# Hardness of Flowable Resin Composite from Rice Husk

*Nazrul M. Yusoff, Yanti Johari*, Ismail Ab Rahman, Dasmawati Mohamad, Mohd Fadhli Khamis, Adam Husein, Zaihan Ariffin*
*School of Dental Sciences, Health Campus, Universiti Sains Malaysia, 16150 Kubang Kerian, Kelantan.*

**Corresponding author: yjohari@usm.my*

ABSTRACT

*The aim of this study was to evaluate the hardness of two experimental dental flowable resin composites (FRCs) from rice husk in comparison to other commercial flowable resin composites. The nanohybrid silica used as the filler for the experimental FRCs was extracted from rice husk. Two commercial FRCs namely Filtek Z350 flow and Tetric N flow and the experimental FRCs with different loading of Bis-GMA at 50 % (EC50B) and 40 % (EC40B) were used. Ten cylindrical specimens (5 x 2mm) for each material were prepared in acrylic mould, light cured and polished. Prior to hardness test, all the composites were immersed in distilled water at 37 °C for 24 h. The Vickers' hardness number (VHN) was measured using Vickers' hardness tester and their surface morphology was investigated using scanning electron microscopy (SEM). The data was analyzed by one way ANOVA followed by Dunnett T3 post hoc test. Results showed that Filtek Z350 flow was statistically significantly higher in regards to VHN compared to the other FRCs tested. There was no statistically significant difference in VHN between the Tetric N flow and the two experimental FRCs. SEM showed a well distributed embedded spherical filler particle in all FRCs. In conclusion, the two experimental FRCs were comparable to Tetric N flow however Filtek Z350 flow exhibited the highest hardness. This was possibly attributed to different filler loading. Between the experimental composites, EC40B had a higher hardness which can be explained by dilution effect on monomer system. In general, mechanical properties improved with increased filler loading and dilution of base monomer.*

**Keywords:** *hardness, flowable resin composite, nanohybrid silica, rice husk.*

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## 1. Introduction

The introduction of flowable resin composite (FRC) has started in late 1996 [1] as the enhancement to putty like consistency of resin based composite by improving its handling ability. This less viscous composite resin is produced by reducing the filler content or altering the viscosity of the monomer mixture itself [2, 3]. As a less viscous material, it improves the wettability to the tooth surface so it can flow into the desired areas whereas the flexibility helps as a stress reliever during polymerization shrinkage of composite resin [4]. These flow characteristic and the syringe system contribute to the ease of placement hence proving useful during the restoration of cavity preparation with difficult access [5]. The major uses of flowable resin composite are preventive resin restorations, pit and fissure sealant, cavity liners, class II and class V abfraction lesions restoration [6].

Fundamentally, the FRC consists of three major different materials which are the organic matrix, filler and coupling agent to bond the filler to the organic matrix [7]. The filler and organic matrix affect its viscosity, handling, application and manipulation of the material. The mechanical properties also vary with different type and loading of fillers [8] and organic phases [9]. With reduced amount of fillers, the FRC has its drawbacks compared to the conventional composite, be it high curing shrinkage [10] and low physical and mechanical strength [11]. As a result, it might not be suitable in high stress areas.

The organic matrices used in dental composite are largely based on the methacrylate chemistry such as 2,2-bis[4-(2-hydroxy-3-methacryloyloxypropyl)phenyl]propane (Bis-GMA), ethoxylated Bis-GMA (EBPDMA), 1,6-bis-[2-methacryloyloxyethoxycarbonylamino]-2,4,4-trimethlhexane (UDMA), dodecanediol dimethacrylate ($D_3MA$) or triethyleneglycol dimethacrylate (TEGDMA) [9]. There are various types of resin, nevertheless the most commonly used by manufacturers are Bis-GMA and TEGDMA as the monomer and diluent respectively.

It has been known that Bis-GMA is stiffer than TEGDMA, so any alteration or dilution of the monomer can affect or lower its mechanical properties. However there is no definite correlation between the dilution and mechanical strength because other factors such as degree of conversion [12], refractive index and viscosity [13] of the monomer can also affect the mechanical properties. A study investigated the effect of viscosity on the degree of conversion showed resin viscosity has a marginal influence on the mechanical properties of composites [14]. Another study examined the influence of base monomer and its diluent on the degree of double bond conversion. It was found that a decrease in base monomer gave an increase in degree of double bond conversion but lowered the mechanical properties [15].

In this study, the FRC using nanohybrid silica from rice husk had been prepared by altering the monomer mixture to decrease its viscosity. Nanohybrid silica used as the filler had been prepared using the sol-gel method for dental composite fabrication according to Noushad and colleagues [16]. Thus, the aim of this study was to investigate the hardness of experimental FRC using nanohybrid silica extracted from rice husk in comparison to other commercial FRCs.

## 2. Materials and method

### 2.1. Fabrication of flowable resin composite

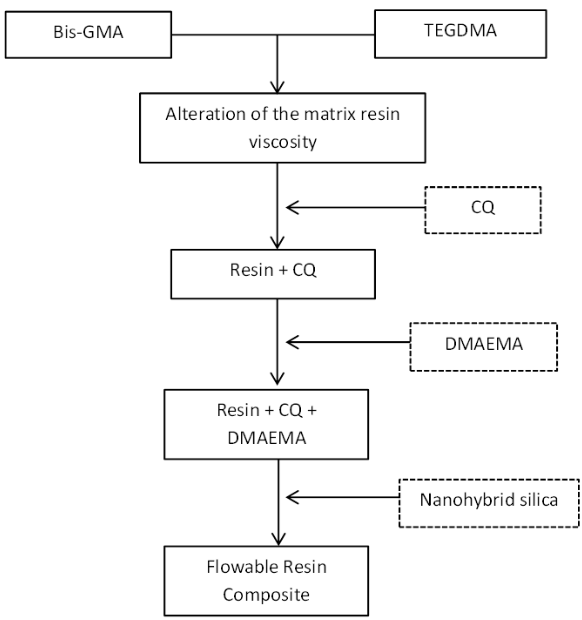



Figure 1. Flow chart for the preparation of flowable resin composite.

Figure 1 depicts the procedure to prepare FRC comprising of nanohybrid silica extracted from rice husk. The Bis-GMA (Esstech, USA) as a base monomer was manually mixed with TEGDMA (Esstech, USA) as a diluent at various concentrations to achieve different levels of viscosity. Two experimental composites consisting of Bis-GMA and TEGDMA at ratio of

50:50 (EC50B) and 40:60 (EC40B) were made. Both composites had the same amount of filler (nanohybrid silica, 50 wt. %), 0.02 g of CQ (Merck, Germany) and two drops of DMAEMA (Merck, Germany). Nanohybrid silica from rice husk was synthesized according to Noushad and colleagues [16]. A disposable 1 mL B-D syringe (Becton Dickinson & Co. Franklin Lakes, New Jersey) without a needle tip was filled with the experimental FRCs and wrapped with aluminum foil. The composition of the tested FRCs is shown in Table 1. Information of Filtek Z350 flow and Tetric N flow were obtained from the manufacturers.

Table 1. Composition of flowable resin composites investigated in this study.

| | Composition | | |
|---|---|---|---|
| **Material** | **Matrix** | **Filler** | **Filler content** |
| Filtek Z350 flow | Bis-GMA<br>UDMA<br>TEGDMA | 0.1-0.5 µm ytterbium triflouride<br>20, 75 nm silica,<br>4-11 nm Zirconia | (65 wt. %) |
| Tetric N flow | Bis-GMA<br>TEGDMA | 40-3000 nm barium glass, ytterbium triflouride, highly dispersed silica and mixed mode | (63 wt. %) |
| EC50B | Bis-GMA (50 wt. %)<br>TEGDMA (50 wt. %) | 48-534 nm nanohybrid silica | (50 wt. %) |
| EC40B | Bis-GMA (40 wt. %)<br>TEGDMA (60 wt. %) | 48-534 nm nanohybrid silica | (50 wt. %) |

### 2.2. Characterization

Ten specimens for each studied group were prepared in acrylic moulds (5 x 2 mm), with placement of Mylar strips over and below the mould before light curing for 40 s. They were stored in distilled water for 24 h at 37 ℃. The hardness was tested using a Vickers' hardness tester (Model VM 50, FIE) under 1 kg load for 15 s dwell time. Three indentations were made for each sample.

Surface morphology of the samples and the distribution of fillers in the composites were examined using a scanning electron microscope (SEM) (Fei, Quanta FEG 450) operating at 5 kV under low vacuum.

Data was statistically analyzed using one way ANOVA. Dunnett T3 post hoc test was used to determine the difference in hardness between the types of flowable resin composite. The significance level was set at 5%.

## 3. Result and Discussion

Hardness is influenced by material composition such as the type and size of the fillers and the monomers used. In this study, the hardness of two experimental FRCs, EC50B and EC40B containing the same amount of fillers but different amount of base monomer (50 % and 40 % Bis-GMA) were tested and compared to the commercial FRCs.

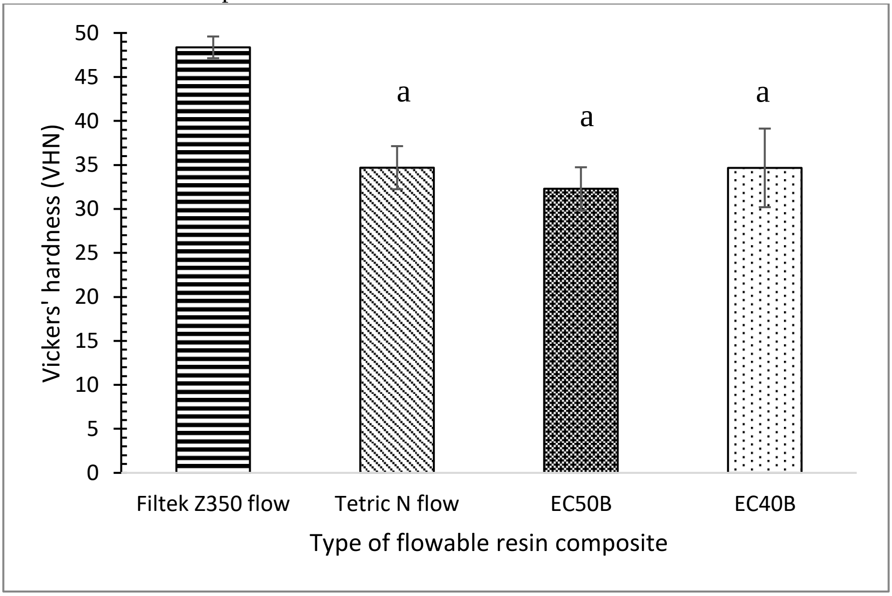
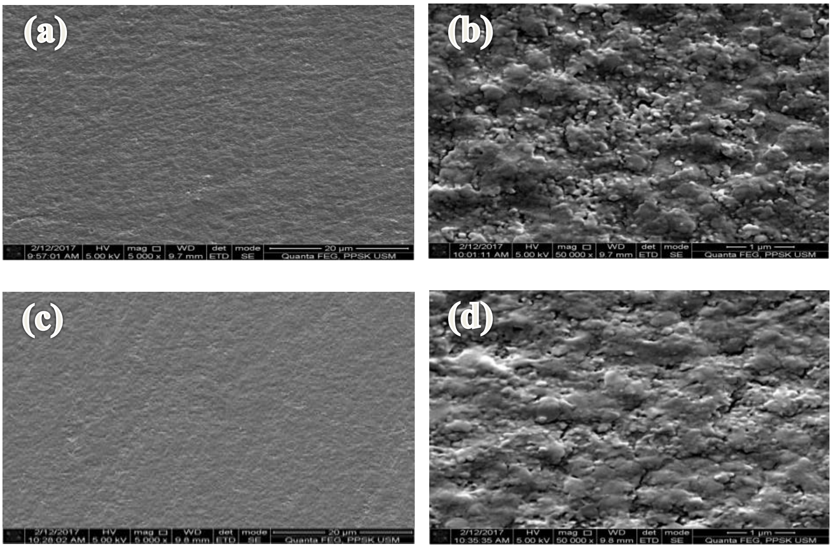



Figure 2. Vickers' hardness of the tested flowable resin composites. Letter 'a' depicts statistically significant different relationship with Filtek Z350 flow ($p$<0.05).

Figure 2 and Figure 3 present Vicker's hardness and surface morphology of all FRCs respectively. SEM showed a well distributed embedded spherical filler particles in the FRCs. All FRCs in the study showed comparable surface morphology at lower magnification (5000x) in Figure 3(a), (c), (e), and (g) and their differences can be clearly seen at higher magnification (50000x) in Figure 3 (b), (d), (f) and (h). One way ANOVA showed that there were statistically significant differences in the hardness of the four groups [$F(3,36) = 64.2$; $p<0.001$]. A Dunnett T3 post hoc test revealed that the hardness of Filtek Z350 flow was statistically significantly greater compared to other tested flowable composites ($p<0.05$). The hardness values as mean ± SD were 48.4 ± 1.24, 34.7 ± 2.45, 32.3 ± 2.44 and 34.5 ± 4.47 (VHN) for Filtek Z350 flow, Tetric N flow, EC50B and EC40B respectively. This was probably due to its high filler loading as shown in Table 1. The manufacturer has claimed that Filtek Z350 flow contains zirconia filler, adding an extra value to the hardness as zirconia is well known for its good mechanical strength [17]. In addition, Filtek Z350 flow comprised the smallest particle which can be seen by its smooth surface appearance under SEM (Figure 3(f)). Furthermore, the fillers vary in size thus giving a better packing arrangement due to the smaller fillers occupy the gap in between the larger one.



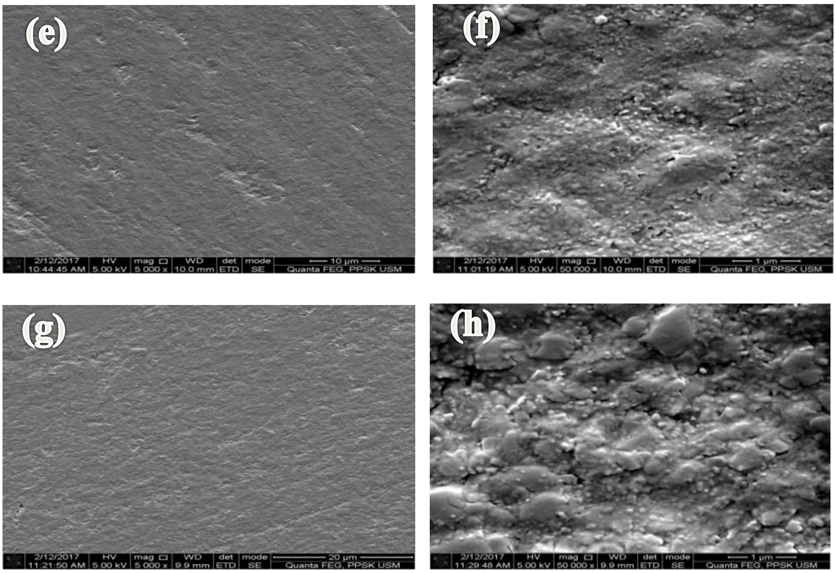



Figure 3. Surface morphology of the flowable resin composites (a) EC40B (5000x), (b) EC40B (50 000x), (c) EC50B (5000x), (d) EC50B (50 000x), (e) Filtek Z350 flow (5000x), (f) Filtek Z350 flow (50 000x), (g) Tetric N flow (5000x) and (h) Tetric N flow (50 000x).

Although the filler loading of experimental FRCs (50 wt. %), were less than Tetric N flow (63 wt. %), the result of hardness showed no statistically significant different ($p>0.05$). This distinction can be explained by their filler size. The smaller filler size of the experimental composites (EC40B and EC50B) as shown in Figure 3, has allowed the transmittance of visible light more efficiently to initiate the crosslinking process of the monomer system. This finding was in agreement with previous study where smaller particle increased the hardness of the composite [18]. Smaller filler size also allowed more densely pack resin composite. Both of the factors gave extra strength to experimental FRCs, making it comparable to Tetric N flow. Under SEM, Tetric N flow yielded large agglomerates of filler particles compared to the experimental composites (Figure 3(h)).

Less viscous composite resin can be attained by altering the monomer mixture itself. Diluting the monomer can be achieved through decreasing Bis-GMA while increasing the less viscous TEGDMA. Theoretically, stiffer and higher molecular weight monomer of the experimental flowable composite which contains 50 % Bis-GMA should give better hardness value but this was not observed in this study as shown in

Figure 2. This is possibly attributed to the degree of conversion of the monomer system. The degree of conversion displayed an increase trend with the dilution of Bis-GMA as it has lower limiting degree of conversion compared to TEGDMA [12]. Dimethacrylates as in TEGDMA show relatively high degree of conversion due to favourable stereochemistry [19]. Therefore, in the case of similar filler loading, the composite which consists of higher portion of TEGDMA has higher mechanical strength as demonstrated in Figure 2. This finding was corroborated by other studies [7, 14] which had demonstrated the effects of base monomer dilution on hardness of resin composites. In both studies, dilution of the base monomer could lead to increase in hardness of resin composites in their study.

## 5. Conclusion

In the present work, FRCs fabricated using nanohybrid silica from rice husk show a comparable hardness to commercial FRC namely Tetric N flow. The dilution of Bis-GMA resulted in an increase of hardness due to higher portion of TEGDMA which enhanced the degree of conversion. This is the first step to evaluate one of the mechanical strength of experimental flowable resin composite to ensure that the new product is comparable to the commercial product. It is hoped that it will bring another potential sustainable product in dentistry.

## Acknowledgement

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## 6. References

[1] S. Beun, C. Bailly, J. Devaux, and G. Leloup, “Rheological properties of flowable resin composites and pit and fissure sealants,” Dental Materials 24(4), 548-555 (2008).

[2] N. Attar, L. E. Tam, and D. McComb, “Flow, strength, stiffness and radiopacity of flowable resin composites,” J. Canadian Dental Association 69(8), 516-21 (2003).

[3] P. C. Moon, M. S. Tabassian, and T. E. Culbreath, “Flow characteristics and film thickness of flowable resin composites,” Opreative Dentistry 27(3), 248-53 (2002).

[4] A. Hervas-Garcia, M. A. Martinez-Lozano, J. Cabanes-Vila, A. Barjau-Escribano, and P. Fos-Galve, “Composite resins. A review of

the materials and clinical indications," Medicina Oral Patologia Oral y Cirugia Bucal 11(2), 215-20 (2006).

[5] K. Al-Ahdal, N. Silikas, and D. C. Watts, "Rheological properties of resin composites according to variations in composition and temperature," Dental Materials 30(5), 517-524 (2014).

[6] K. Baroudi and J. C. Rodrigues, "Flowable Resin Composites: A Systematic Review and Clinical Considerations," J. Clinical Diagnostic Research 9(6), 18-24 (2015).

[7] S. G. Pereira, R. Osorio, M. Toledano, and T. G. Nunes, "Evaluation of two Bis-GMA analogues as potential monomer diluents to improve the mechanical properties of light-cured composite resins," Dental Materials 21(9), 823-830 (2005).

[8] S. Beun, C. Bailly, J. Devaux, and G. Leloup, "Physical, mechanical and rheological characterization of resin-based pit and fissure sealants compared to flowable resin composites," Dental Materials 28(4), 349-59 (2012).

[9] N. Moszner and U. Salz, "New developments of polymeric dental composites," Progress in Polymer Science 26(4), 535-576 (2001).

[10] K. M. C. Oliveira, A. C. R. A. Lancellotti, R. A. Ccahuana-Vásquez, and S. Consani, "Influence of filling techniques on shrinkage stress in dental composite restorations," J. Dental Sciences 8(1), 53-60 (2013).

[11] L. M. Petrovic, D. M. Zorica, I. L. Stojanac, V. S. Krstonosic, M. S. Hadnadjev, and T. M. Atanackovic, "A model of the viscoelastic behavior of flowable resin composites prior to setting," Dental Materials 29(9), 929-934 (2013).

[12] I. Sideridou, V. Tserki, and G. Papanastasiou, "Effect of chemical structure on degree of conversion in light-cured dimethacrylate-based dental resins," Biomaterials 23(8), 1819-1829 (2002).

[13] R. M. Ku, C. C. Ko, C. M. Jeong, M. G. Park, H. I. Kim, and Y. H. Kwon, "Effect of flowability on the flow rate, polymerization shrinkage, and mass change of flowable composites," Dental Materials 34(2), 168-74 (2015).

[14] L. Musanje and J. L. Ferracane, "Effects of resin formulation and nanofiller surface treatment on the properties of experimental hybrid resin composite," Biomaterials 25(18), 4065-4071 (2004).

[15] C. J. E. Floyd and S. H. Dickens, "Network structure of Bis-GMA- and UDMA-based resin systems," Dental Materials 22(12), 1143-1149 (2006).

[16] M. Noushad, I. Ab Rahman, A. Husein, and D. Mohamad, "Nanohybrid dental composite using silica from biomass waste," Powder Technology 299, 19-25 (2016).

[17] I. Denry and J. R. Kelly, "State of the art of zirconia for dental applications," Dental Materials 24(3), 299-307 (2008).

[18] K. Fujita, T. Ikemi, and N. Nishiyama, "Effects of particle size of silica filler on polymerization conversion in a light-curing resin composite," Dental Materials 27(11), 1079-1085 (2011).

[19] A. Peutzfeldt, "Resin composites in dentistry: the monomer systems," European J. Oral Sciences 105(2), 97-116 (1997).