

# Microleakage and Polymerization Shrinkage of Various Polymer Restorative Materials

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## ABSTRACT

**Purpose:** The aim of this in vitro study was to evaluate the polymerization shrinkage and the microleakage of direct resin-based restorative materials commonly used in pediatric dentistry.

**Methods:** Standardized Class V cavities overlapping the cemento-enamel junction were prepared on the buccal and the lingual surfaces of 40 extracted human mandibular third molars (36 specimens, 4 controls). The cavities were restored with 4 different materials: a packable resin composite (Filtek P60), a compomer (Compoglass F), an ormocer (Admira) and their associated bonding agents (Scotchbond 1, Excite, and Admira Bond, respectively), and a resin-modified glass ionomer (Fuji II LC). The teeth were then immersed in methylene blue solution for 48 hours. Dye penetration was evaluated for all materials, which were analyzed using a multivariate model ( $\alpha=0.05$ ): influence of microleakage score, margin location (enamel/cementum), and preparation location (buccal/lingual). Multivariate analysis was performed using a polychotomous logistic regression. Polymerization shrinkage was evaluated by the disk deflective method. The percentage of polymerization shrinkage ( $N=3$ ) was evaluated by ANOVA and Tukey test.

**Results:** Regarding polymerization shrinkage, the P60 demonstrated the lowest value, followed by ADM and COF, whereas FLC presented the highest shrinkage-strain ( $P<.0001$ ). The preparation location had no significant effect on dye penetration ( $P=.86$ ). Margin location (enamel or cementum) had a significant effect on microleakage (odds ratio [OR]=24.61). Significant differences in the microleakage patterns and scores were also observed between the 4 restorative materials. Admira exhibited the lowest overall microleakage. In comparing Filtek P60, Compoglass F, and Fuji II LC to Admira, P60 showed significantly less microleakage (OR=1.30) than Fuji II LC (OR=1.47), whereas Compoglass F demonstrated the greatest significant overall microleakage (OR=3.15).

**Conclusion:** Within the experimental conditions of this in vitro study, the microleakage was significantly lower at the enamel margins than at the cementum margins for the four restorative materials tested. The ormocer and the packable resin composite exhibited the best sealing ability, as well as the lowest polymerization shrinkage. It could not be demonstrated in this study, however, that the higher the polymerization shrinkage was, the lower the marginal sealing ability was. (J Dent Child 2008;75:125-33)

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Today, resin-based restorative materials (RBRM) are the most frequently used restorative material in both pediatric dentistry and general practice.<sup>1,2</sup> Although these materials present numerous benefits to both practitioners and patients, the conversion of resin monomers into a polymer network leads to bulk contraction and stresses.<sup>3</sup>

The polymerization shrinkage is known to be the main cause of marginal gap formation, subsequent microleakage, and pulpal pathosis.<sup>4</sup>

The sealing ability of RBRM to the calcified tooth tissues is of great significance in many restorative procedures.<sup>5,6</sup> Despite numerous improvements within modern dentin adhesive systems, the bond strength and marginal adaptation of RBRM to dentin remain less predictable than those to enamel.<sup>7</sup> Many restorative cavities, specifically in posterior teeth, present margins located in cementum or in dentin.<sup>8,9</sup> The difficulty in achieving marginal sealing and correct hygiene, especially at the cervical margin of deep cavities, increases marginal microleakage and the subsequent recurrence of caries.<sup>10</sup>

Although no current product satisfies all the requirements of an ideal restorative material, adhesive techniques enable some procedures that cannot be provided with amalgam restorations.<sup>11</sup> RBRM procedures, however, are technique sensitive compared to amalgam restorations. Besides, the restorative material type plays an important role in restoration longevity. The type of resin composite is partly determined by the amount and the size of filler particles.<sup>11</sup> A high amount of filler increases the strength and the elasticity modulus and reduces: polymerization shrinkage<sup>12,13</sup>, coefficient of thermal expansion, and water sorption.<sup>14,15</sup> Recently, a new generation of composites has been proposed with a new filler design, allowing a more efficient packing into cavity preparations.<sup>16</sup> Packable composites have generally larger filler particles, because the resin matrix is chemically modified to allow this slight increase of filler amount.<sup>16</sup>

Another new approach in restorative dentistry has been the introduction of ormocer (organically modified ceramics) in 1998. Instead of bisphenol glycidyl methacrylate (Bis-GMA), urethane dimethacrylate (UDMA) and triethylene glycol dimethacrylate (TEGDMA), multifunctional urethane- and thioetheracrylate alkoxy silanes as sol-gel precursors have been developed for the synthesis of inorganic-organic copolymer ormocer resin composites as RBRM.<sup>17</sup> The hydrolysis and the polycondensation reactions of alkoxy silyl groups allow the formation of an inorganic Si-O-Si network, and the acrylate groups are available for photochemically induced organic polymerization. After incorporation of filler particles, the so-called ormocer composites can be manipulated like hybrid composites.<sup>18</sup>

About 2 decades ago, 2 hybrid restorative material types originated from combining glass ionomer and methacrylate resin technology<sup>19</sup>:

1. The first type includes the resin-modified glass ionomers (RMGI), which resemble the composition of conventional glass ionomers. Resin-modified glass ionomers are said to bond to enamel and dentin with a:
  - a. chemical bonding mechanism identical to conventional glass ionomers; and
  - b. micromechanical bonding similar to that of resin composites.<sup>20</sup> The retention and the marginal sealing capacity could be determined by such a double adhesion mechanism.
2. The second hybrid material type is represented by the polyacid-modified resin composites—so-called compomers. Compomers have close relationship with conventional resin composites and are thought to bond to dentin by micromechanical bonding, as resin composites do.<sup>21,22</sup> Moreover, the fluoride content of these materials might present a cariostatic effect, which would last several weeks.<sup>23</sup>

In vitro evaluations remain an essential method for an initial screening of dental materials and might act as a predictable indicator of in vivo leakage.<sup>24</sup> The primary factors affecting sealing ability and, consequently, clinical durability, are:

1. chemical variations of the tooth substrate<sup>9</sup>;
2. adhesive properties of the materials<sup>5,9</sup>; and
3. differences in the coefficient of thermal expansion of restorative materials with the tooth substrate.<sup>25</sup>

It is known that many different and varied techniques have been used to test the in vitro cavity-sealing ability of restorations. These have included the use of: dyes, chemical tracers, radioactive isotopes, air pressure, bacteria, neutron activation analysis, scanning electron microscopy, artificial caries techniques, and electrical conductivity.<sup>26</sup>

The purpose of this study was to evaluate, via dye penetration and in relation to their polymerization shrinkage, the marginal leakage in Class V cavities of 4 different restorative materials associated with their respective adhesive systems: a packable resin composite, an ormocer, a compomer, and a resin-modified glass ionomer.

## METHODS

### MICROLEAKAGE EVALUATION

The same operator performed all the procedures according to ISO specification 11 405:2003.<sup>27</sup> Forty freshly extracted human third molars, stored for less than 3 months, were selected as study specimens. All gingival remnants were removed, and the crowns were thoroughly cleaned with prophylactic rotary instruments (Screw-in Cups, W&H, Bürmoos, Austria). Before storage, the teeth were examined under binocular microscope (X10, model S2H; Olympus Corp, Tokyo, Japan) to ensure that the specimens were exempt from any decay, cracks, or previous restorations. In order, the teeth were stored in:

1. 0.1% T chloramine (Prolabo, Paris, France) at 4°C for 1 week;
2. distilled water at 4°C for 3 months at most;
3. distilled water at 23°C±1°C during the last 12 hours before use.

**Table 1. Materials Used\***

Code	Material (manufacturer)	Type	Main components	Batch no.
ADM	Admira (Voco D-27457, Cuxhafen, Germany)	Organically modified ceramic (ormocer)	<ul style="list-style-type: none"> <li>Monomers: Bis-GMA, di-UDMA, TEGDMA</li> <li>Fillers (78 wt %=56% vol): Ba-Al-B-silicate glass (90%, ca 0.7µm), SiO<sub>2</sub> (10%)</li> <li>3-dimensionally curing anorganic-organic copolymers, additive aliphatic and aromatic dimethacrylates</li> </ul>	03655
COF	Compoglass F (Vivadent Ets, FL-9494 Schaan, Liechtenstein)	Polyacid-modified resin composite (compomer)	<ul style="list-style-type: none"> <li>Monomers (22.5 wt %): UDMA, poly-EGDMA, CADCADM</li> <li>Fillers (77 wt %): Ba-Al-fluorosilicate glass (1 µm), ytterbium trifluoride, spheroid mixed oxide</li> <li>Additional contents (0.25 wt %): Catalysts, stabilizers, pigments</li> </ul>	B06383
FLC	Fuji II LC (GC Corp, Tokyo, Japan)	Resin-modified glass ionomer	<ul style="list-style-type: none"> <li>Powder: Aluminofluorosilicate glass</li> <li>Liquid: Polyacrylic acid, HEMA, 2-2-4 trimethyl hexamethylene dicarbonate, TEGDMA, water</li> </ul>	Powder 030461 Liquid 080771
P60	Filtek P60 (3M/ESPE dental products, St. Paul, Minn)	Resin composite	<ul style="list-style-type: none"> <li>Monomers: UDMA, TEGDMA, Bis-EMA</li> <li>Fillers (81 wt %=61% vol): ZrSiO<sub>4</sub></li> </ul>	9AN

\* Bis-GMA=Bisphenol-A glycidylmethacrylate; Bis-GA=Bisphenol-A glycidylpolyacrylate; UDMA=urethane dimethacrylate; TEGDMA=triethyleneglycol dimethacrylate; poly-EGDMA=polyethyleneglycol dimethacrylate; CADCADM=cycloaliphatic dicarboxilic acid dimethacrylate; Bis-EMA=ethoxylated bisphenol-A glycol dimethacrylate; HEMA=2-hydroxyethyl methacrylate.

In 4 series of 9 teeth, standardized, nonbeveled Class V (U shaped) cavities were prepared and finished on the buccal and lingual surfaces with 90 µm and 20 µm diamond rotary cutting instruments (no. 802 314 009 and 801 314 023, Komet, Lemgo, Germany) under constant air-water spray. The margins were located:

1. on both sides of the cemento-enamel junction (CEJ);
2. in enamel for the occlusal aspect of the preparation; and
3. in cementum at the gingival margin.

The dimensions of each preparation (mesiodistal width=4.6 mm; occlusogingival height=2.6 mm; pulpal depth=2 mm) were verified with a 10-µm accurate electronic caliper (Digimatic, model no. 500-181U; Mitutoyo Corp, Tokyo, Japan). Between each step of the experiment, the teeth were stored in distilled water at 23°C±1°C. Following the conditioning and priming of the teeth according to the manufacturers' instructions, the cavities were filled with a single increment of 4 different materials:

1. a packable resin composite (Filtek P60 [P60], 3M ESPE Dental Products, St Paul, MN, USA);
2. a compomer (Compoglass F [COF], Vivadent, Schaan, Liechtenstein);

3. an ormocer (Admira [ADM], Voco, Cuxhaven, Germany) and the associated bonding agents Scotchbond 1 (3M ESPE Dental Products, St Paul, MN, USA), Excite (Vivadent, Schaan, Liechtenstein), and Admira Bond (Voco, Cuxhaven, Germany) respectively; and
4. a resin-modified glass ionomer (Fuji II LC [FLC], GC Corp, Tokyo, Japan).

The main components of all the materials are summarized in Tables 1 and 2.

The specimens were randomly assigned to receive a pair of restorative materials according to the possible pairing combinations (P60/COF, P60/ADM, P60/FLC, COF/ADM, COF/FLC, ADM/FLC). The random allocations were carried on by drawing lots. A visible-light polymerizing unit with an irradiating diameter of 9 mm (XL 3000 curing light, 3M/ESPE, St Paul, MN) was used to polymerize the bonding agents and the restorative materials. Light activation energy was controlled at regular intervals to assure a minimum value of 600 mW/cm<sup>2</sup>. The distance of the light tip from the specimens was maintained between 1 and 2 mm.

**Table 2. Bonding Agents Used\***

Code	Material (manufacturer)	Type	Main components	Batch no.
ADB	Admira Bond (Voco D-27457, Cuxhafen, Germany)	ADM	Bis-GMA, HEMA, organic acids, complex 3-dimensionally curing anorganic-organic copolymers, acetone	03653
EXC	Excite (Vivadent Ets FL-9494, Schaan, Liechtenstein)	SOL2	Bis-GMA, HEMA, dimethacrylate phos- phoric acid, acrylate, highly dispersed silica, ethanol (25 wt % )	B33276
SCO	Scotchbond 1† (3M ESPE dental products, St. Paul, Minn)	P60	Bis-GMA, HEMA, Bis-phenol A glycerolate dimethacrylate, copolymer of polyacrylic and polyitaconic acids, water, ethanol	0EE

\* **B is-GMA=Bisphenol-A glycidylmethacrylate; UDMA=urethane dimethacrylate;**  
**HEMA=2-hydroxyethyl methacrylate; 4-META=4- methacryloxyethyl-trimellitic acid.**  
 † **Scotchbond 1=Scotchbond Single Bond in USA**

Immediately following polymerization, the specimens were immersed in distilled water at 23°C±1°C for 24 hours. Polishing was then conducted with flexible disks (Sof-Lex XT Pop On, 3M/ESPE, Seefeld, Germany). The root apices were then sealed with a resin composite (Z100, 3M/ESPE, Seefeld, Germany) without a bonding agent. The teeth were entirely coated with 2 layers of nail varnish (L'Oreal, Paris, France), except for the location of the restorations and 1 mm around the restoration margins. Four teeth were prepared as controls:

- 2 specimens with a filled cavity and entirely coated with nail varnish served as negative controls; and
- 2 specimens with an empty cavity, and without varnish coating, served as positive controls.

All of the specimens were then soaked in a 1% methylene blue dye (Prolabo, Paris, France) for 48 hours. To evaluate dye penetration, after having been rinsed with distilled water, the specimens were sectioned with a 500-µm thick, slow-speed, diamond-coated disk (Isomet-Plus; Buehler, Lake Bluff, Ill) under water coolant. A first section was centered along the mesiodistal axis to separate the buccal and the lingual surfaces. Three 1-mm thick buccolingual sections were then made for each half-specimen: (a) 1 in the center of the restoration; (b) 1 in the mesial margin; and (c) 1 in the restoration's distal margin. The sections were examined on each side under a binocular microscope (X10, model S2H, Olympus Corp, Tokyo, Japan). Dye penetration was measured on the 6 enamel margins and 6 cementum margins, for a total of 12 measurements per cavity. As there were 18 cavities per material, 216 measurements were taken for each restorative material. The degree of dye penetration was identified according to ISO specification 11 405:2003:

- 0=no penetration;
- 1=penetration to the enamel or cementum aspect of the preparation wall;

- 2=penetration to the dentin aspect of the preparation wall, but not including the pulpal floor; and
- 3=penetration including the pulpal floor of the preparation.<sup>27</sup>

Statistical analyses were performed using the statistical software SAS/STAT, (v. 8.2, SAS Institute Inc, Cary, NC). Using a multivariate model ( $\alpha=0.05$ ), the following factors were analyzed: microleakage score, margin location (enamel or cementum), and preparation location (buccal or lingual). The model provided regression coefficients of independent variables (margin location, preparation location, and restorative materials). Odds ratios (OR) expressed the effect, when changing an independent variable, on the probability of having one unit higher score value, holding other variables in the equation model constant (OR are given with the 95% confident limits). Multivariate analysis was performed using an ordinal polychotomous logistic regression.

### **POLYMERIZATION SHRINKAGE**

Polymerization shrinkage was measured with the "deflecting disk" technique used by Watts and Cash.<sup>28</sup> The tested polymer was placed within a 1.64-mm high brass ring—with an internal diameter of 15 mm—and attached with an adhesive to a glass microscope slide, the surface of which had been sandblasted. A flexible glass cover slip (0.1-mm thick) was placed on the ring rim in contact with the tested material. A linear vertical displacement transducer (LVDT GTX 2500, RDP Electronics, UK; sensitivity>0.1 µm) was gently located in contact with the cover slip's upper surface. The temperature of the glass plate and the specimen platform was set to 37°C±0.5°C, and the room temperature was maintained at 23°C±1°C with a relative humidity of 50%.

The light guide initiated the polymerization from below the unset specimen-disk, using a light-curing unit (XL 3000

Curing Light, 3M Dental Products, St Paul, Minn) activated for 60 seconds, for all the samples (N=3 for each material). The light activation energy was regularly controlled to assure a minimum value of 600mW/cm<sup>2</sup>. For each specimen, the cover slip was attracted downwards in an axial way, as shrinkage took place. The displacement of the cover slip was recorded over time. According to Watts and Cash,<sup>28</sup> with the displacement of the disk upper surface being uniform, measurements at the center were representative of the whole. The cover slip displacement was not only recorded during the light activation time, but also for 100 seconds after light activation stopped—for a total duration of 160 seconds.

The shrinkage-strain,  $\epsilon(t)$ , was expressed as a percentage, according to the equation:

$$\epsilon(t)_{\%} = 100 \times \Delta L / L_0$$

where  $L_0$  is the initial specimen height and  $\Delta L$  is the cover slip displacement expressed in  $\mu\text{m}$ . Data were statistically analyzed by 1-way ANOVA and Tukey tests.

## RESULTS

### MICROLEAKAGE

The negative controls showed no evidence of dye penetration, whereas the dye completely penetrated the positive control cavities. The dye penetration data (Table 3) and the polychotomous stepwise logistic regression results (Table 4) are presented in accordance with each element of the study, including the: preparation location, various restorative materials, and margin location.

The buccal or the lingual preparation location on the teeth had no significant influence on the dye penetration

**Table 3. Leakage Scores\***

Variables		Total	Score 0	Score 1	Score 2	Score 3
Materials (code)	<b>Total</b>	<b>864</b>	<b>96</b>	<b>328</b>	<b>115</b>	<b>325</b>
	Admira (ADM)	216	27	101	25	63
	EM N=108		25	73	7	3
	CM N=108		2	28	18	60
	Compoglass F (COF)	216	17	63	26	110
	EM N=108		17	62	9	20
	CM N=108		0	1	17	90
	Fuji II LC (FLC)	216	20	85	41	70
	EM N=108		19	67	17	5
	CM N=108		1	18	24	65
	Filtek P60 (P60)	216	32	79	23	82
	EM N=108		32	61	12	3
	CM N=108		0	18	11	79
Margin location	<b>Total</b>	<b>N=864</b>	<b>96</b>	<b>328</b>	<b>115</b>	<b>325</b>
	Enamel	N=432	93	263	45	31
	Cementum	N=432	3	65	70	294
Preparation location	<b>Total</b>	<b>N=864</b>	<b>96</b>	<b>328</b>	<b>115</b>	<b>325</b>
	Buccal	N=432	46	167	53	166
	Lingual	N=432	50	161	62	159

\* EM=enamel margins; CM=cementum margins.

**Table 4. Factors Associated With the Score Variation (N=864)**

		OR (95% CI)*
Margin location	Enamel	1
	Cementum	24.61 (17.94-33.74)
Materials	ADM	1
	COF	3.15 (2.16-4.58)
	FLC	1.47 (1.03-2.10)
	P60	1.30 (0.91-1.85)

\* OR=odds ratio; CI=confidence interval.

( $P=.86$ ). Conversely, the polychotomous stepwise logistic regression results established that margin location was the strongest and most consistent predictor of increased microleakage (OR=24.61). The statistical model confirmed that microleakage may have been dependant upon the material type. Admira significantly exhibited the lowest overall microleakage. Comparing Filtek P60, Compoglass F, and Fuji II LC to Admira, P60 showed significantly less microleakage (OR=1.30) than Fuji II LC (OR=1.47). Compoglass F, however, demonstrated the greatest significant overall microleakage (OR=3.15). The ordinal polychotomous stepwise logistic regression revealed statistical interactions referring to the score variation between the margin location and the luting cements.

This study confirmed that the occurrence of microleakage is higher at the cementum margin than at the enamel one. The effect of the 4 materials on the microleakage score

**Table 5. Shrinkage-strain Data\***

Materials	Mean apparent shrinkage strain (%) at 37°C±(SD)	
	t=60 seconds	t=160 seconds
Admira	2.64±0.11 <sup>a</sup>	2.82±0.14 <sup>a</sup>
Compoglass F	2.96±0.02 <sup>b</sup>	3.22±0.01 <sup>b</sup>
Fuji II LC	3.25±0.17 <sup>c</sup>	3.46±0.13 <sup>c</sup>
Filtek P60	1.99±0.06 <sup>d</sup>	2.13±0.04 <sup>d</sup>

\* Within datasets for each material, superscript letters indicate homogenous sub-sets (at the 0.05 level).

at the margin locations is presented in Figure 1. In each occurrence, the score values were higher at the cementum margin than at the enamel margin. The increase of microleakage at the cementum margins was significantly greater for COF and for P60 than for ADM and FLC ( $P<.05$ ).

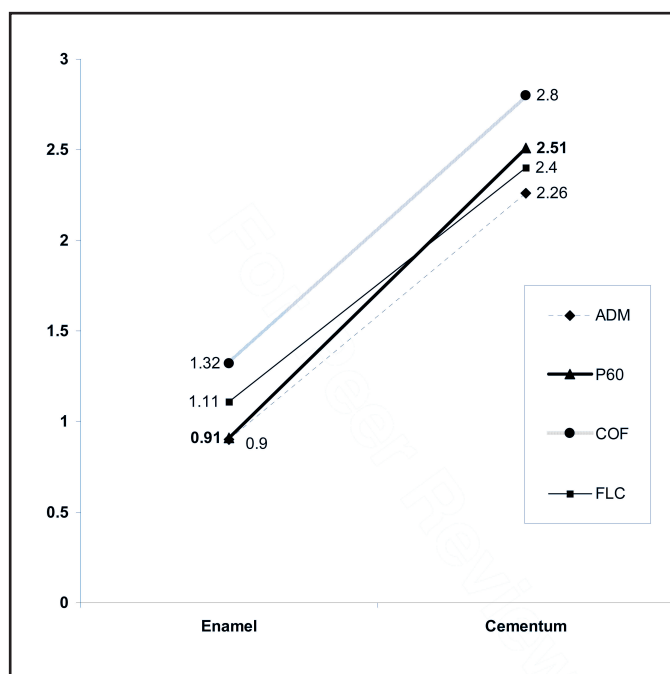
## POLYMERIZATION SHRINKAGE

A representative, time-dependent curve for each material is displayed in Figure 2, and the summary data are given in Table 5. Figure 2 measurements were made at 160 seconds—sufficient duration to obtain a good estimate of the final shrinkage-strain. Statistical analysis established the same value ranking and an equivalent statistical significance at the different shrinkage measurement times (60 seconds, 160 seconds). FLC had the highest polymerization shrinkage ( $P<.0001$ ), whereas P60 showed the lowest shrinkage ( $P<.0001$ ), followed by ADM ( $P<.0001$ ) and COF ( $P<.0001$ ).

## DISCUSSION

According to Mannhart et al,<sup>7</sup> all the restorations selected in this study exhibited interfacial microleakage as was expected.<sup>8,16,21,22</sup> A possible explanation relied upon the adhesive bond, which declined or even broke because of dimensional changes, occurring during polymerization.<sup>29</sup> Therefore, an interfacial gap would occur if the adhesion of the restorative material to the tooth structures did not compensate for the shrinkage stress exerted by the material in the setting's very first setting.<sup>30</sup>

The polymerization of the resin matrix modified the material from a flowing viscous-plastic phase into a rigid-elastic phase—a gel. Resin shrinkage occurred before reaching the gel point, when the monomer-polymer was still flowable. It could be partially compensated by a movement of molecules in the resin composite from the restoration's free surfaces.<sup>29,31</sup> The compensation could not occur after the gelation point, however, and large stresses consequently developed in the RBRM. Thus, polymerization contraction stress might be the primary cause for microleakage when the experimental restorations were not subjected to thermocycling and/or mechanical load cycling<sup>22</sup> as the present study did. Shrink-



**Figure 1. Graph of restorative material effect on microleakage score vs margin locations (N=864).**

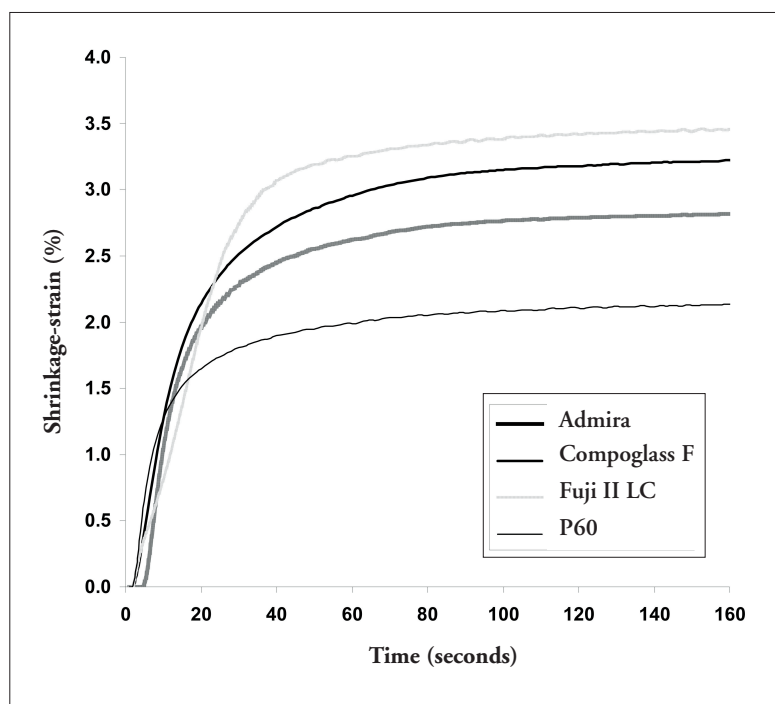


Figure 2. Time-dependence of shrinkage-strain.

age has to be specially considered when a bulk placement technique in Class V cavity was used, because a large bonding area (high C-factor) allowed greater stress development at the adhesive interface.<sup>13,30,31</sup>

Nevertheless, if only contraction stresses determined the extent of microleakage, FLC used in this study was expected to obtain the highest scores of dye penetration and P60 was expected to show the best sealing ability. ADM demonstrated the overall lowest dye penetration scores, however, despite a shrinkage-strain value superior to P60. Also, COF presented the highest leakage scores, when its shrinkage-strain value was inferior to FLC. The good sealing ability of ADM and P60 could be related to their high filler load, which withstands the polymerization contraction stress.<sup>17</sup> In addition, ormocers might undergo less leakage because of their specific 3-dimensional structure and extremely high molecular weight.<sup>15</sup>

Regarding the difference between the marginal leakage of FLC and COF, a major factor might be the bonding ability to tooth structures. At the same time FLC showed the highest penetration score and a less overall leakage than COF, particularly at the cementum margins. On top of an acid-base reaction, COF sets with a free-radical polymerization process. Therefore, the shrinkage stress would be sufficiently enhanced to produce a marginal gap and more subsequent leakage. The setting procedure of FLC is essentially achieved by an acid-base reaction.<sup>19</sup> A polymerization reaction also occurs with the HEMA and urethane dimethacrylate monomers of the resin matrix that produce additional shrinkage.<sup>13</sup> The fact that FLC demonstrated weaker bond strength to both enamel and dentin could explain the high leakage scores.<sup>32</sup> Furthermore, the experimental conditions

themselves contributed to increase FLC microleakage scores.

Despite a constant water-storage at  $23^{\circ}\text{C} \pm 1^{\circ}\text{C}$  between each experimental step, the extracted teeth tissue did not contain sufficient water to prevent dehydration.<sup>33</sup> Thus, microleakage might result from a bonding breakdown and from dye absorption into the layer of porous material—a phenomena which is due to the changes in the physical properties of the material.<sup>34</sup> Actually, methylene blue penetration was observed in this study in both the tooth/material gap and the FLC material. This physicochemical behavior might be explained by: hygroscopic expansion<sup>33</sup>, reduced setting stress owing to water absorption<sup>35</sup>; and improved bonding ability or polymerization during the water storage by the increased value of flexural modulus.<sup>6</sup>

These properties, however, may also compensate for the relative weakness of the bond strength on dental hard tissues.<sup>34</sup> The water sorption mechanism may affect the mechanical properties and decrease the sealing ability of the composite resin materials.<sup>36</sup> In fact, the water solubility of some matrix components could create some modification in the material structure and, subsequently, allow tracer penetration.<sup>23</sup>

This study's results confirmed that polymerization contraction was not the only parameter implicated in the adhesion mechanisms and durability. Other factors have to also be considered:

1. the extent of the marginal gap;
2. varying coefficients of thermal expansion for tooth and restoration materials at the interfacial level<sup>25</sup>;
3. degradation of the bonding joint or of the restorative materials<sup>37</sup>; or
4. the dissolution of liners or smear layers.<sup>25,37</sup>

This study agrees with others that demonstrated a stronger adhesion to enamel than to dentin or cementum.<sup>16,17</sup> The crystalline prismatic structure conferred to enamel an anisotropic behavior. That means that cleavage plan location was perpendicular to the enamel surface and, thus, a high bond strength.<sup>5</sup> Once again, this study clearly indicates that cementum cannot offer sufficient crystalline structure to provide a high micromechanical bond with the RBRM.<sup>38</sup> Consequently, the leakage scores for all the studied materials were lower at the enamel margins than at the cementum margins.

The sealing ability of FLC at the cementum margin was better than that of P60 and COF and might result from a complex adhesion mechanism. The short-time conditioning treatment might remove the smear layer and expose the collagen fibrils up to a  $0.5\text{-}\mu\text{m}$  depth, leaving a large part of the crystal content linked to the collagen fibrils.<sup>19</sup> A chemical bonding—a consequence of the acid-base reaction in the glass ionomer setting reaction—occurred with the calcium of the hydroxyapatite crystals.<sup>19</sup> HEMA

and urethane dimethacrylate monomers might interdiffuse in collagen fibrils, providing a micro-mechanical bond in partly demineralized dentin.<sup>19</sup>

The RBRMs did not chemically bond to tooth structure, when glass ionomers do thanks to the acid-base process. The sealing ability of the RBRMs was said to be mainly based on a micromechanical overlap in the conditioning tooth crystalline structures. The diphenyl sulfone derivative in COF might also have this ability.<sup>39</sup> COF, however, demonstrated higher leakage scores than FLC at the cementum margin.

To be able to establish whether a correlation might exist between this study's results and those previously conducted,<sup>24</sup> this study conformed to the ISO/TS 11405:2003 standard guidelines.<sup>27</sup> The employment of this recognized approach addressed whether in vitro studies are able to reproduce the clinical behavior of dental materials. Some authors claim that in vivo specimens are able to exhibit greater microleakage than in vitro specimens, even if the latter are thermal cycled or mechanically loaded.<sup>22</sup> Others report differences between in vitro and in vivo bond strengths, which are related to microleakage.<sup>5,24</sup>

It is accepted that the present study may have underestimated the actual leakage by comparing a sectioning method with other methods, which screen the entire volume of the preparations.<sup>26</sup> Some authors have indicated that such a 2-dimensional approach may explain the lack of correlation between in vitro microleakage measurements and in vivo performance.<sup>40</sup> Moreover, the ISO 11405 specifications<sup>27</sup> may create unfavorable polymerization conditions for the RMGIC, especially during uncontrolled dehydration conditions. Thus, the microleakage may be overestimated for this material. For these reasons, this experiment has to be completed by evaluating the microleakage after a mechanical loading treatment in an acidic environment. It is hoped that such parameters might better reflect the clinical situation.

## CONCLUSIONS

Within the experimental results of this in vitro study, the following conclusions were drawn:

1. None of the 4 restorative materials prevented dye penetration. For all the materials tested, dye penetration was higher at the cementum margin. This observation suggests that the restorative materials tested must be carefully considered for subgingival preparations.
2. Regarding material types, the ormocer and the packable resin composite exhibited the lowest microleakage scores, followed by the resin-modified glass ionomer. The compomer, however, showed the highest marginal leakage values.
3. Regarding polymerization shrinkage, the packable resin composite demonstrated the lowest values, followed respectively by the ormocer and the compomer; the resin-modified glass ionomer, however, presented the highest values. In this way, ormocers and packable composites appear to be more suitable than compomers and resin-modified glass ionomer cements to

meet long-term requirements. This appears to be a point of major concern, particularly considering the dental care of young patients.

4. This study, however, could not demonstrate that higher polymerization shrinkage resulted in lower marginal sealing ability. Thus, polymerization shrinkage was confirmed not to be the only factor inducing interfacial microleakage.

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