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

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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

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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.[1] Wear is among those recognized properties, and is a consequence of cyclic loading during regular occlusal and masticatory functiond. It also is a factor that can contribute to composite failure.[2]

The wear of composites depends on the characteristics of the inorganic filler particles, particularly on the concentration and size of those particles.[3] 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.[4] Therefore, the size, shape, and percentage of inorganic fillers provide resin-based composites with rather specific, desirable mechanical properties.[5]

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.[6] Nanotechnology applied to resin composites is aimed toward the production of composite resins with improved mechanical and esthetic characteristics attributed to the reduced size and wide distribution of the fillers.[5] These nanofilled composites also possess differences in their organic formulation, which may lead to distinct mechanical performance.[7] 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.[8] 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.[7] Conversely, there are reports that indicate nanofilled composites are not likely to improve wear and fatigue performance over the traditional composite resins.[1]

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.[9–13] The acids produced by bacterial metabolism lead to pH variations, which, in turn, also result in the degradation of resin-based materials.[13]

Restorative materials are not only subjected to masticatory forces, occlusal forces, and attack by chemical acids,[10,14,15] 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.[10,14,15] 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.[16]

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.[17]

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 (T_0), after pH-cycling (T_c), and after simulated toothbrushing (T_b). Both resin products were subjected to pH-cycling and simulated toothbrushing. The response variable was roughness (ηm) 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

Table 1. The dental resin-based composites tested.

Resin-Based Composites	Type	Basic Composition*	Inorganic Filler Level (wt%)	Average Particle Size	Manufacturer
Filtek Z250	Microhybrid	Zirconia, silica, Bis-GMA, UDMA, Bis-EMA	82	0.01–3.5 µm	3M ESPE Dental Products, St. Paul, MN, USA
Filtek Supreme	Nanofilled	Zirconia/silica cluster filler, Bis-GMA, Bis-EMA, UDMA, TEGDMA	52	40 ηm	3M ESPE Dental Products, St. Paul, MN, USA

*As disclosed by the manufacturer. Bis-GMA = bisphenol glycidyl methacrylate; UDMA = urethane dimethacrylate; Bis-EMA = bisphenol A polyetheylene glycol diether dimethacrylate; TEGDMA = tetraethyleneglycol dimethacrylate.

between 700 and 800 mW/cm^2. 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.

pH-Cycling

In order to simulate oral conditions, a pH-cycling model was chosen[18] because it is suitable for replicating acid-challenge conditions. The specimens of both groups were submitted to demineralization-remineralization cycles at 37°C. Each cycle was comprised of 6 hours of immersion in 5 mL of demineralizing solution followed by 18 hours of immersion in 5 mL of a remineralizing solution. The demineralizing solution was composed of 74 mM acetate buffer at pH 4.3 containing 2.0 mM calcium and 2.0 mM phosphate. The composition of the remineralizing solution was 20 mM TRIS buffer at pH 7.4 containing 1.5 mM calcium, 0.9 mM phosphate, and 150 mM KCl. The re-demineralization cycle was performed over a period of 10 consecutive days.

Simulated Toothbrushing

The specimens of both groups were subjected to 50,000 cycles in a simulated toothbrush machine (MSEt machine, Federal University of Alagoas, Maceio, Brazil) with the cycle speed set at 374 strokes per minute and 200 grams of load/weight. Soft nylon bristle toothbrush heads (Sanifill Leader Vip, Facilit Industry, Curitiba, PR, Brazil) were fixed to the brushing machine and replaced after 25,000 cycles. A toothpaste slurry was prepared from Colgate toothpaste (Maximum Anti-Cavity Protection, Colgate-Palmolive, Osasco, SP, Brazil) and distilled water (1:3 w/v). This slurry was injected onto the composite resin samples at a rate of 0.4 ml/min. Afterwards, the specimens were removed from the toothbrushing machine and ultrasonically cleaned with water for 10 minutes.

Atomic Force Microscopy (AFM)

Composite surface roughness was determined by AFM (SPM 9500 J3, Shimadzu Corp, Tokyo, Japan) limited to a 125-µm^2 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 Si_3N_4 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 (T_0), after pH-cycling (T_c), and finally after toothbrushing (T_b).

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 (T_c) when compared to T_0 of both groups (Table 2).

AFM

The AFM image of Filtek Z250 (Figure 1) and Filtek Supreme (Figure 2) at the baseline (T_0) showed surface smoothness when compared to T_c (Figures 3 and 4) and T_b (Figures 5 and 6).

Especially after brushing (T_b), 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 (T_0), after pH-cycling (T_c), and after simulated toothbrushing (T_b).

Resin-Based Composites	Three Time Intervals			
	T_0	T_c	T_b	
Filtek Z250	55.8 (13.2)	51.8 (18.2)	228.7 (21.8)	A
Filtek Supreme	55.2 (6.4)	52.2 (11.0)	237.1 (33.7)	A
	B	B	a	

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).

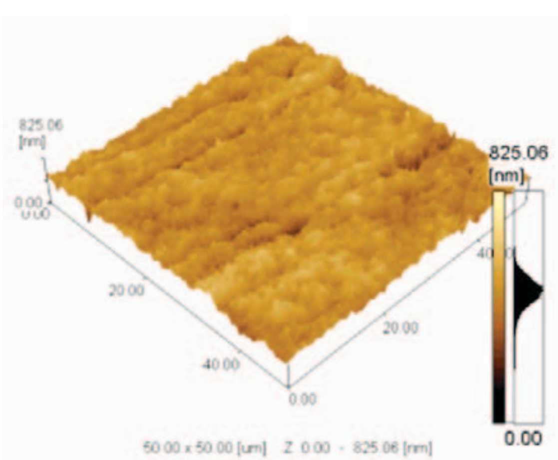

Figure 1. AFM image of Filtek Z250 resin composite at the baseline (T_0). Before treatment, the surface of this composite appears regular with no peaks or deep valleys detectable.

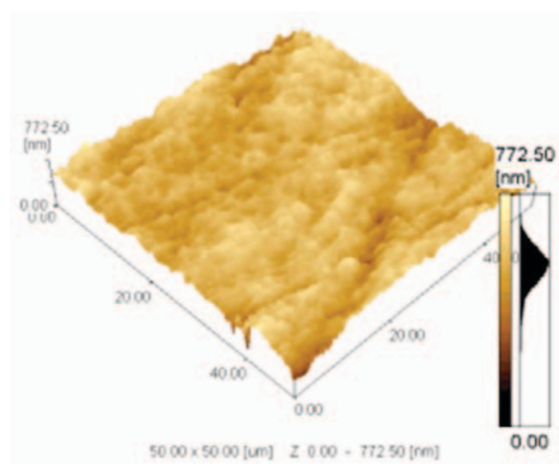

Figure 2. AFM image of Filtek Supreme resin composite at the baseline (T_0). Before pH-cycling and brushing, the surface is even with imperceptible irregularities.

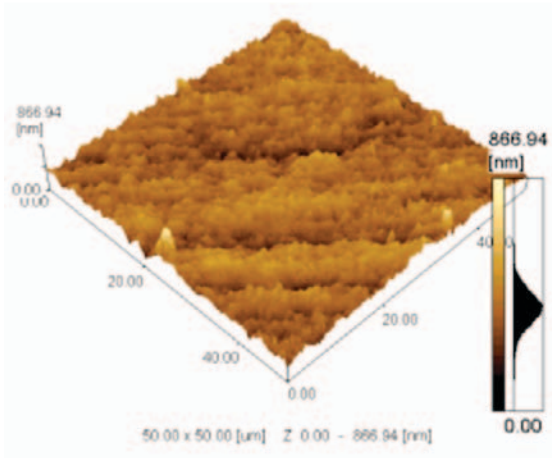

Figure 3. AFM image of Filtek Z250 resin composite after pH-cycling (T_c). After cycling, some irregularities are observed on the composite surface, but no significant differences were noted compared to the baseline.

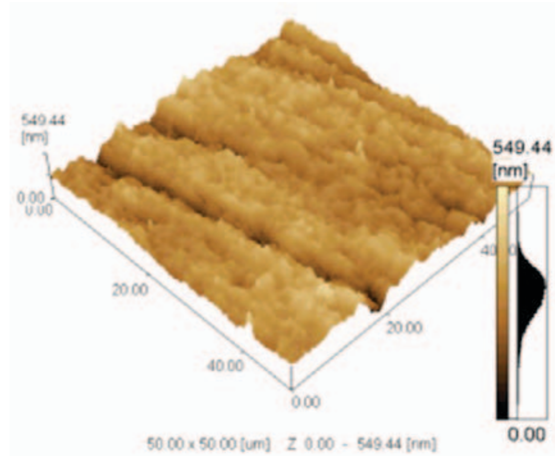

Figure 4. AFM image of Filtek Supreme resin composite after pH-cycling (T_c). The Filtek Supreme specimens were observed to be similar to Filtek Z250 with the development of surface grooves and fissures but no significant differences from the baseline (T_c).

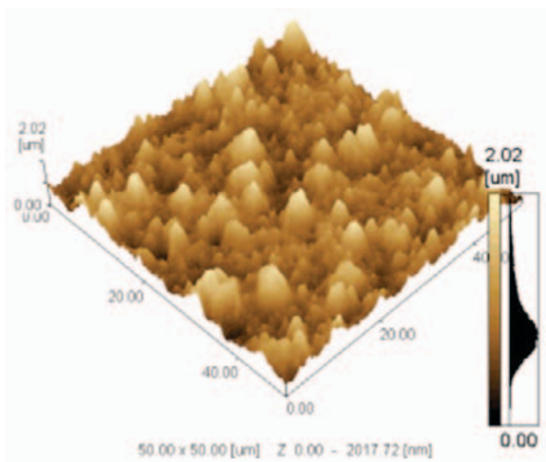

Figure 5. AFM image of Filtek Z250 after simulated toothbrushing (T_b). A significant increase in surface roughness is evident. Removal of the organic matrix and dislodgment of inorganic fillers are a direct result of simulated toothbrushing.

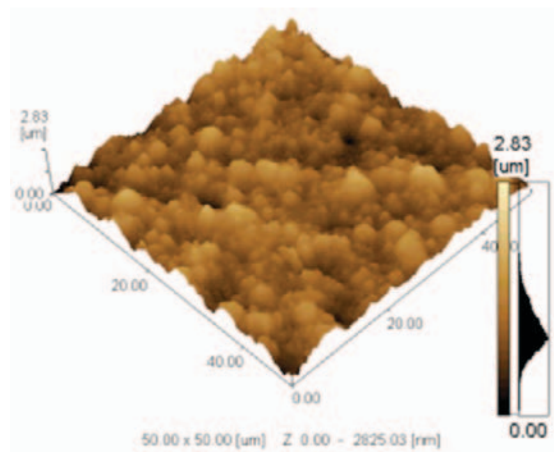

Figure 6. AFM image of Filtek Supreme after simulated brushing (T_b). A remarkable change in the composite surface is noted after toothbrushing, much like that seen in Figure 5 for Filtek Z250.

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 al[20] 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 a significant increase in composite roughness, and the uneven surfaces (Figures 5 and 6) most likely represented resin matrix loss and the dislodgement of the filler particles promoted by mechanical brushing. Evidence has shown that simulated brushing increases the surface roughness of composites because the act of brushing is able to wear away the organic matrix, thereby exposing the inorganic particles.[10,21,22] However, under oral conditions, the silane coating at the resin-filler interface also may disintegrate due to low pH regimen and lead to exfoliation of the filler particles.

It was expected that after pH-cycling and simulated brushing, the nanofilled composite would present lower surface roughness values than the microhybrid resin. Conversely, no differences were observed between the two composites at the baseline (T_0), after pH-cycling (T_c), or after toothbrushing (T_b). In fact, at baseline, similarities between composites were expected and actually desirable because they allowed comparison between these polymer-based materials. Given that these materials present some differences in formulation, one would expect that they might behave differently after pH-cycling and toothbrushing. Teixeira et al[22] observed differences between Filtek Z250 and Filtek Supreme after simulated brushing. These authors reported that Filtek Supreme had lower wear rates after 20,000, 50,000, and 100,000 cycles but higher surface roughness than Filtek Z250 after 50,000 and 100,000 cycles. The authors confirmed that the surface roughness increased after 10,000, 20,000, 50,000, and 100,000 brushing strokes for both nanofilled and microhybrid composites.[22] The differences in the results may be due to the fact that Teixeira et al[22] used a 250-g vertical load at a rate of 1.5 Hz, whereas in the present study brushing was simulated under 200 g of vertical load at a rate of 1.0 Hz.

The mechanical properties of nanofilled composites (such as the modulus of elasticity, flexural strength, tensile strength, and fracture toughness) are known to be similar to those of hybrid composite resins.[5] Because no differences were found between the nanofilled and microhybrid composites when submitted to toothbrushing abrasion, it could be speculated that nanofilled composites would exhibit mechanical behavior similar to that of microhybrid composites. Some studies have demonstrated the effect of filler size on wear and 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.[3] 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.[5,7]

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 nanoifilled 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

1. Turssi CP, Ferracane JL, Ferracane LL. Wear and fatigue behavior of nano-structured dental resin composites. J Biomed Mater Res B Appl Biomater. 2006; 78(1):196–203.
2. Heintze SD. How to qualify and validate wear simulation devices and methods. Dent Mater. 2006; 22(8):712-34.
3. Turssi CP, Ferracane JL, Vogel K. Filler features and their effects on wear and degree of conversion of particulate dental resin composites. Biomaterials. 2005; 26(24):4932–7.
4. Ikejima I, Nomoto R, McCabe JF. Shear punch strength and flexural strength of model composites with varying filler volume fraction, particle size and silanation. Dent Mater. 2003;19(3):206–11.
5. Beun S, Glorieux T, Devaux J, Vreven J, Leloup G. Characterization of nanofilled compared to universal and microfilled composites. Dent Mater. 2007; 23(1):51-9.

6. Choi MS, Lee YK, Lim BS, Rhee SH, Yang HC. Changes in surface characteristics of dental resin composites after polishing. J Mater Sci Mater Med. 2005; 16(4):347-53.
7. Mitra SB, Wu D, Holmes BN. An application of nanotechnology in advanced dental materials. J Am Dent Assoc. 2003; 134(10):1382-90.
8. Terry DA. Direct applications of a nanocomposite resin system: Part 1—The evolution of contemporary composite materials. Pract Proced Aesthet Dent. 2004; 16(6):417-22.
9. Sarrett DC, Coletti DP, Peluso AR. The effects of alcoholic beverages on composite wear. Dent Mater. 2000; 16(1):62-7.
10. Turssi CP, de Magalhães CS, Serra MC, Rodrigues Júnior AL. Surface roughness assessment of resin-based materials during brushing preceded by pH-cycling simulations. Oper Dent. 2001; 26(6):576-84.
11. Ortengren U, Andersson F, Elgh U, Terselius B, Karlsson S. Influence of pH and storage time on the sorption and solubility behaviour of three composite resin materials. J Dent. 2001; 29(1):35-41.
12. Yap AU, Chew CL, Ong LF, Teoh SH. Environmental damage and occlusal contact area wear of composite restoratives. J Oral Rehabil. 2002; 29(1):87-97.
13. Prakki A, Cilli R, Mondelli RF, Kalachandra S, Pereira JC. Influence of pH environment on polymer based dental material properties. J Dent. 2005; 33(2):91-8.
14. Attin T, Buchalla W, Hellwig E. Effect of topical fluoride application on toothbrushing abrasion of resin composites. Dent Mater. 2006; 22(4):308-13.
15. Wang L, Garcia FC, Amarante de Araújo P, Franco EB, Mondelli RF. Wear resistance of packable resin composites after simulated toothbrushing test. J Esthet Rest Dent. 2004; 16(5):303-14.
16. Mondelli RF, Wang L, Garcia FC, Prakki A, Mondelli J, Franco EB, Ishikiriama A. Evaluation of weight loss and surface roughness of compomers after simulated toothbrushing abrasion test. J Appl Oral Sci. 2005; 13(2):131-5.
17. Watari F. Compositional and morphological imaging of CO_2 laser irradiated human teeth by low vacuum SEM, confocal laser scanning microscopy and atomic force microscopy. J Mater Sci Mater Med. 2001; 12(3):189-94.
18. Serra MC, Cury JA. The *in vitro* effect of glass-ionomer cement restoration on enamel subjected to a demineralization and remineralization model. Quintessence Int. 1992; 23(2):143-7.
19. Charig A, Winston A, Flickinger M. Enamel mineralization by calcium-containing-bicarbonate toothpastes: assessment by various techniques. Compend Contin Educ Dent. 2004; 25(9 Suppl 1):14-24.
20. Turssi CP, Hara AT, de Magalhães CS, Serra MC, Rodrigues AL Jr. Influence of storage regime prior to abrasion on surface topography of restorative materials. J Biomed Mater Res B Appl Biomater. 2003; 65(2):227-32.
21. Senawongse P, Pongprueksa P. Surface roughness of nanofill and nanohybrid resin composites after polishing and brushing. J Esthet Restor Dent. 2007; 19(5):265-73.
22. Teixeira EC, Thompson JL, Piascik JR, Thompson JY. *In vitro* toothbrush-dentifrice abrasion of two restorative composites. J Esthet Restor Dent. 2005; 17(3):172-80.

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