

Flexural Strength Of Microhybrid Composites After Immersion In Distilled Water And Citric Acid Solutions

Kekuatan Lentur Komposit Mikrohibrid Setelah Perendaman Dalam Larutan Air Distil Dan Asam Sitrat

Manendra Muhtar, Ellyza Herda, Bambang Irawan

Department of Dental Materials, Faculty of Dentistry, Universitas Indonesia, Jakarta, Indonesia
Correspondence email to: ellyza.herda@ui.ac.id

ABSTRAK

Sifat mekanis restorasi komposit resin dapat terdegradasi oleh cairan di lingkungan mulut, termasuk makanan dan minuman seperti buah-buahan. Tujuan dari penelitian ini adalah untuk membandingkan kekuatan lentur dua komposit mikrohibrid yang berbeda setelah perendaman dalam air suling dan larutan asam sitrat dengan konsentrasi buah jeruk yang biasa dikonsumsi: jeruk mandarin dan lemon. Empat puluh G-aenial PosteriorTM dan empat puluh spesimen batang uji bengkok 3 titik Filtek Z250TM dibagi menjadi empat kelompok, 1 hari penyimpanan kering, 30 hari perendaman dalam air suling, 30 hari perendaman dalam asam sitrat 0,06 mol/L (jeruk mandarin konsentrasi), dan 30 hari perendaman dalam 0,30 mol/L asam sitrat (konsentrasi lemon). Setelah itu, kekuatan lentur spesimen diukur dengan Shimadzu, Japan AGS-X Universal Testing Machine. Data dianalisis menggunakan uji statistik One-Way ANOVA dan Tamhane Post Hoc. Analisis statistik menunjukkan penurunan yang signifikan dalam kekuatan lentur G-aenial PosteriorTM. Komposit Filtek Z250TM juga menunjukkan penurunan kekuatan lentur yang signifikan dari nilai $138,72 \pm 6,26$ MPa setelah 1 kali penyimpanan kering, menjadi $93,58 \pm 7,69$ MPa setelah perendaman dalam akuades, $96,39 \pm 3,15$ MPa setelah perendaman dalam asam sitrat 0,06 mol/L, dan $96,14 \pm 5,39$ MPa setelah direndam dalam asam sitrat 0,30 mol/L. Perbedaan kekuatan lentur yang signifikan ditunjukkan antara kedua bahan mikrohibrid, pada masing-masing kelompok perlakuan, dengan nilai Filtek Z250TM yang jauh lebih tinggi. Disimpulkan bahwa perendaman dalam air dan asam sitrat memiliki pengaruh yang signifikan terhadap kekuatan lentur komposit mikrohibrid.

Kata Kunci: Resin komposit mikrohibrid, kuat lentur, perendaman dalam akuades, perendaman dalam asam sitrat

ABSTRACT

The mechanical properties of a resin composite restoration can be degraded by liquids in the oral environment, including food and drinks such as fruits. The aim of this study was to compare the flexural strength of two different microhybrid composites after immersion in distilled water and citric acid solutions with concentrations of commonly consumed citrus fruits: mandarin orange and lemon. Forty G-aenial PosteriorTM and forty Filtek Z250TM 3-point bend test bar specimens are divided into four groups, 1 day of dry storage, 30 days of immersion in distilled water, 30 days of immersion in 0.06 mol/L citric acid (mandarin orange concentration), and 30 days of immersion in 0.30 mol/L citric acid (lemon concentration). Afterwards, the flexural strength of the specimens are measured with a Shimadzu, Japan AGS-X Universal Testing Machine. The Data was analyzed using a One-Way ANOVA statistical test and Tamhane Post Hoc. Statistical analysis shows a significant reduction in the flexural strength of G-aenial PosteriorTM. Filtek Z250TM composite also showed a significant reduction in flexural strength from a value of 138.72 ± 6.26 MPa after 1 dry storage, to 93.58 ± 7.69 MPa after immersion in distilled water, 96.39 ± 3.15 MPa after immersion in 0.06 mol/L citric acid, and 96.14 ± 5.39 MPa after immersion in 0.30 mol/L citric acid. A significant difference in flexural strength is shown between the two microhybrid materials, in each of the treatment groups, with a significantly higher value for Filtek Z250TM. It is concluded that immersion in water and citric acid has a significant effect on the flexural strength of the microhybrid composites.

Keywords: Microhybrid composite resin, flexural strength, immersion, distilled water, citric acid

INTRODUCTION

Microhybrid composite resins are a type of composite restoration with a combination of fine filler particles (average particle size of 0.04-1 µm) and microfine filler particles (average particle size of 0.04-0.2 µm). These composites generally have good mechanical properties and can be used in areas with high stress, such as the posterior region.¹

The longterm clinical success of a restoration has a strong correlation with its mechanical properties. The mechanical properties of a composite resin depend on its composition and microstructure. One of the important mechanical properties is flexural strength, as a restoration with poor flexural strength has increased possibility of fracture. Restorations in the posterior region have high stress and a large occlusal load; therefore, it is very important for them to have good mechanical properties, including flexural strength. Flexural strength measures the maximum stress that a material can withstand before fracturing.² According to ISO 4049 standards, a posterior restoration needs to have a minimal flexural strength of 80 MPa.³

G-aenial Posterior™ is a microhybrid composite product that can be used for posterior restorations. It contains prepolymerized fillers, and the matrix doesn't contain Bis-GMA.⁴ Prepolymerized fillers are usually added to composite resins to reduce the amount of unpolymerized resin, reduce polymerization shrinkage, increase filler loading, increase polishability, and enhance esthetics by increasing light reflection and scattering.⁵ However, Blackham *et al.* found lower strength values for composite resins that contain prepolymerized fillers compared to traditional hybrid composites.⁶ Composites with prepolymerized fillers can also have increased water sorption.⁵

G-aenial Posterior™ matrix consists of UDMA and dimethacrylate co-monomers, without Bis-GMA. Sideridou *et al.* found that UDMA and co-polymers of UDMA and Bis-GMA have strength values larger than Bis-

GMA, despite that a significant reduction of tensile strength was found in UDMA after immersion in distilled water for 30 days, while Bis-GMA resin didn't show a significant reduction of strength.⁷

Saliva, food, drinks, and the interaction between these components can cause degradation of resin composite mechanical properties. Based on the FDA (Food and Drug Administration) guide, several food simulating liquids like citric acid can be used as aging medias for the testing of the composite mechanical properties.⁸ In this study, the flexural strength of G-aenial Posterior™ is compared with Filtek Z250™ (a microhybrid composite that contains prepolymerized fillers and has Bis-GMA resin), the flexural strength is tested after dry storage, immersion in distilled water, and immersion in two citric acid concentrations. The citric acid solutions simulate two commonly consumed citrus fruits, which are mandarin oranges with a 0.06 mol/L concentration and lemons with a 0.30 mol/L concentration.⁹ Immersion in distilled water is tested as a control group.

RESEARCH METHOD

In the present research study, two types of microhybrid composites were used, G-aenial Posterior™ (GC, Japan) and Filtek Z250™ (3M, USA). The composition of the two materials can be seen in Table 1.

Forty G-aenial Posterior™ and forty Filtek Z250™ 3-point bend test bar shaped specimens with a 25 x 2 x 2 mm size (ISO specification) were light cured for 20 seconds using a LED curing unit with an irradiance of 700 mW/cm² (LED Max Hilux); afterwards, they were divided into 4 treatment groups: 1 day of dry storage, 30 days immersion in distilled water, 30 days immersion in 0.06 mol/L citric acid (mandarin orange concentration), and 30 days immersion in 0.30 mol/L citric acid (lemon concentration). All the specimen groups were stored in an incubator at 37°C.

Table 1. Materials Details and Manufacturers' Specification

Material	Batch number	Filler loading (%)	Organic matrix	Filler type	Manufacturer
G-aenial Posterior™	GC-0130F 078-0020	65% vol and 77% wt	UDMA and dimethacrylate co monomers	Two types of 16-17 µm prepolymerized fillers containing 16 nm silica and 400 nm strontium, 100 nm lantahnoid fluoride, 16 nm fumed silica and 850 nm fluoroaluminosilicate glass	GC Corporation, Japan
Filtek Z250™	70-2010-5201-9	60% vol and 78% wt	Bis-GMA, UDMA, Bis-EMA6 and TEGDMA	0.01 – 3.5 µm silica/zirconia fillers	3M Company, USA

The pH of the citric acid and distilled water solutions were also measured using a pH meter (Cyberscan pH 510, Eutech). After treatment, the flexural strength of the Specimens were tested using a Shimadzu, Japan AGS-X Universal Testing Machine, with a crosshead speed of 0.5 mm/ min and a load of 50 kgF.

A One-Way Anova statistical test was used to analyze the differences between all the treatment groups (G-aenial Posterior™ and Filtek Z250™ composites after 1 day of storage and after immersion in the distilled water and citric acid solutions). To determine which groups have a significant difference, a Tamhane Post Hoc was done. All data were subjected to a normality and homogeneity test.

RESULTS

pH tests showed a value of 7 for distilled water, 2,19 for the 0.06 mol/L citric

acid solution and 2 for the 0.30 mol/L citric acid solution. The results of the flexural strength tests for G-aenial Posterior™ and Filtek Z250™ is shown in Table 2 and Figure 1. Filtek Z250™ has a significantly larger flexural strength value compared to G-aenial Posterior™ in all of the treatment groups (1 day dry storage, 30 days immersion in distilled water, 30 days immersion in 0.06 mol/L citric acid and 30 days immersion in 0.30 mol/L citric acid). In each of the microhybrid types, a significant reduction of flexural strength is observed from the 1 day dry storage group towards the 30 days immersion in distilled water and citric acid groups. Finally, the flexural strength values of each microhybrid composite types were not different between the distilled water, 0.06 mol/L citric acid and 0.30 mol/L immersion groups.

Table 2. Average Flexural Strength Values of Two Microhybrid Composites After Several Treatments

Treatment	Average G-aenial Posterior™ Flexural Strength Value (MPa) ± SD	Average Filtek Z250™ Flexural Strength Value (MPa) ± SD
1 day dry storage	86,32 ± 3,54 ^{*A}	138,72 ± 6,26 ^{*C}
30 days immersion in distilled water	70,14 ± 4,37 ^{*B}	93,58 ± 7,69 ^{*D}
30 days immersion in 0.06 mol/L citric acid	71,58 ± 4,09 ^{*B}	96,39 ± 3,15 ^{*D}
30 days immersion in 0.30 mol/L citric acid	71,01 ± 3,28 ^{*B}	96,14 ± 5,39 ^{*D}

*SD: standard deviation

* Same letters indicate no significant differences between the treatment groups, while different letters indicate a significant difference (p<0.05)

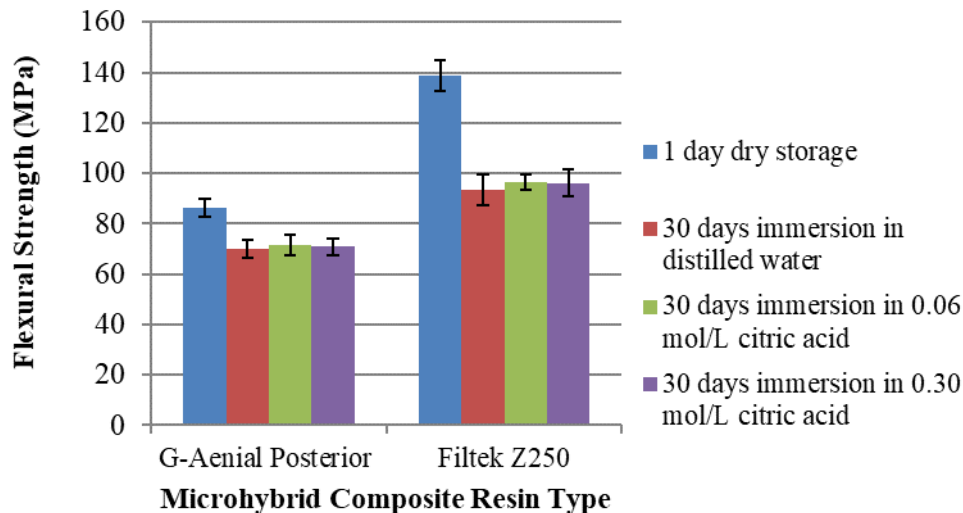


Figure 1. Average Flexural Strength Values of Two Microhybrid Composites After Several Treatments

DISCUSSION

In the present study, a significantly smaller flexural strength was found for G-aenial PosteriorTM compared to Filtek Z250TM in all of the treatment groups (1 day of dry storage, 30 days of immersion in distilled water, 30 days of immersion in 0.06 mol/L citric acid, and 30 days of immersion in 0.30 mol/L citric acid). The flexural strength values found in G-aenial PosteriorTM after immersion in water and the citric acid solutions were under 80 MPa, which is the minimum required flexural strength after immersion in water, based on ISO 4049.¹⁰ Meanwhile, all the flexural strength values found in Filtek Z250TM after the immersion groups were higher than the ISO criteria. A flexural strength value under the ISO criteria might imply that the composite resin has a higher risk of fracture. The difference in flexural strength can be attributed to the difference in composition between the two microhybrids.³

The flexural strength value of a composite resin can be influenced by its filler loading, filler distribution, and the type of filler and matrix that it has.^{1,11} A composite resin with higher filler loading usually also has higher strength values, however, G-aenial PosteriorTM and Filtek Z250TM have similar filler loading: G-aenial PosteriorTM with a 65% filler volume and 77% filler weight, and Filtek Z250TM has a 60% filler volume and 78% filler weight. Therefore, the significant difference in

strength between the two microhybrids might be primarily caused by the difference in filler and matrix composition. G-aenial PosteriorTM contains prepolymerized fillers that, despite increasing filler loading and enhancing esthetics, can cause strength values to be lower. Prepolymerized fillers in composites have been previously cured during the fabrication of the composite, which might cause the prepolymerized fillers to have few active bonding sites that can bond with the composite matrix, therefore having a poor integration in the matrix that might cause lower strength values.¹² Besides that, the fluoroaluminosilicate glass found in G-aenial PosteriorTM might also attribute to the lower strength values. Fluoroaluminosilicate glass is a relatively brittle glass material, Huang *et al.* found that the strength value of G-aenial PosteriorTM is smaller compared to G-aenial AnteriorTM, which contains silica fillers instead of fluoroaluminosilicate glass.¹³ Filtek Z250TM contains micro and nano sized filler particles with varied sizes (0,01-3,5 µm) that can fill one another, thus attributing to high strength values. Filtek Z250TM also doesn't contain prepolymerized fillers that might reduce its strength values. Regarding matrix, both G-aenial PosteriorTM and Filtek Z250TM contain UDMA resin, which is a type of resin with high flexural strength (around 92.3 MPa).⁷ Therefore, the significant differences in flexural strength between the two microhybrid

materials are probably mainly caused by the differences in filler types between them.

Microhybrid composites underwent a significant reduction of flexural strength after immersion in distilled water. Water can cause degradation in resin composites by degrading siloxane bonds (Si-O-Si) which are bonds found in the silanol of fillers and silane coupling agents. Degradation occurs through hydrolysis, in which H^+ ions from water bind to oxygen in the siloxane bonds, causing the bonds to break and the filler to debond.^{14,15} Moreover, the negative effects of water on the resin matrix include softening of the matrix because of polymer chain separation and the release of unreacted monomers. Weak organic acid solutions also have a similar degradation effect on composite resins, through impairment by H^+ ions and negative effects on the resin matrix.¹⁶

After immersion in distilled water and citric acid, the microhybrid Filtek Z250TM had a significantly higher flexural strength value compared to G-aenial PosteriorTM, but the average reduction of strength in Filtek Z250TM (31.24%) is larger compared to the reduction of strength in G-aenial PosteriorTM (17.97%). The Filtek Z250TM matrix contained Bis-GMA, Bis-EMA6, UDMA, and a little bit of TEGDMA, while G-aenial PosteriorTM contained UDMA and dimethacrylate comonomers. Bis-GMA and TEGDMA are more hydrophylic compared to UDMA¹⁶, and since those two monomers are found in Filtek Z250TM but not in G-aenial PosteriorTM, this might imply that Filtek Z250TM has a bigger water sorption value, thus causing the bigger reduction in strength.

Several studies state that 100% silica or quartz is a type of filler that is relatively inert towards water compared to other filler types such as barium, zirconia, and strontium.¹⁷⁻²⁰ Neither G-aenial PosteriorTM nor Filtek Z250TM contain 100% silica. G-aenial PosteriorTM contains a combination of silica, fluoroaluminosilicate glass, strontium, and prepolymerized fillers. Filtek Z250TM contains zirconia and silica fillers. According to its manual, the fillers in Filtek Z250TM were not given silane treatment, which may cause a poor bond between the filler and matrix. Therefore, despite Filtek Z250TM having a high filler load (78% weight), a weak filler matrix bond interface could provide gaps for water to

diffuse into, thus causing a degradation in the composite resin.²¹ G-aenial PosteriorTM did show a smaller reduction in flexural strength; however, this material contains prepolymerized filler that has a relatively poor integration in the resin matrix. This might cause a smaller filler density, thus increasing the sorption of liquid.¹²

The strength values found in the two microhybrids after immersion in citric acid, both in the mandarin orange concentration (2,19 pH) and lemon concentration (2 pH), were slightly larger but not significantly different from the strength values after immersion in distilled water; therefore, in this present study, the effect of pH on the results is not seen. The findings in this present study are in accordance with the studies done by Mohammadi *et al.* and Marghalani *et al.* that found flexural strength values of nanohybrids Filtek Z350TM and Tetric N-CeramTM were slightly larger after 0,02 N citric acid immersion compared to immersion in distilled water, but a significant difference was not found.¹⁸⁻²⁰ Resin composites undergo the biggest degradation and strength reduction in solutions that have a similar solubility parameter as the composite resin. The more similar the solubility parameter is, the easier it is for the solvent molecules to penetrate and extract the resin components.¹⁶ Studies have shown that distilled water has a solubility parameter value of $[(4.8 \times 10^{-4} \text{ J/m}^3)^{1/2}]$, which isn't similar with composite resin parameters, and therefore the degradation effect of distilled water isn't as bad as solvents that have a similar solubility parameter with composite resins.²² Based on this present study, the pH difference doesn't seem to have a significant effect on the strength reduction of the composite resins. The solubility parameter of the solvents seems to have the most significant effect on the degradation and strength reduction of the composites therefore, citric acid solutions seem to have a solubility parameter that isn't similar to that of composite resins, just like distilled water.²²

The strength values after immersion in the mandarin orange concentration of citric acid (2,19 pH) and lemon concentration of citric acid (2 pH) are almost the same, despite a slightly lower pH and a bigger concentration of acid in the lemon concentration solvent. This shows that in the current study, a significant

effect of pH on the strength reduction of composite resins wasn't seen.

Sorption and solubility values can explain the reduction in strength in composite resins. The bigger the sorption, the bigger the amount of solvent that is absorbed by the composite resin. The higher the solubility, the more unreacted monomer is released from the composite resin into the solvent. Higher sorption and solubility values usually correlate with a bigger strength reduction in composite resins.¹⁶ Sunbul *et al.* found a sorption value of 1.24% and a solubility of 0.09% for Genial Posterior™ after 180 days of immersion in distilled water and a 120 day reconditioning period.²³ For Filtek Z250™, Alshali *et al.* found a sorption value of 1,19% and a solubility of 0,33% after a 360 day immersion in distilled water and a 90 day reconditioning period.²¹ However, a study hasn't been done that compares the sorption and solubility of these materials at the same time after immersion in distilled water and citric acid; therefore, a study regarding this could be done so that it can further explain the results found in this current study.

CONCLUSION

The microhybrid composite resin G-aenial Posterior™ had a lower flexural strength value compared with the microhybrid Filtek Z250™. A significant reduction in flexural strength is seen in both microhybrid composite resins after immersion in distilled water and citric acid solutions.

REFERENCES

1. Sakaguchi R, Powers J. Craig's Restorative Dental Materials. 13th ed. USA: Mosby; 2012.
2. Ferracane JL. Resin composite - State of the art. *Dent Mater.* 2011;27(1):29–38.
3. Heintze SD. Features of a Good Composite Material. *J Cosmet Dent.* 2013;28(4):108–19.
4. G-aenial Anterior & Posterior technical manual. 2012. Available from: https://cdn.gceurope.com/v1/PID/gaenial/manual/MAN_G-aenial_Anterior-Posterior_en.pdf. [Last Accessed 2 March 2018]
5. Carrilho M, D'Alpino PHP. Dental Composite Materials for Direct Restorations. 1st ed. Switzerland: Springer; 2018.
6. Blackham JT, Vandewalle KS, Lien W. Properties of Hybrid Resin Composite Systems Containing Prepolymerized Filler Particles. *Oper Dent.* 2009;34(6):697–702.
7. Sideridou ID, Karabela MM, Bikiaris DN. Aging studies of light cured dimethacrylate-based dental resins and a resin composite in water or ethanol/water. *Dent Mater.* 2007; 23(9): 1142–9.
8. Eweis AH, Yap AUJ, Yahya NA. Dynamic analysis of bulk-fill composites: Effect of food-simulating liquids. *J Mech Behav Biomed Mater.* 2017;74(June):183–8.
9. Laboratory Chemical Safety Summary. Citric acid | C6H8O7 - PubChem. 2018.
10. ISO 4049_2009 - Dentistry -- Polymer-based restorative materials. Geneva: International Organization for Standardization. 2009. Available from: <https://www.iso.org/standard/42898.html>. [Last Accessed 1 November 2018]
11. K.Anusavice, C.Shen HR. Phillips Science of Dental Materials. 12th ed. USA: Elsevier Ltd; 2012.
12. Randolph LD, Palin WM, Leloup G. Filler characteristics of modern dental resin composites and their influence on physico-mechanical properties. *Dent Mater.* 2016;32(12):1586–99.
13. Huang Q, Huang S, Liang X, Qin W, Liu F, Lin Z, et al. The antibacterial, cytotoxic, and flexural properties of a composite resin containing a quaternary ammonium monomer. *J Prosthet Dent.* 2018;1–8.
14. Noraihan T, Tuan A, Mohamad D, Akil H, Ab I. Water sorption characteristics of restorative dental composites immersed in acidic drinks. *Dent Mater.* 2012;28(6):e63–70.
15. Hengtrakool C, Kukiattrakoon B, Kedjarune-leggat U. Effect of Naturally Acidic Agents on Microhardness and Surface Micromorphology of Restorative Materials. *Eur J Dent.* 2011;5(January):89–100.
16. Ferracane JL. Hygroscopic and hydrolytic effects in dental polymer networks. *Dent Mater.* 2006;

- 22(3):211–22.
17. Akova T, Ozkomur A, Uysal H. Effect of food-simulating liquids on the mechanical properties of provisional restorative materials. *Dent Mater.* 2006;22(12):1130–4.
18. Marghalani HY. Effect of chemical solvents on flexural properties of bulk-fill composites. *Dent Mater.* 2018;31:e15–6.
19. Yesilyurt C, Yoldas O, Altintas SH, Kusgoz A. Effects of food-simulating liquids on the mechanical properties of a silorane- based dental composite. *Dent Mater J.* 2009;28(3):362–7.
20. Mohammadi E, Pishevar L, Boroujeni PM. Effect of food simulating liquids on the flexural strength of a methacrylate and silorane-based composite. *PLoS One.* 2017;12(12):1–8.
21. 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;43(12):1511–8.
22. Münchow EA, Ferreira ACA, Machado RMM, Ramos S, Rodrigues-junior SA, Zanchi CH. Effect of Acidic Solutions on the Surface Degradation of a Micro-Hybrid Composite Resin. *Braz Dent J.* 2014;25(4):321–6.
23. Sunbul H Al, Silikas N, Watts DC. Surface and bulk properties of dental resin- composites after solvent storage. *Dent Mater.* 2016;32(8):987–97.