated until a constant mass \((m_1)\) was obtained, i.e., until the mass loss of each specimen was not more than 0.2 mg per 24 h cycle.

Thereafter, the specimens were individually stored in a volume of water for immersion, at least 6 mL at 37 ºC for 7 days. The specimens were weighed again daily after being carefully wiped with absorbent paper (Gala®, Bragança Paulista, SP, Brazil). When the constant weight was obtained, this value was recorded as \(m_2\). After this weighing, the specimens were returned to the first desiccator and the entire mass re-conditioning cycle was repeated and the constant mass was recorded as \(m_3\).

The thickness of each specimen was measured at three different points using a digital electronic caliper (Mitutoyo Corporation, Tokyo, Japan). The values for water sorption and solubility, in micrograms per cubic millimeter, were calculated using the following equations:

\[
WS = \frac{(m_2 - m_3)}{V} \\
WSB = \frac{(m_1 - m_3)}{V}
\]

The assumptions of equality of variances and normal distribution of errors were checked for the tested response variables. Since the assumptions were satisfied, the data was submitted to one-way ANOVA and Tukey post hoc testing \((p < 0.05)\).

**RESULTS**

Water sorption and solubility values (in µg/mm³) are summarized in tables 2 and 3, respectively.

<table>
<thead>
<tr>
<th>Table 2 – Mean and (standard deviations) of water sorption among tested luting cements in µg/mm³</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>MATERIALS</strong></td>
</tr>
<tr>
<td>M</td>
</tr>
<tr>
<td>BC</td>
</tr>
<tr>
<td>R</td>
</tr>
<tr>
<td>CB</td>
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<tr>
<td>BF</td>
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<td>CP</td>
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<tr>
<td>E</td>
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<td>A</td>
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</tbody>
</table>

\(N=8; \) *Different uppercase letter denotes statistical differences among materials

The highest WS was observed for the M and the lowest for the A, as showed in Table 2. The M material was statistically different than all the tested materials \((p < 0.05)\). The self-etching luting cement BC showed the second highest mean values of WS, with no differences when compared to the R, which was the third most susceptible to WS. The R material was not statistically different than the CB, BF and CP. Self-adhesive luting cement BC was statistically more prone to WS when compared to the dual cure cements BF, A, E, and to the chemical cure cements CB and CP.

When compared to the glass-ionomer luting cement, the self-adhesive materials demonstrated higher WS.

According to the values of WSB, M showed the lowest means, which was statistically different than all the other tested materials \((p < 0.05)\), as shown in table 3.

**DISCUSSION**

Since polymerization of light-cured or dual-cured luting cements commonly implies in an interposition of the tooth structure or the restoration material between the cement and the light-activation unit tip, this study was conducted using an usual ceramic material disk in order to reproduce a clinical situation [23,24].

In spite of the features regarding curing, [25] it is important to consider the composition of the materials in their own performance [10,26,27]. Water- sorption is strongly associated to an organic matrix polymeric nature. The curing process can interfere in their polymeric arrangement since more cross-link and
Water sorption is a continuous process as previously stated by Chutinan et al. [18] and Huang et al. [29], which verified an increase of volume of materials submitted to a water sorption test after 56 days. Archegas et al. [34] related greater water sorption when prolonged storage occurs.

The other relevant property analyzed in the present study is related to the solubility of materials in water storage. Leachability of components into the dentinal tubules can provoke inflammation in the dentin pulp complex [20] and bonding interface degradation [4,35].

Only the glass-ionomer luting cement was statistically different from the other tested materials. Glass-ionomer cements allow fluoride lixivia and so solubility is an inherent property of this material category [9,36]. The lower the pH level of the environment, the more fluoride leaching occurs [35].

Negative values of solubility of glass-ionomer cements point out the high capability of water absorption and adsorption, masking its solubility [4,9]. It was previously demonstrated that Meron C, as a glass-ionomer cement, contains water in its composition and consequently the desiccation method applied in this study might not actually represent glass-ionomer sorption and solubility [18,33].

There were no differences among all the resin-based materials. In the RelyX Unicem reaction, fluoride release is expected, however, it did not support any difference in the BisGMA, which did not present fluoride in its composition [37]. Costa et al. [21] verified that RelyX Unicem didn’t release particles into the dentin, suggesting low solubility. Additionally, since it didn’t constituted tags formation, it might not influence a release into the dentinal tubules.

A material that greatly absorbs water does not exactly present high solubility and vice versa. In the present study, the Biscem was more prone to sorption compared to the Rely X Unicem, but the opposite was verified related to solubility even no significant difference was detected. Malacarne et al. [4] reported similar results, since they showed that less hydrophilic resin presented higher solubility compared to more hydrophilic resin.

Self-adhesive luting cements can be attributed to glass-ionomer materials, considering its ability to adhere to tooth structures and due to its acidic composition and calcium content. However, the results of the present study can support values more similar to resin-based materials compared to glass-ionomer cements.

Even though the water sorption of the self-
adhesive materials was greater than the other resin-based materials, it was significantly lower than the glass-ionomer cement. This information is essential to highlight these materials as a particular category and does not erroneously anticipate one to indicate it as a glass-ionomer cement. Although materials prone to water sorption represent a critical point to facilitate degradation, other authors were encouraged to attest that it does not necessarily indicate more clinical fragility, since it is represented by the glass-ionomer cement’s clinical performance [38,39]. In a clinical point of view, a moist environment is the actual oral situation and thus, resin-based materials will be susceptible not only to their own hydrolysis but also to the hybrid layer and collagen fibrils degradation over time [2,29]. Studies such as Viotti et al. [3] recently showed that most self-adhesive luting agents provided low bond strength values on the dentin surfaces, which indicate the need of interaction of the analysis of different properties of these materials to correctly indicate their use or not.

The overall findings apparently suggest that the solubility of self-etching luting cements does not seem to be more hazardous, as indicated, to other resin-based materials. The great water sorption suffered by the tested self-etching luting cements indicates the need for further investigations to verify whether their acidic monomers hydrolytic stability is sufficient, mainly under critical clinical situations. Even though it is high, a previous study demonstrated more stability over time compared to other resin-based cements [8].

**Conclusion**

Self-adhesive luting cements perform differently than resin-based available cements, and are more prone to water sorption. Conversely, they did not inhibit more aggressiveness comparatively to resin-based materials by means of solubility.

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