TEN-MONTH IN VITRO STUDY OF MARGINAL ADAPTATION AND MICROPERMEABILITY IN CLASS II RESTORATIONS MADE WITH MATRIX-MODIFIED COMPOSITES

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Abstract

The aim of the present in vitro study was to compare the marginal adaptation and micropermeability of class II restorations with a gingival wall, located 1 mm below the enamel-cementum junction, restored with two conventional dimethacrylate composites (Filtek P60 and Filtek Ultimate), and two innovative composites with a modified polymer matrix (Venus Diamond and Kalore) over ten months of ageing. The marginal adaptation to the proximal margins of the cavity was analyzed and evaluated by the SEM. The dye method was used to assess the micropermeability, and the analysis was performed using a stereomicroscope. The same samples were used for both studies. A correlation analysis was done based on the data to establish the relationships between marginal adaptation and micropermeability.

A statistically significant difference in the estimates for marginal adaptation to the gingival, vestibular, and lingual proximal edges for values of excellent marginal adaptation ($p < 0.005$) was found. F.P60 differed significantly from V. Diamond and F. Ultimate. Kalore is arranged in the middle and does not differ considerably from the above two groups. According to micro permeability, a significant difference was found between F. Ultimate with better results and F.P60 and Kalore ($p < 0.005$). There was a significant correlation ($p = 0.008$) between the excellent scores for marginal adaptation to the edge of the gingival wall and micropermeability. It can be concluded that innovative high-molecular-weight composites do not significantly improve...
marginal adaptation and micropermeability in class II cavities, with a gingival wall located entirely in dentin.

**Key words:** Kalore, Venus Diamond, marginal adaptation, micropermeability

**Introduction.** The application of resin-composite restorations in the molar area is still associated with problems. Clinical data suggest that the most common causes for failure of these restorations are secondary caries and restoration fractures \[^{1,2}\].

The polymerization shrinkage of the organic matrix is among the main problems of composite materials. It generates stress inside the material, the cavity/restoration border, and the tooth structure itself. As a result, forces arise that can separate the restoration from the cavity walls \[^{3,4}\]. This discrepancy allows micropermeability, dental hypersensitivity, secondary caries, damage to the dental pulp. Furthermore, when the gingival wall of the cavity is below the enamel-cement junction, the problem is complicated by more difficult drying of the field, difficult adaptation of the material, and a more unreliable adhesive bond only to the dentin \[^{5}\].

Over the last decade, more diligent and successful work has been done on changing the composition of the polymer matrix of resin composites, dictated mainly by reducing the polymerization shrinkage. One strategy to reduce the polymerization shrinkage involves using high molecular weight monomers in the polymer matrix composition \[^{6}\]. The presence of high molecular weight monomers reduces the initial amount of double bonds, which in the polymerization of the matrix leads to a reduction in bulk shrinkage while maintaining a high level of conversion \[^{7}\]. As a result, monomers such as TEGDMA and Bis-GMA are excluded from the composition of the materials.

The present in vitro study compares the marginal adaptation and micropermeability of class II restorations made with two conventional dimethacrylate composites and two innovative composites with a modified polymer matrix over ten months of ageing.

Working hypothesis: Low-shrinkage composites with modified polymer matrix lead to a reduction of micropermeability. They allow building an acceptable, marginal connection to both the enamel and the dentin proximal margins of the class II cavities.

**Materials and methods.** The materials included in the study are presented in Table 1.

Forty standardized class II cavities (MO/DO) on extracted intact human third molars were prepared. The molars were extracted for orthodontic purpose and collected with patients’ informed consent.

The parameters of the cavities were approximately the following: vestibular-lingual size = 3 mm; medial-distal size = 2 mm; axial size = 5 mm. The gingival
Low shrinkage resin composites, included in the study

<table>
<thead>
<tr>
<th>Material</th>
<th>Organic matrix</th>
<th>Material type and filler loading</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kalore (G.C.)</td>
<td>DuPont monomer; UDMA; dimethacrylate</td>
<td>Nanohybrid contains prepolymerized particles</td>
</tr>
<tr>
<td></td>
<td>co-monomers</td>
<td>Filled 82% wt</td>
</tr>
<tr>
<td>Venus Diamond (Heraeus</td>
<td>TCD-urethane; UDMA</td>
<td>Nanohybrid, Filled 81% wt</td>
</tr>
<tr>
<td>Kulzer)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Filtek P60 (3M-ESPE)</td>
<td>Bis-GMA; TEGDMA; UDMA; Bis-EMA</td>
<td>Condensable, Filled 80% wt</td>
</tr>
<tr>
<td>Filtek Ultimate (3M-ESPE)</td>
<td>Bis-GMA; TEGDMA; UDMA; Bis-EMA</td>
<td>Nanocomposite, Filled 79% wt</td>
</tr>
</tbody>
</table>

The wall was located 1 mm below the enamel-cementum junction. The edges of the cavities were not bevelled.

The cavities were prepared with turbine cylindrical burs with a rounded tip (Z880-140-FG012M-NTI). The vestibular and lingual walls of the cavities were approximately parallel to each other. After preparing the cavities, the teeth were divided randomly with ten (n = 10) cavities per studied material.

All materials were applied after the application of the same three-step adhesive system Optibond FL (Kerr). The adhesive system was used according to the manufacturer’s instructions and after placement of a contoured metal matrix (MetaFix-Kerr) and a wooden interdental wedge.

All materials were applied in 2 mm layers. Each layer was polymerized for 40 s with a diode lamp (Elipar FreelightII; 3M-ESPE). The soft-start mode of photoactivation was used.

After the restoration of the cavities, finishing and polishing were performed. Once the fillings were completed, the teeth were placed in saline for 24 h, then thermocycled every 1000 cycles (5–55 °C for 1 min in each bath with a 30 s interval between immersions/thermocycling apparatus T H E 1100/1200).

After completing the thermal loading, the samples were placed in saline for ten months and stored at room temperature. Every two weeks, the solution was renewed. After ten months, the samples were removed from the solution, dried, and their apexes were sealed with adhesive wax. The restorations were insulated 1 mm from their border with nail polish and stained for 12 h in 2% methylene blue. After removal from the dye, the teeth were washed under running water for 20 min and cleaned.

The samples thus prepared were coated with gold by low vacuum cathodic sputtering (JEOL JFC-1200) and observed on a scanning electron microscope (SEM-JEOL JSM-5510) for direct evaluation of the marginal edges of the restorations. The peripheral connection was examined sequentially for each edge, a 25× magnification field, and a 250× magnification field to detail the SEM image. The images were measured with KLONK Image Measurement 14.1.1.4, Copyright 1528 S. Yantcheva
2013, Klonk Sm Ba. Estimates were presented as a percentage of the total length of the relevant edge. The following ratings were given: 1. Excellent peripheral connection; 2. Hypercontoured restoration in the area of the peripheral connection; 3. Hypocontour of the restoration in the area of the peripheral connection; 4. The gap between the edges of the cavity and the restoration.

When the SEM analysis was completed, the teeth were included into epoxy blocks and cut longitudinally on a Leica SP 1600 microtome (1 mm slice thickness). An assessment of microleakage follows. It was performed on a stereomicroscope (Leica MZ6) at 40×. In this way, three slices are obtained for each sample. The middle section can be observed medially and distally. Thus, a total of four surfaces per sample are analyzed. This method increases the reliability of the micropermeability study.

The penetration level of the dye was assessed on the following scale: 0. no due penetration; 1. due penetration up to 1/3 of the gingival wall; 2. due penetration up to 1/2 of the gingival wall; 3. due penetration over 1/2 of the gingival wall, but the axial wall is not affected; 4. due penetration reaches and covers the axial wall of the cavity.

The results obtained from both studies were statistically processed. The IBM SPSS Statistics statistical package was used for statistical data processing. ANOVA (analysis of variance), Chi-Square Analysis, Post-hoc LSD Analysis were performed.

Correlation coefficients (according to Spearman) between the microleakage and the scores for marginal adaptation to the proximal edges of class II cavities were calculated.

Results. The median values of the results recorded in terms of marginal adaptation to the proximal edges of class II cavities (SEM analysis) are presented in Tables 2 and 3.

A statistically significant difference was found between the materials in terms of marginal adaptation to the edge of the gingival base. The differences were in terms of the following scores: excellent marginal adaptation ($p = 0.005$).

Post-Hoc ($p = 0.003$) analysis showed that F.P60 differed significantly from the group of V. Diamond, F. Ultimate. On the other hand, Kalore is arranged in the middle and does not differ considerably from the above two groups.

There is a statistically significant difference in the estimates for marginal adaptation of materials to the vestibular and lingual proximal edges: excellent marginal adaptation to the vestibular and lingual marginal edge ($p = 0.012 < 0.05$). Post-Hoc analysis showed that F.P60 differed significantly from V. Diamond and F. Ultimate ($p = 0.017$). Kalore is arranged in the middle and does not differ significantly from either of the above two groups.

The micropermeability results are shown in Table 4.

According to the summarized results for each of the materials, the Post-hoc analysis showed that Filtek Ultimate differed significantly from Filtek P60 and
### Table 2
Marginal adaptation to the gingival margin of the class II cavities

<table>
<thead>
<tr>
<th>Material</th>
<th>Values (%)</th>
<th>Excellent peripheral connection</th>
<th>Hypercontour of the restoration</th>
<th>Hypocontour of the restoration</th>
<th>The gap between the edges of the cavity and the restoration</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>n</td>
<td>Median</td>
<td>$\bar{x} \pm SD$</td>
<td>Median</td>
<td>$\bar{x} \pm SD$</td>
</tr>
<tr>
<td>Filtek P60</td>
<td>10</td>
<td>66.31</td>
<td>4.65 ± 10.91</td>
<td>54.58 ± 29.27</td>
<td>0</td>
</tr>
<tr>
<td>Filtek Ultimate</td>
<td>10</td>
<td>46.54</td>
<td>41.61 ± 27.04</td>
<td>3.82</td>
<td>20.50 ± 32.51</td>
</tr>
<tr>
<td>Kalore</td>
<td>10</td>
<td>25.23</td>
<td>25.14 ± 20.33</td>
<td>30.23</td>
<td>35.65 ± 29.23</td>
</tr>
<tr>
<td>Venus Diamond</td>
<td>10</td>
<td>34.44</td>
<td>34.29 ± 29.96</td>
<td>29.44</td>
<td>39.58 ± 35.58</td>
</tr>
</tbody>
</table>

### Table 3
Marginal adaptation to the vestibular/lingual margins of the class II cavities

<table>
<thead>
<tr>
<th>Material</th>
<th>Values (%)</th>
<th>Excellent peripheral connection</th>
<th>Hypercontour of the restoration</th>
<th>Hypocontour of the restoration</th>
<th>The gap between the edges of the cavity and the restoration</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>n</td>
<td>Median</td>
<td>$\bar{x} \pm SD$</td>
<td>Median</td>
<td>$\bar{x} \pm SD$</td>
</tr>
<tr>
<td>Filtek P60</td>
<td>10</td>
<td>36.12</td>
<td>36.54 ± 8.49</td>
<td>48.20</td>
<td>40.14 ± 26.72</td>
</tr>
<tr>
<td>Filtek Ultimate</td>
<td>10</td>
<td>56.86</td>
<td>56.65 ± 29.88</td>
<td>21.60</td>
<td>19.52 ± 19.26</td>
</tr>
<tr>
<td>Venus Diamond</td>
<td>10</td>
<td>56.87</td>
<td>56.52 ± 10.85</td>
<td>19.85</td>
<td>22.48 ± 19.53</td>
</tr>
</tbody>
</table>


**Table 4**

Results of micropermeability evaluation

<table>
<thead>
<tr>
<th>Materials</th>
<th>Results</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>Filtek P60</td>
<td>40</td>
<td>5</td>
</tr>
<tr>
<td>Filtek Ultimate</td>
<td>40</td>
<td>7</td>
</tr>
<tr>
<td>Kalore</td>
<td>40</td>
<td>6</td>
</tr>
<tr>
<td>Venus Diamond</td>
<td>40</td>
<td>6</td>
</tr>
<tr>
<td>$\chi^2 = 40.472$</td>
<td></td>
<td>Kruskal Wallis, $p = 0.005$</td>
</tr>
</tbody>
</table>

Kalore ($p < 0.001$). Venus Diamond did not differ statistically significantly from any of the materials ($p = 0.179$).

The four microleakage values for each sample of each material were averaged. The correlation coefficients (according to Spearman) between the microleakage and the excellent scores for marginal adaptation to the proximal edges of class II cavities were calculated. There is a significant correlation ($p = 0.008$) between the excellent scores for marginal adaptation to the edge of the gingival base and micropermeability: correlation coefficient = $-0.338$ ($R = -0.443$, $p < 0.001$).

There are no significant correlations between the excellent scores for vestibular and lingual proximal margin and microleakage to the axial wall of the cavities.

**Discussion.** We chose the II class of direct composite restorations, as they have the most problems in marginal adaptation and stability over time. To avoid the questionable significance of cervical enamel on the integrity of the composite adaptation to the gingival margin, the cavities included in the present in vitro study were prepared with a gingival wall 1 mm apically from the enamel-cementum junction. The adhesive bond to the dentin is weaker than that to the enamel due to the higher organic content of the dentin tissue, moisture from a dentinal fluid; orientation of the dentinal tubules; the presence of odontoblastic growths; difficult elimination of the contaminant layer [8,9].

The importance of marginal adaptation to the dentinal gingival margin is crucial for limiting micropermeability. When searching the correlations between marginal adaptation and the level of micropermeability in class II cavities, we found that the materials in which more excellent scores for the marginal gingival edge: F. Ultimate and V. Diamond, showed lower levels of micropermeability. There is a significant correlation between the excellent scores for the edge of the gingival wall and the micropermeability. Following the correlation coefficients between the estimates for the presence of gaps (open peripheral connection) and the degree of micropermeability, we do not find significant correlations anywhere.
Analyzing the results obtained from the present study, we can conclude that the continuous, excellent marginal seal of the gingival margin in class II cavities limits the micropermeability to the axial wall. This conclusion is in line with the previous six-year, parallel in vitro/in vivo study \cite{10}, which establishes a correlation between laboratory and clinical studies results. Furthermore, it demonstrates that the continuous contour of class II restorations is crucial for their clinical durability.

The analysis of the present study results leads to the rejection of the initial hypothesis, namely that the matrix-modified composites will lead to better marginal adaptation and lower micropermeability of class II restorations. A significant difference was found in terms of excellent results for marginal adaptation to the edges of the gingival margin, located entirely in dentin only between F P60 (conventional dimethacrylate composite) on the one hand, and a heterogeneous group of materials – V. Diamond and F. Ultimate, on the other, as well as for excellent values relative to the enamel edges of the cavity, where F.P60 again shows significantly more unsatisfactory results than V. Diamond and F. Ultimate. Among the materials that showed significantly better results is F. Ultimate, which has a conventional dimethacrylate matrix.

Filtek P60 and Filtek Ultimate are composites with the same polymer matrix. They were control materials in the present study. They were selected to see the influence of the inorganic phase on the sealing properties of the materials. F. Ultimate is a nanocomposite. The nanocomposites were created to reduce the polymerization shrinkage (up to 1.92%) and increase the fillings’ mechanical stability and aesthetics \cite{2}. F. Ultimate has round-shaped filler particles, which, according to some authors, further contribute to reducing the polymerization stress \cite{11}. A study by Santos et al. \cite{12} analyses the properties of marginal adaptation of Filtek Ultimate and Filtek P60 in cavities with different C-factor. The study shows that despite the exact composition of the matrix differences in the filling phase and modulus of elasticity (11.7 GPa Filtek P60/10.5 GPa Filtek Ultimate), different behaviours concerning polymerization stress and gap formation. The nanocomposite with a lower modulus of elasticity generates less stress and leads to more minor marginal defects \cite{12}. The lower level of polymerization stress and the structure of the inorganic filler phase are a possible explanation for the registered satisfactory, marginal adaptation of Filtek Ultimate in the present study. The nanomaterial present in the materials showed the most excellent results for marginal adaptation to both the enamel and dentin edges of the cavity. The material has the most excellent values registered in the area of the dentinal gingival margin, which leads to lower micropermeability in the direction of the axial wall of the cavities.

Venus Diamond is a low-shrinkage nanohybrid composite based on TCD-urethane. TCD-urethane is a high-molecular-weight, low-viscosity methacrylate monomer that does not require the addition of density-correcting agents, such
as those used in Bis-GMA composites, and leads to increased shrinkage. The polymerization shrinkage reduction is 1.6–1.7% \cite{13,14}, a more significant achievement than materials based on classical methacrylate chemistry. The most important advantage of the material is the reduced level of polymerization stress – 2.8 MPa \cite{13,15}. Stress is reduced compared to other low-shrinkage dimethacrylate composites. According to researchers, this is due to the increased elasticity of the TCD molecule and the slower polymerization, leading to stress relaxation \cite{16}. In addition, the polymer matrix structure allows reducing the modulus of elasticity of the material despite the high content of inorganic filler (64 vol.%) \cite{13}.

In the present study of the peripheral connection in class II cavities, V. Diamond showed relatively better marginal adaptation to the vestibular and lingual peripheral enamel edges (56%) than the dentinal edge of the gingival base (34%). Despite reduced contraction stress, no significant improvement in marginal adaptation is observed. Nevertheless, the material is in the groups with better marginal adaptation than the condensable conventional material F.P60. Scientific laboratory studies of the marginal adaptation of V. Diamond and other established dimethacrylate composites \cite{14} confirm the better adaptation of TCD-material to enamel than to dentin. The recorded values are similar to those reported for the nanocomposite (Filtek Supreme) \cite{14}.

Kalore (G.C.) is an example of a composite material whose matrix is a polymer structure containing UDMA (urethane dimethacrylate) and the high molecular weight methacrylate monomer DX-511 \cite{15,16}. The shrinkage during polymerization is 1.7–1.8% \cite{16,17}. The generated stress is 1.92 MPa \cite{18}. Kalore has prepolymers in its filling phase. They further reduce the compressive strength and increase the hardness, pressure resistance, and the possibility of good polishing \cite{19}.

The material does not differ statistically with an excellent peripheral connection either from the materials with a conventional matrix (Filtek P60, Filtek Ultimate) or the other innovative composite Venus Diamond. Similar results are present in the scientific literature \cite{20}.

Other researchers stated that reduced shrinkage at the expense of a lower concentration of reactive groups in Kalore is not as appropriate as the change in a matrix made in V. Diamond \cite{18}. Evidence of this is the weaker physicomechanical properties of Kalore compared to those of V. Diamond and the established dimethacrylate hybrid material (Filtek Z250). The results of the present study on marginal adaptation and related micro permeability complement these views.

In Kalore and V. Diamond, the increase in the molecular weight of the organic matrix and the increase in the proportion of filler particles led to increased viscosity of the materials. From a clinical point of view, this hinders the good marginal adaptation of the composite materials to the walls and corners of the cavities. The material density can be a crucial factor for good adaptation to the cavity elements \cite{19,20} despite the reduced shrinkage and reduced stress levels.
Based on the results and limitations of the present in vitro study, the following conclusions can be made:

1. Innovative high-molecular-weight composites do not lead to a significant improvement in marginal adaptation and micropermeability in class II cavities, with a gingival wall located entirely in dentin.

2. There is a significant correlation between the excellent marginal adaptation to the edge of the gingival wall, located entirely in dentin, and the micropermeability at the direction of the axial wall of the class II cavities.

REFERENCES


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