

IN VITRO MICROHARDNESS EVALUATION OF BULK FILL RESINS AFTER SIX MONTHS OF WATER STORAGE

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ABSTRACT

Introduction: Bulk-fill resin composites reduce clinical time of a restoration because they can be cured at larger increments (4 to 5 mm) than conventional composites. This study determined the Knoop microhardness of bulk-fill and conventional composites at 24 hours and 180 days after storage in a solvent. **Methods:** Twenty-four specimens were divided into four groups: Z2 (control, conventional resin composite), AB (Aura bulk fill), TB (Tetric bulk fill) and FB (Filtek bulk fill). Knoop microhardness (KHN) was determined at the top and bottom surfaces of each specimen.

Results: KHN values for the top measurements at 24 hours and 180 days were: Z2 - $95.02^{Aa} \pm 10.72$ and $63.50^{Ab} \pm 7.98$; AB - $60.92^{Ba} \pm 5.43$ and $39.43^{Bb} \pm 3.02$; TB - $66.38^{Ba} \pm 7.38$ and $41.05^{Bb} \pm 6.29$; FB - $60.30^{Ba} \pm 8.42$ and $44.61^{Bb} \pm 6.65$. For the bottom surface, values were: Z2 - $79.65^{Aa} \pm 9.67$ and $52.36^{Ab} \pm 5.6$; AB - $38.79^{Ba} \pm 6.46$ and $30.16^{Bb} \pm 4.34$; TB - $40.14^{Ba} \pm 7.79$ and $31.55^{Bb} \pm 5.62$; FB - $42.67^{Ba} \pm 5.94$ and $33.69^{Bb} \pm 4.15$. The AB, TB and FB groups had significantly lower values than the control group for both top and bottom measurements at the two time points. **Conclusion:** Storage in solvent reduced the microhardness of all composites, with no statistically significant differences between them. Only the control group had clinically acceptable results. As bulk-fill resin composites should not be exposed to humid environments, an occlusal layer filled with a conventional composite should be used.

KEYWORDS:

Resin Composites. Hardness. Permanent Dental Restoration.

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INTRODUCTION

Resin composites are some of the contemporary materials most often used to restore teeth that have lost dental structure. Indicated for small and large cavities, they withstand masticatory forces well. Since the development of the Bis-GMA monomer, the main component of a resin matrix, there has been a revolution in composite formulation. Changes in the size and shape of filler particles led to improved resistance to wear, better color stability and good final polishing.¹ Such characteristics are direct contributions to adequate clinical functioning.

Conventional composites demand longer operational times because of the need for incremental filling. Light penetration during light curing, one of the main factors to justify the demand for the incremental technique, promotes the adequate conversion of monomers into polymers. For that purpose, increments should not be greater than 2 mm.^{2,3,4} Not all monomers will be converted during the conversion from gel to solid, as they may degrade at different rates when in contact with water. Depending on the amount of unconverted monomers in polymers, hardness may be reduced, and, consequently, materials may have a shorter clinical longevity.

The conversion of monomers into polymers requires the use of a source of visible light of 470-nm wavelength at irradiances greater than 500 mW/cm.^{3,7} Resin composite polymerization is also associated with the distance between the material to be polymerized and the light source.⁴ The greater the distance, the lower the light intensity, which compromises the polymerization of the restorative material.⁸

Resin composite stability is directly dependent on the degree of conversion of its monomers. Insufficient polymerization may contribute to degradation by loss of components when in contact with water, as in the humid oral environment. The release of these components may produce biological and physical effects that are harmful to the organism. In insufficient polymerization, water and solvent may diffuse in the resin matrix, which results in loss of components and the consequent chemical degradation. This degradation affects the physical and mechanical properties of the material, which results in color changes and reductions of resistance to wear and superficial hardness.⁹

The volume of water sorption by a resin composite depends on its hydrophilicity, amount of crosslinks,¹⁰ spaces between the polymeric chains and water diffusion in the matrix. The main results of water and solvent diffusion into the resin matrix are the expansion of the polymer network and, in some cases, chemical degradation. Water sorption may lead to hygroscopic expansion and plasticization,¹¹ with an increase in the space between the polymer chains. This way, the free monomers, caught in the polymer network, diffuse into the solvent because of their molecular size and affinity with the solvent. Material solubility compromises biocompatibility

and reduces volume, which negatively affects mechanical properties, reducing resistance to wear and hardness and increasing discoloration.

Bulk-fill resin composites have been developed to reduce clinical time. They may be used in increments of up to 5 mm and have low shrinkage and polymerization stress¹³. This study determined the microhardness of bulk-fill resin composites in comparison with a conventional resin after polymerization and at 180 days of storage in distilled water. The null hypothesis was that resin microhardness does not undergo changes after immersion in an aqueous environment for the length of storage time under study.

MATERIAL AND METHODS

The microhardness trial was conducted in the Dental Materials Laboratory (LAMAD) of the School of Dentistry of the Federal University of Rio Grande do Sul, Porto Alegre, Brazil. Twenty-four specimens were prepared using a metal matrix with numbered perforations and 5 mm diameter¹⁴ (Fig 1). These specimens were divided into four groups of six units each. The same procedures were used to prepare all specimens. A strip of polyester film was placed in the lower part of the metal matrix, followed by a strip of black insulation tape to block light. After that, single increments of the different types of resin composite were added to the matrix and slightly pressed down to avoid the incorporation of air. Then they were covered again with a polyester matrix and a glass plate on it,¹⁴⁻¹⁶ so that excess material may run off, and the surface was smooth and flat in the absence of oxygen (Fig 2). Increments were light-cured according to the time recommended by the manufac-

turer using a dental composite curing light (Op-tilux 401, Demetron, USA) at a mean polymerization intensity of 900 mW/cm², which might vary ± 10 mW/cm², and monitored using a radiometer (Ecel R-D7) for every five samples. The samples were divided into groups: control (Z2) - conventional composite, Z250 (3M ESPE, USA) with increments of 2-mm thickness; AB - Aura bulk-fill resin composite (SDI, Australia) with a thickness of 4 mm; TB - Tetric N-Ceram bulk-fill resin composite (Ivoclar-Vivadent, Schaan, Liechtenstein) with 4-mm increments; and FB - bulk-fill Filtek (3M ESPE, USA) with 5-mm increments (Fig 3). The groups are shown in Table 1.

Specimen microhardness at the top (surface in direct contact with light source) and bottom (surface away from light source) was determined 24 hours after light curing. Later, the specimens were stored in distilled water, changed every 15 days, for 180 days, when top and bottom microhardness was determined again.

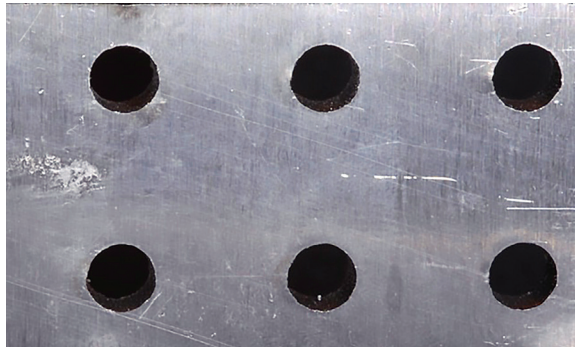


Figure 1:
Metal matrix showing orifice diameter.

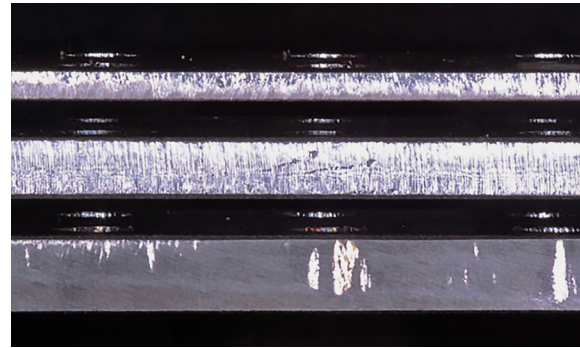


Figure 2:
Metal matrix showing thickness of each matrix, which corresponds to depth of each specimen.

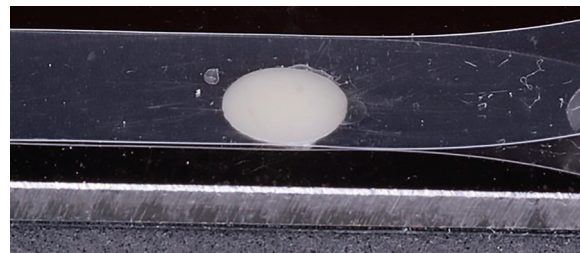


Figure 3:
Resin placed in matrix with a piece of polyester tape for polymerization.

Table 1:
Resin composites used in this study.

GROUP	MATERIAL	MANUFACTURER/ BATCH	SHADE	PHOTOINITIATOR	MONOMER	LOAD %/W	POLYMERIZATION TIME*
Z2	Filtek Z250 Universal	3M ESPE 390045	A3	Cq.	UDMA BisEMA BisGMA TEGDMA	75-85	20s
AB	Aura Bulk Fill	SDI 150557	Uni	N.d.	N. d.	N. d.	20s
TB	Tetric N-Ceram Bulk Fill	Ivoclar-Vivadent T47219	U IVB	Cq.	Bis-GMA, UDMA	75-77	20s
FB	Filtek Bulk Fill	3M ESPE N685667	A3	Cq.	Bis-GMA, BisEMA, UDMA	76,5	20s

*Polymerization time at ± 900 mW/cm²

Knoop microhardness (KN) was estimated as the mean value of three indentations on each surface, measured using an automatic microhardness tester (Shimadzu, Japan) equipped with a 40x lens at 10 gf loads for five seconds. Microhardness was calculated using the following equation:

Knoop hardness = $\frac{14228 \cdot c}{d^2}$, where:

d^2

14228 is a constant, c is the load in grams, and d is the length of indentation along its long axis expressed in μm (Fig 4).

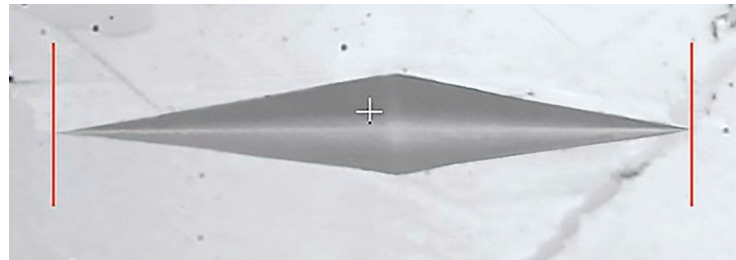


Figure 4:

Image of indentation and limits of area measured.

The values acquired were analyzed statistically using a mixed-design analysis of variance followed by the Tukey test for multiple comparisons. The level of significance was set at 5%.

RESULTS

Results are shown in Table 2.

Table 2:

Mean and standard deviation values of Knoop microhardness at the top and bottom surfaces of specimens, and percentage of hardness reduction after storage.

GROUPS	AFTER 24 H	AFTER 6 MONTHS	% REDUCTION
Z2 top	95.02 ± 10.72 ^{Aa}	63.50 ± 7.98 ^b	32.7 ± 8.6 ^A
Ab top	60.92 ± 5.43 ^{Ca}	39.43 ± 3.02 ^b	35.1 ± 3.7 ^A
Tb top	66.38 ± 7.38 ^{B,Ca}	41.05 ± 6.29 ^b	37.5 ± 7.6 ^A
Fb top	60.30 ± 8.42 ^{Ca}	44.61 ± 6.65 ^b	24.8 ± 13.9 ^A
Z2 bottom	79.65 ± 9.67 ^{Ba}	52.36 ± 5.6 ^b	33.1 ± 13.1 ^A
Ab bottom	38.79 ± 6.46 ^{Da}	30.16 ± 4.34 ^b	26.3 ± 14.1 ^A
Tb bottom	40.14 ± 7.79 ^{Da}	31.55 ± 5.62 ^b	24.2 ± 15.1 ^A
Fb bottom	42.67 ± 5.94 ^{Da}	33.69 ± 4.15 ^b	20.5 ± 8.8 ^A
p values	p=0.004	p<0.001	p=0.216

Means followed by different capital letters in the column or by different lowercase letters indicate statistically significant differences.

The samples stored in distilled water for 180 days had a mean microhardness reduction of 35% at the top and 33% at the bottom surface, which was statistically significant; therefore, the null hypothesis was false. However, the percentage of microhardness reduction after storage was not statistically different between groups ($p=0.216$). There was a statistically significant difference between top and bottom hardness of composites in the same group and at the different measurement time points. The Z2 group had the highest top and bottom measurements, and the AB, TB and FB groups did not have any statistically significant differences from each other in either top or bottom measurements. In addition, there was a statistically significant difference ($p<0.05$) from the Z2 group when exposed to mean irradiance values of 900 mW/cm^2 , as recommended by the manufacturer. The percentage reductions for the time evaluated were not statistically different between groups.

DISCUSSION

The microhardness of a light-cured resin composite is directly associated with the percentage of conversion of monomers into polymers, which depends on the penetration of the activating light into the resin composite and requires the use of a light-curing unit that emits visible light above 500 mW/cm^2 .^{2,3,7,14} Some of the factors that may affect polymerization depth are composite type, opacity and translucency, increment depth, distance from the tip of the

light-curing unit, and distribution of filler particles.⁴ The degree of conversion peaks at 30 min and stops increasing at 24 h after light curing. Because of that, Knoop hardness of the specimens was measured 24 h after light curing.

This study corroborates findings by Alshali et al in 2015,¹² who reported that one of the consequences of the solubility of restorative materials is the reduction of their hardness. In our study, hardness reduced a mean 35% at the top of specimens and 25% at the bottom, both statistically significant reductions. This may be seen as a predictor of a poorer clinical performance and longevity of restorations.¹⁸

Alqahtani et al. found that the increase in material depth and the consequent distance from the light-curing source affected microhardness unfavorably.¹⁹ The top surface of the resin receives greater light intensity, which results in better polymerization. As the depth increases, resin gradually receives less photons, which reduces the degree of polymerization. These data were confirmed in this study, which found that the values of specimen hardness were lower at the bottom than at the top.

The mechanical properties of resin composites depend, primarily, on their monomer systems, amount and size of fillers and filler-polymer binding. Bulk-fill resin composites have lower hardness values at both the top and bottom surfaces than conventional composites. This may be explained by the fact that they have a smaller amount of filler by weight: 75% for the bulk-fill resin composites and 85% for conventional composites.¹²

The data obtained in this study showed that all the groups had a statistically significant hardness reduction after storage, on both top and bottom surfaces. Such changes, the result of water sorption in the porosities and intermolecular spaces inside the composite, lead to physical and chemical composite degradation.^{18,20} The sorption of more or less water molecules depends on the degree of polymerization, structural crosslink density and hydrophilicity.^{2,11}

Extensive water sorption may be the cause of cuspal deflections and microfissures of restored teeth.^{21,22} It may also lead to microhardness reductions due to plasticization of specimens after water storage. In addition to the results mentioned above, water diffusion leads to progressive material degradation.^{2,5} Storage time (180

days) was defined according to Ferracane et al. in 1998, who found that water sorption occurs up to the point of saturation and affects composite properties after two and three months of water storage, without any greater microhardness reduction after that time.⁶ The lower Knoop hardness values found for bulk-fill resin composites may be explained by their monomer system, which contains UDMA. This monomer is more likely to dissolve in solvents than Bis-GMA.²³ Another possible explanation is that its composition includes diluent monomers, whose purpose is to reduce shrinkage stress.¹⁸

CONCLUSION

Bulk-fill resin composites had statistically lower top and bottom Knoop hardness values than the conventional resin at the two evaluation time points.

Bulk-fill resin composites had statistically similar results, regardless of commercial brand.

The percentages of Knoop hardness reductions for bulk-fill and conventional resin composites were not statistically difference during storage time.

Because of the low values of superficial hardness of bulk-fill resin composites, the authors suggest the use of a conventional composite occlusal layer to avoid the contact of the bulk-fill composite with the oral environment.

REFERENCES

- Al Sunbul H, Silikas N, Watts DC. Resin-based composites show similar kinetic profiles for dimensional change and recovery with solvent storage. *Dent Mater*. 2015 Oct;31(10):201-17.
- Alshali RZ, Salim NA, Satterthwaite JD, Silikas N. Long-term sorption and solubility of bulk-fill and conventional resin composites in water and artificial saliva. *J Dent*. 2015 Dec;43:1511-8.
- Flury S, Peutzfeldt A, Lussi A. Influence of increment thickness on microhardness and dentin bond strength of bulk fill resin-composites. *Dent Mater*. 2014 Oct;30(10):1104-12.
- Alrahlah A, Silikas N, Watts DC. Post-cure depth of cure of bulk fill dental resin-composites. *Dent Mater*. 2014 Feb;30(2):149-54.
- Gonçalves L, Noronha Filho JD, Guimarães J, Poskus L, Silva E. Solubility, salivary sorption and degree of conversion of dimethacrylate based polymeric matrixes. *J Biomed Mater Res B Appl Biomater*. 2008 May;85(2):320-5.
- Ferracane JL, Berge HX, Condon JR. In vitro aging of dental composites in water-effect of degree of conversion, filler volume, and filler/matrixcoupling. *J Biomed Mater Res*. 1998 Dec;42(3):465-72.
- Van Dijken JW, Pallesen U. A randomized controlled three year evaluation of "bulk-filled" posterior resin restorations based on stress decreasing resin technology. *Dental Materials*. 2014 Sept;30(9):245-51.
- Finan L, Palin WM, Moskwa N, McGinley EI, Fleming GJ. The influence of irradiation potential on the degree of conversion and mechanical properties of two bulk-fill flowable RBC base materials. *Dent Mater*. 2013 Aug;29(8):906-12.
- Van Ende A, De Munck J, Lise DP, Van Meerbeek B. Bulk-fill composites: a review of the current literature. *J Adhes Dent*. 2017;19(2):95-109.
- Yujie Z, Jingwei XU. Effect of immersion in various media on the sorption, solubility, elution of unreacted monomers, and flexural properties of two model dental composite compositions. *J Mater Sci Mater Med*. 2008 June;19(6):2477-83.
- Wei YJ, Silikas N, Zhang ZT, Watts DC. The relationship between cyclic hygroscopic dimensional changes and water sorption/desorption of self-adhering and new resin-matrix composites. *Dent Mater*. 2013 Sept;29(9):218-26.
- Alshali RZ, Salim NA, Sung R, Satterthwaite JD, Silikas N. Analysis of long-term monomer elution from bulk-fill and conventional resin composites using high performance liquid chromatography. *Dental Materials*. 2015 Dec;31(12):1587-98.
- Zorzín J, Maier E, Harre S, Fey T, Belli R, Lohbauer U, et al. Bulk-fill resin composites: polymerization properties and extended light curing. *Dent Mater*. 2015 Mar;31(3):293-301.
- Godoy E. Avaliação da capacidade de polimerização e elevação de temperatura produzida por aparelhos fotopolimerizadores [dissertação]. Ponta Grossa (PR): Universidade Estadual de Ponta Grossa; 2008.
- Alshali RZ, Salim NA, Satterthwaite JD, Silikas N. Post-irradiation hardness development, chemical softening, and thermal stability of bulk-fill and conventional resin-composites. *J Dent*. 2015 Feb;43(2):209-18.
- Alshali ZR, Silikas N, Satterthwaite JD. Degree of conversion of bulk-fill compared to conventional resin-composites at two time intervals. *Dent Mater*. 2013 Sept;29(9):e213-7.
- Al-Ahdal K, Ilie N, Silikas N, Watts DC. Polymerization kinetics and impact of post polymerization on the degree of conversion of bulk-fill resin-composite at clinically relevant depth. *Dent Mater*. 2015 Oct;31(10):1207-13.
- Sunbul HA, Silikas N, Watts DC. Surface and bulk properties of dental resin-composites after solvent storage. *Dent Mater*. 2016 Aug;32(8):987-97.
- Alqahtani MÇ, Michaud PI, Sullivan B, Labrie D, Alshaafi MM, Price RB. Effect of high irradiance on depth of cure of a conventional and a bulk fill resin-based composite. *Oper Dent*. 2015 Nov-Dec;40(6):662-72.
- Watts DC, Amer OM, Combe EC. Surface hardness development in light-cured composites. *Dental Materials*. 1987;3(5):265-9.
- Wei YJ, Silikas N, Zhang ZT, Watts DC. Diffusion and concurrent solubility of self-adhering and new resin-matrix composites during water sorption/desorption cycles. *Dent Mater*. 2011 Feb;27(2):197-205.
- Alrahlah A, Silikas N, Watts DC. Hygroscopic expansion kinetics of dental resin-composites. *Dental Mater*. 2014 Feb;30(2):143-8.
- Sajjani AR, Hegde MN. Leaching of monomers from bulk-fill composites: an in vitro study. *J Conserv Dent*. 2016 Sept-Oct;19(5):482-6.

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