Evaluation of Surface Roughness of Microhybrid and Nanofilled Composites after pH-Cycling and Simulated Toothbrushing

Roberta Alves Pinto Moura Penteado, DDS, MSc; Josealdo Tonholo, MSc, PhD; José Ginaldo Júnior, MSc, PhD; Milton Fernando de Andrade Silva, DDS, MSc, PhD; Celso da Silva Queiroz, DDS, MSc, PhD; Vanessa Cavalli, DDS, Ms, PhD; Marcos Augusto do Rego, DDS, Ms, PhD; Priscila Christiane Suzy Liporoni, DDS, MSc, PhD

Abstract

Aim: This study evaluated the surface roughness patterns of two resin-based composite restorative materials, a microhybrid (Filtek Z250, 3M ESPE) and a nanofilled (Filtek Supreme, 3M ESPE), subjected to a regimen that simulated dynamic pH-cycling and toothbrushing.

Methods and Materials: Twelve standardized cylindrical specimens of each resin-based composite material were prepared, finished, and mechanically polished. The experimental units were submitted to a pH-cycling regimen followed by 50,000 toothbrushing cycles, after which the surface roughness was measured using an atomic force microscope (AFM). AFM surface roughness was evaluated at three intervals: (1) immediately after specimen preparation (baseline), (2) after pH-cycling, and (3) after simulated toothbrushing. The results were then analyzed using a split-plot design and followed by linear regression and a Tukey’s test at a significance level of \( p < 0.05 \).

Results: The results obtained indicated that simulated toothbrushing provoked a remarkable increase in surface roughness for both types of composite resins tested \( (p=0.0031) \). However, pH-cycling did not alter the surface of the composite under the conditions of this experiment.

Conclusions: Based on the results obtained, it was concluded that simulated toothbrushing was capable of increasing the surface roughness of the microhybrid (Filtek Z250) and the nanofilled (Filtek Supreme) composites tested.

Clinical Significance: Surface roughness of nanofilled and microhybrid composites is significantly increased after toothbrushing, although pH-cycling, as tested in this study, does not appear to affect the morphology of either composite material.

Keywords: Composite resin, pH-cycling, surface roughness, toothbrushing

Introduction

Some of the aims of contemporary research of resin-based composites are to identify and overcome any limitations in the materials and then refine their mechanical properties. Wear is among those recognized properties, and is a consequence of cyclic loading during regular
occlusal and masticatory function. It also is a factor that can contribute to composite failure.

The wear of composites depends on the characteristics of the inorganic filler particles, particularly on the concentration and size of those particles. The percentage composition of inorganic filler content is directly related to an increase in the compressive strength, hardness, flexural strength, and elastic modulus and a decrease of polymerization shrinkage of the filler. Therefore, the size, shape, and percentage of inorganic fillers provide resin-based composites with rather specific, desirable mechanical properties.

The average size of the filler particles in a microfilled composite is approximately 0.04 µm, whereas in microhybrid composites the particle sizes may range between 0.01 and 2.0 µm. Recently, new filler materials with sizes between 5 and 100 nm have been developed. Nanotechnology applied to resin composites is aimed toward the production of composite resins with improved mechanical and aesthetic characteristics attributed to the reduced size and wide distribution of the fillers. These nanofilled composites also possess differences in their organic formulation, which may lead to distinct mechanical performance. The reduced size and wide distribution of the nanofillers may increase filler load and, consequently, improve the mechanical properties of these new materials, such as their polymerization shrinkage, tensile strength, compressive strength, resistance to fracture, and reduced wear. It has been observed that nanocomposites promote translucency and polish, and retain that polish similar to microfilled composites but with physical properties and wear resistance equivalent to those of hybrid or universal composites. Conversely, there are reports that indicate nanofilled composites are not likely to improve wear and fatigue performance over the traditional composite resins.

Although wear contributes greatly to material failure, in an oral environment chemical degradation also damages composites because alcoholic and acidic solutions may actually remove portions of the polymer matrix. The acids produced by bacterial metabolism lead to pH variations, which, in turn, also result in the degradation of resin-based materials.

Restorative materials are not only subjected to masticatory forces, occlusal forces, and attack by chemical acids. They also are subjected to the potentially harmful effect of toothbrushing, which may be held responsible for the wear and surface roughening of these resin-based materials. Most in vitro studies simulate toothbrushing under rather specific, standardized conditions, and several authors have shown that this is an effective method of evaluating the wear resistance of different restorative materials.

In order to determine the surface roughness of composite resins, atomic force microscopy (AFM) has become an important tool for imaging surfaces and analysis. An AFM scans the material surface to provide a topographic image revealing the subnanometer resolution and microroughness features of composites. In addition, AFM offers quantitative data on surface morphology.

In light of the promising features of nanofilled composites compared to most other composite resins and the oral conditions these materials must withstand, this study was designed to assess the effects of pH-cycling and toothbrushing on the surface of both microhybrid and nanofilled composite resin restorative materials.

**Methods and Materials**

**Experimental Design**

Two experimental groups consisted of 12 cylindrical specimens for each composite. The factors to be assessed were (1) resin composite type (Table 1): Filtek Z250 and Filtek Supreme; and (2) time: initial (T0), after pH-cycling (Tc), and after simulated toothbrushing (Tb). Both resin products were subjected to pH-cycling and simulated toothbrushing. The response variable was roughness (µm) measured by atomic force microscopy.

**Specimen Preparation**

Cylindrically shaped specimens (n=12) for both composite resins were prepared in a Teflon mold (6.0-mm diameter and 2.0-mm thickness) that was filled in a single increment with either composite Filtek Z250 or Filtek Supreme (3M/ESPE, St. Paul, MN, USA) and covered with a polyester matrix. The top surfaces were cured for 40 seconds with a visible-light curing unit device, Optilux 401 (Demetron Kerr, Danbury, CT, USA) operating...
between 700 and 800 mW/cm². Immediately after irradiation, specimens were retrieved from the mold, wrapped in gauze, and immersed in 3 mL of deionized water at 37°C for 24 hours. Afterwards, the specimens were mechanically polished with fine and then superfine Sof-Lex (3M ESPE) discs for 15 seconds in a single direction.

**Atomic Force Microscopy (AFM)**
Composite surface roughness was determined by AFM (SPM 9500 J3, Shimadzu Corp, Tokyo, Japan) limited to a 125-µm² scanning area (x-y directions) and 8-µm height displacement (z-range) through the contact mode, in which a sharp pyramid-shaped silicon nitride tip scans the surface with a light, constant load. Probes of Si₃N₄ each with a 200-µm-long lever, 0.15-N m⁻¹ spring constant, and a resonance frequency of 24 kHz were furnished by Olympus. The acquisition parameters were a scan frequency of 1 Hz with a contact mode and x-y scan of 50 µm. As a result, an image was produced in gray shades: the highest peaks corresponded to light gray and the valleys to dark gray. In this study, three randomly selected subareas of each specimen (50 µm × 50 µm) were scanned and the mean roughness average (Ra) was determined and expressed in nanometers at the baseline (T₀), after pH-cycling (Tₐ), and finally after toothbrushing (Tₜ).

**Statistical Analysis**
The assumptions of equality of variances and normal distribution of errors were checked with Hartley and Shapiro-Wilk tests for the response variable data roughness average (Ra). The assumptions were satisfied for Ra, and two-way analysis of variance (ANOVA) and Tukey tests (α=0.05) were applied to identify differences among levels. The analyses were performed with the SAS System 6.11 software (SAS Institute Inc., Cary, NC, USA).
Results

Roughness Surface
No significant interaction was noted between resin-based composites and time (p=0.61) or between Filtek Z250 and Filtek Supreme regarding surface roughness at the three test intervals (p=0.0031) (Table 2).

There was, however, a significant increase in surface roughness for both products after simulated toothbrushing (p=0.0031). But no significant changes in resin composite were observed after pH-cycling (Tc) when compared to T0 of both groups (Table 2).

AFM
The AFM image of Filtek Z250 (Figure 1) and Filtek Supreme (Figure 2) at the baseline (T0) showed surface smoothness when compared to Tc (Figures 3 and 4) and Tb (Figures 5 and 6). Especially after brushing (Tb), the resin composite appeared to possess an uneven and irregular surface with attenuated differences between peaks and valleys (Figures 5 and 6).

Discussion
In the present study, AFM was used to quantitatively determine the surface roughness of two composite resins, composed of different organic and inorganic compounds, that were subjected to pH-cycling and simulated toothbrushing. AFM studies were performed to obtain information about surface morphology and smoothness. Furthermore, this technique provides a true, three-dimensional surface profile, and no special specimen preparation is required. Therefore, this method of analysis will neither damage nor irreversibly change the sample.19

Table 2. Mean average roughness (ηm) and standard deviations (sd) of resin-based composites at the baseline (T0), after pH-cycling (Tc), and after simulated toothbrushing (Tb).

<table>
<thead>
<tr>
<th>Resin-Based Composites</th>
<th>Three Time Intervals</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>T0</td>
</tr>
<tr>
<td>Filtek Z250</td>
<td>55.8 (13.2)</td>
</tr>
<tr>
<td>Filtek Supreme</td>
<td>55.2 (6.4)</td>
</tr>
</tbody>
</table>

The means followed by different letters are significantly different based on Tukey's test (p=0.0031). Capital letters compare differences between groups (columns) and lowercase letters compare differences in time (lines).

Table 2. Mean average roughness (ηm) and standard deviations (sd) of resin-based composites at the baseline (T0), after pH-cycling (Tc), and after simulated toothbrushing (Tb).

Figure 1. AFM image of Filtek Z250 resin composite at the baseline (T0). Before treatment, the surface of this composite appears regular with no peaks or deep valleys detectable.

Figure 2. AFM image of Filtek Supreme resin composite at the baseline (T0). Before pH-cycling and brushing, the surface is even with imperceptible irregularities.
that prior to abrasion, pH-cycling, deionized water, and artificial saliva did not change the surface roughness of resin composites. The pH-cycling used by these authors was the same as that used in the present investigation. This outcome also may indicate that the cycling period of 10 days was too short to promote polymer degradation or hydrolysis. It is important to note that the composition of the remineralizing (pH=7.4) and demineralizing (pH=4.3) solutions may not affect the composite bulk, as would concentrated acids or very alkaline solutions.13

Polymer-based materials can degrade through hydrolysis or oxidation processes, and pH levels can change organic composition through the hydrolysis of the ester groups in the matrix.11,13 Hydrolysis of these ester bonds promotes the formation of free, carboxylic acid groups that can lower the pH level inside the polymeric matrix.11,13 This chemical degradation most likely did not occur in the present study because no surface changes after pH-cycling were observed for either composite resin product. In support with our findings, Turssi et al20 reported that prior to abrasion, pH-cycling, deionized water, and artificial saliva did not change the surface roughness of resin composites. The pH-cycling used by these authors was the same as that used in the present investigation. This outcome also may indicate that the cycling period of 10 days was too short to promote polymer degradation or hydrolysis.

The simulation of acid-challenge and toothbrushing is based on the fact that these are clinical and ordinary circumstances in which resin composites may wear.14,16 Polymer-based materials can degrade through hydrolysis or oxidation processes, and pH levels can change organic composition through the hydrolysis of the ester groups in the matrix.11,13 Hydrolysis of these ester bonds promotes the formation of free, carboxylic acid groups that can lower the pH level inside the polymeric matrix.11,13 This chemical degradation most likely did not occur in the present study because no surface changes after pH-cycling were observed for either composite resin product. In support with our findings, Turssi et al20 reported that prior to abrasion, pH-cycling, deionized water, and artificial saliva did not change the surface roughness of resin composites. The pH-cycling used by these authors was the same as that used in the present investigation. This outcome also may indicate that the cycling period of 10 days was too short to promote polymer degradation or hydrolysis. It is important to note that the composition of the remineralizing (pH=7.4) and demineralizing (pH=4.3) solutions may not affect the composite bulk, as would concentrated acids or very alkaline solutions.13

Simulated toothbrushing was performed after pH-cycling, whereby specimens were subjected to 50,000 brushing strokes. Brushing promoted
concluded that, for a fixed volume fraction of filler, a reduced filler particle size results in less interparticle spacing, the improved protection of resin matrix, and less filler plucking. All of these features enhance the wear resistance of the resin-based material. Hence, according to the manufacturers and researchers, it is likely that nanofilled composites would resist occlusal loads and may provide an extra esthetic advantage due to their nanofill particles.

**Conclusion**

Simulated intraoral conditions including pH-cycling and toothbrushing showed that nanofilled and microhybrid composite surface roughness was not altered by pH-cycling but changed remarkably following mechanical toothbrushing. But no differences were found between the microhybrid composite resin, Filtek Z250, and the nanofilled composite, Filtek Supreme, compared in this study.

**Clinical Significance**

The surface roughness of nanofilled and microhybrid composites is significantly increased after toothbrushing, although pH-cycling, as tested in this study, did not appear to affect the surface morphology of either composite material.

**References**


About the Authors

Roberta Alves Pinto Moura Penteado, DDS, MSc
Dr. Penteado is a professor of Operative Dentistry at the Federal University of Alagoas, Maceio, Brazil.

Josealdo Tonholo, MSc, PhD
Dr. Tonholo is an associate professor of the Chemistry Division at the Federal University of Alagoas, Maceio, Brazil.

José Ginaldo Júnior, MSc, PhD
Dr. Ginaldo Jr is an associate professor of the Chemistry Division at the Federal University of Alagoas, Maceio, Brazil.

Milton Fernando de Andrade Silva, DDS, MSc, PhD
Dr. Silva is an associate professor of the Dentistry Department at the Federal University of Alagoas, Maceio, Brazil.
Celso da Silva Queiroz, DDS, MSc, PhD

Dr. Queiroz is an associate professor of the postgraduate program, Cariology Division, at Paulista University, São Paulo, Brazil.

Vanessa Cavalli, DDS, Ms, PhD

Dr. Cavalli is an associate professor in the Operative Dentistry Division at the University of Taubaté, São Paulo, Brazil. She is a member of the International Association for Dental Research (IADR) and the IADR Brazilian division (SBPqO).

Marcos Augusto do Rego, DDS, Ms, PhD

Dr. Rego is an associate professor in the Operative Dentistry Division at the University of Taubaté, São Paulo, Brazil. He is a member of the International Association for Dental Research (IADR) and the IADR Brazilian division (SBPqO).

Priscila Christiane Suzy Liporoni, DDS, MSc, PhD (Corresponding Author)

Dr. Liporoni is currently chair of the Operative Dentistry Division at the University of Taubaté, São Paulo, Brazil. She is a member of the International Association for Dental Research (IADR) and the IADR Brazilian division (SBPqO).

e-mail: prili@yahoo.com