Mechanical Properties of Newly Developed Flowable Composite Derived from Rice Husk at Different Monomer Ratios

Nor Ain Fatihah Azlisham, Dasmawati Mohamad, Mohd Firdaus Yhaya
Unit of Biomaterials, School of Dental Sciences, Universiti Sains Malaysia Health Campus, 16150 Kubang Kerian, Kelantan, MALAYSIA

*Yanti Johari**
Unit of Prosthodontics, School of Dental Sciences, Universiti Sains Malaysia Health Campus, 16150 Kubang Kerian, Kelantan, MALAYSIA
**yjohari@usm.my*

Zuliani Mahmood
Unit of Paediatric Dentistry, School of Dental Sciences, Universiti Sains Malaysia Health Campus, 16150 Kubang Kerian, Kelantan, MALAYSIA

ABSTRACT

An ideal flowable composite (FC) requires good mechanical properties to fulfill its role as a dental restorative material and ensure durability and clinical efficacy, with these properties being influenced by different monomer ratios. The study aimed to evaluate the mechanical properties of the newly developed FC on Vickers hardness, flexural strength, and flexural modulus at different monomer ratios. The newly developed FC employed nanohybrid silica from rice husk and zirconia as fillers, and the ratio of urethane dimethacrylate to triethylene glycol dimethacrylate (UDMA:TEGDMA) at 20:80, 30:70, 50:50, 60:40, 80:20 and 90:10 as monomers. Filtek Supreme Ultra Flowable Restorative, Revolution Formula 2, and G-aenial Universal Flo served as control groups. The data were statistically analyzed using one-way ANOVA with post-hoc Bonferroni test. Generally, the higher UDMA ratio in the newly developed FCs exhibited higher mechanical properties. All newly developed FCs demonstrated a significant decrease in Vickers hardness than control groups. The flexural strength values at ratios of 50:50, 60:40, 80:20, and 90:10 were comparable with control groups and passed the International Organization for Standardization 4049 requirement. Meanwhile, the flexural

ISSN 1823-5514, eISSN 2550-164X
Received for review: 2024-01-04
Accepted for publication: 2024-05-06
Published: 2024-09-15

https://doi.org/10.24191/jmeche.v21i3.27347

modulus of the newly developed FC at the ratio of 90:10 was comparable to Revolution Formula 2. In summary, the differences in monomer ratios affect the mechanical properties of the newly developed FCs. The improved mechanical properties at ratios of 80:20 and 90:10 could potentially substitute sustainable dental restorative materials, subjected to further investigations.

Keywords: *Flexural Modulus; Flexural Strength; Flowable Composite; Nanohybrid Silica; Vickers Hardness*

Introduction

Flowable composites (FCs) primarily consist of inorganic fillers and organic resin monomers. These composites typically have a filler loading ranging from 37% to 53% by volume, and a higher proportion of monomers. As a result, they exhibit reduced viscosities compared to conventional resin composites, imparting a flowable nature that simplifies their handling. They spread uniformly and intimately across prepared tooth surfaces, facilitating their application [1].

Fillers play an important role in strengthening the composites. Among these fillers, silica, also known as silicon dioxide (SiO_2), is one of the most commonly used fillers in dental restorative materials as it possesses good stability, robust mechanical properties, refractive index compatibility with the resin monomers matrix, and ease of production [2]. Remarkably, the locally produced silica derived from rice husks has demonstrated promising potential as a filler in the development of resin composites in general [3]-[7] and flowable composites in particular [8]-[9]. Similarly, synthetic silica has been effectively utilized as a filler in dental composite materials, offering adaptability through various shapes [10] and surface treatments [11], and contributing to enhanced mechanical properties [12]. Both locally produced silica from rice husk and synthetically made silica have been used as fillers, with the silica derived from rice husk promoting the development and production of green-based flowable composite. However, the use of elements with low atomic weight (Z), such as silicon (Si; Z = 14), results in radiolucent materials [13], which can pose challenges in dental radiography. To address this issue, incorporating radiopacifying agents as additional filler would be of beneficial approach as the radiopacifying agents will enhance the material's radiopacity, enabling accurate diagnoses during radiographic examinations [14]-[15]. At present, zirconium dioxide (ZrO_2), or zirconia, is the preferred radiopacifying agent due to its high atomic number of zirconium (Z = 40), which confers the radiopaque characteristics to dental materials while also demonstrating good biocompatibility, and physical and mechanical properties [16]-[17].

The monomer system in dental restorative materials typically consists of base monomers, including bisphenol A glycidyl methacrylate (Bis-GMA) and urethane dimethacrylate (UDMA). As the base monomers are highly viscous, the diluent monomer triethylene glycol dimethacrylate (TEGDMA), which has a low molecular weight and viscosity, is needed as a diluent to the basic monomers [18]. The combination of these monomers is important as it serves as a reaction medium for the dispersion of fillers and ensures proper clinical handling. Bis-GMA, with its high viscosity and rigid molecular structure, offers several benefits in dental composites, including less volumetric shrinkage, higher modulus, and reduced toxicity due to its lower volatility and diffusivity into tissues [19]. On the other hand, UDMA offers various advantages over Bis-GMA, such as structural flexibility, low molecular weight, reduced water sorption [20], lower viscosity [21], and a closer refractive index to that of silica [22]. The differences in the ratio of monomers play a role in determining the flowability of the resulting materials, thus exerting a substantial influence on the performance of dental resins [23].

To date, there have been no studies evaluating the effect of using UDMA:TEGDMA at different ratios in fabricating newly developed FCs. Nevertheless, incorporating a radiopacifying agent into the newly developed FC to render the material radiopaque is advantageous. Given that zirconia is a well-established radiopacifying agent commonly used in dental restorative materials [24], its incorporation becomes imperative. While ensuring the radiopacity of the newly developed flowable composite, the evaluation of mechanical properties at different ratios should not be overlooked. Therefore, the study aims to evaluate the mechanical properties of the newly developed flowable composite, which incorporates locally produced silica derived from rice husk and zirconia fillers at varying ratios of UDMA and TEGDMA. The development of a flowable composite that is radiopaque and has good mechanical properties is important to fulfill its role as a restorative material and enhance its long-term clinical performance and longevity.

Materials and Methods

Preparation of newly developed FCs

The ratio of filler-to-monomer of the newly developed FC was set at 50:50, as the selected ratio was found to offer the desired flowable characteristic compared to commercial counterparts. The ratios higher than this render it similar to a conventional packable dental composite, while ratios lower than this define it more as a dental fissure sealant material, as it becomes excessively diluted.

The newly developed FC was prepared by incorporating locally produced nanohybrid silica (Patent ID:MY-187327-A) and zirconia (230693, Sigma Aldrich) as fillers at a ratio of 40:10. In this study, zirconium oxide or

zirconia, was incorporated at 10% (w/w) and served as the radiopacifying agent. Urethane dimethacrylate (UDMA) (Sigma Aldrich) was used as the base monomer, while triethylene glycol dimethacrylate (TEGDMA) (Sigma Aldrich) served as the diluent to this base monomer. A preliminary study on flowability covering a wide range of ratios, from low (10:90) to high (100:0) ratios of UDMA:TEGDMA, was initially conducted. However, only a few ratios were selected as they possess flowability levels comparable to commercial FCs. The mixture of these monomers, UDMA:TEGDMA, was manipulated at selected ratios of 20:80, 30:70, 50:50, 60:40, 80:20, and 90:10. These ratios were further classified as low, medium, and high. Their flowability levels, where 20:80 and 30:70 were classified as high flow, 50:50 and 60:40 were classified as medium flow, and 80:20 and 90:10 were classified as low flow. Photoinitiators, 0.5 wt% camphorquinone (CQ) (Merck, Germany) and 0.5 wt% 2-(dimethylamino)ethyl methacrylate (DMAEMA) (Merck, Germany), were then added to the resin matrix. The resin matrix and fillers were homogeneously mixed to obtain the newly developed FC in the injectable form. The schematic diagram illustrating the fabrication process of the newly developed FC is shown in Figure 1.

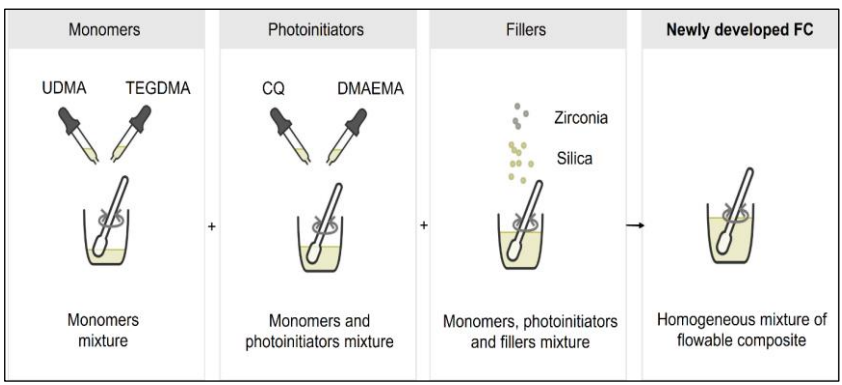

Figure 1: The fabrication process of the newly developed FC

The image of the newly developed FC produced by the dental X-ray imaging unit exhibited radiopaque properties compared to nanohybrid silica alone, confirming the role of zirconia as a radiopacifying agent. Figure 2 shows the radiographic image of zirconia-incorporated silica and nanohybrid silica.

Revolution Formula 2 Flowable Light Cure Composite (Kerr Corporation), Filtek Supreme Ultra Flowable Restorative (3M, ESPE), and G-aenial Universal Flo Universal Light-cured Radiopaque Flowable Composite (GC Corporation, Japan) served as control groups. The composition of the newly developed FCs and control groups is summarized in Table 1.

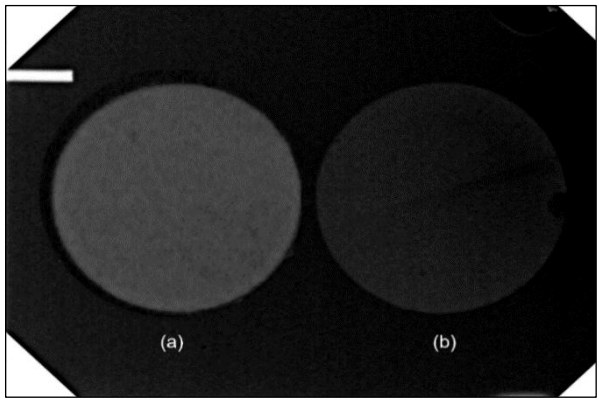

Figure 2: Radiographic image of (a) zirconia-incorporated silica; (b) nanohybrid silica

Table 1: Composition of the newly developed FCs and control groups used in the study

Sample type/Abbreviation	Fillers	Monomers	
		Type	Ratio
EF20U	Nanohybrid silica, zirconia	UDMA/TEGDMA	20:80
EF30U		UDMA/TEGDMA	30:70
EF50U		UDMA/TEGDMA	50:50
EF60U		UDMA/TEGDMA	60:40
EF80U		UDMA/TEGDMA	80:20
EF90U		UDMA/TEGDMA	90:10
Revolution Formula 2	Propriety glass filler	Bis-GMA, TEGDMA	NIL
Filtek Supreme Ultra Flowable Restorative	Silica, zirconia, ytterbium trifluoride	Bis-GMA, TEGDMA, Procrylat	NIL
G-aenial Universal Flo	Silicone dioxide, strontium glass	UDMA, bisphenol A ethoxylate dimethacrylate (Bis-MEPP), TEGDMA	NIL

Vickers hardness

A total of 63 samples (n = 7/group) were prepared in a disk shape (5 mm diameter; 2 mm thickness) for Vickers hardness testing. The samples were irradiated for 20 s both on top and bottom surfaces using a LED light-curing unit (Elipar^{TM} S10, 3M ESPE, Germany) with a light intensity of ~1500 kW/cm^2 and then stored in a water bath at 37 °C for 24 hours before testing. The Vickers hardness number (VHN) of all newly developed FCs and control

groups was recorded using a Vickers hardness tester, with a 1 kg load and a 15-second dwelling time.

Flexural strength and modulus

The flexural strength was determined in accordance with the International Organization for Standardization (ISO) 4049 [25]. A total of 63 samples (n = 7/group) were prepared in a bar shape (25 mm length, 2 mm thickness, 2 mm width) for flexural strength and modulus of elasticity testing. The entire length of the samples was irradiated for 40 seconds using a LED light-curing unit (Elipar^{TM} S10, 3M ESPE, Germany) with a light intensity of ~1500 kW/cm^2 in overlapping conditions for each section, for both the top and bottom of the samples according to ISO 4049. The prepared samples were stored in a water bath at 37 °C for 24 hours before testing. A universal testing machine equipped with a 20 kN cell (Shimadzu AGX Plus) was used, with a crosshead speed of 1.0 mm/min and a span length of 20 mm. The flexural strength in three-point bending was calculated using the following formula, while the modulus of elasticity was obtained from the linear part of the stress-strain curve.

$$Flexural\ strength = \frac{3Fl}{2bh^2} \quad (1)$$

where; F = force, l = span length, b = width of sample, h = thickness of sample.

Statistical analysis

The data were expressed as means and standard deviations and were statistically analyzed using Statistical Package for Social Science (SPSS) software, version 26 (IBM Corporation, USA). One-way analysis of variance (ANOVA) with a post-hoc Bonferroni test was used to determine the significant differences ($p<0.05$) between the newly developed FCs and the control groups.

Results and Discussion

The knowledge of the mechanical properties of dental restorative materials is important as it provides the correct indication for their applications and durability of restorations [26]. In the oral environment, restorations are subjected to stresses from mastication. Hardness serves as an initial indicator of surface changes during mastication and it is defined as the measure of resistance to permanent surface indentation or penetration. In contrast, flexural strength measures the ability of materials to bend before breaking, while the elastic modulus represents the stiffness of a material within the elastic range when exposed to flexural, compressive, or tensile forces. In this context, the elastic modulus can guide the selection of a restorative material with

deformation properties closely resembling those of the material it replaces [27].

Vickers hardness

Table 2 shows the Vickers hardness values of the newly developed FCs and control groups. The Vickers hardness values of the newly developed FCs exhibited an increasing trend, wherein higher ratios of UDMA were associated with increased Vickers hardness. Specifically, the Vickers hardness values of the newly developed FCs followed an ascending order of UDMA:TEGDMA ratios of 20:80 < 30:70 < 50:50 < 60:40 < 80:20 < 90:10. A significant decrease was demonstrated in the Vickers hardness values of the newly developed FCs at all ratios compared to the control groups of Revolution Formula 2, G-aenial and Filtek.

Table 2: Vickers hardness, flexural strength, and modulus of the control groups and newly developed FCs

Sample type	Vicker hardness (VHN)	Flexural strength (MPa)	Flexural modulus (GPa)
Revolution Formula 2	35.06 ± 2.65^{e}	100.38 ± 10.05^{ab}	3.78 ± 0.09^{a}
G-aenial Universal Flo	46.59 ± 0.97^{a}	78.55 ± 20.17^{abc}	5.67 ± 0.17^{f}
Filtek Supreme Ultra Flowable Restorative	46.71 ± 0.51^{a}	97.94 ± 10.10^{ab}	6.21 ± 0.09^{g}
20:80	20.60 ± 0.00^{f}	57.41 ± 12.38^{e}	2.73 ± 0.10^{h}
30:70	22.70 ± 0.39^{g}	66.31 ± 10.35^{cd}	2.78 ± 0.19^{e}
50:50	26.62 ± 0.34^{bcd}	79.61 ± 13.32^{abc}	3.14 ± 0.18^{cd}
60:40	26.17 ± 0.29^{bcd}	83.08 ± 11.11^{abc}	3.21 ± 0.12^{cd}
80:20	27.89 ± 0.94^{bc}	95.24 ± 6.93^{ab}	3.35 ± 0.05^{bc}
90:10	28.01 ± 0.36^{b}	102.14 ± 7.09^{a}	3.58 ± 0.08^{ab}

Mean values ± standard deviations in parentheses. The same superscript letters in a column denote no statistical significance according to the Bonferroni test ($p>0.05$).

The ratios that contain high UDMA within the resin matrix are associated with increased Vickers hardness values, which are attributed to the chemical structure of UDMA. UDMA is a base monomer that is flexible and contains two urethane links in the structure which are able to form hydrogen bonds. The high UDMA ratio in the resin matrix provides many available urethane linkages. These linkages may have improved the intermolecular interactions, mainly hydrogen bonding, which are beneficial for enhancing the mechanical properties of cross-linked dimethacrylate systems [21], [28]-[29]. The formation of this hydrogen bond can increase the mechanical properties of dental composites [20].

The addition of TEGDMA monomer as the diluent to the base monomer has contributed to achieving a higher degree of conversion and improved filler

homogenization [30]. However, it should be noted that the high amount of TEGDMA ratio deteriorated the Vickers hardness of the newly developed FCs in this study, likely attributable to primary cyclization. Higher TEGDMA ratios are associated with an increased likelihood of primary and secondary cyclization reactions within the polymer network. These reactions could potentially affect the polymer packing density, resulting in a more loosely or less densely packed network structure [31]. This phenomenon is proven in the ratios of 20:80 and 30:70, where Vickers hardness values were the lowest.

Moreover, TEGDMA is also characterized by its high hydrophilicity and greater susceptibility to polymerization shrinkage [32]. Given that the newly developed FCs were immersed in distilled water for 24 hours prior to testing, it is possible that there was a significant uptake of water into the resin matrix, resulting in a decrease in mechanical properties. This aligns with the findings of Sideridou et al. [33], who reported that TEGDMA monomer exhibits a high water sorption value compared to UDMA monomer, which corroborates with the reduction of Vickers hardness in this study at high TEGDMA content, 20:80 and 30:70. Conversely, as the ratio of TEDGMA decreases, specifically at 80:20 and 90:10, there is an enhancement in Vickers hardness.

As for the control groups, a significant increase in Vickers hardness compared to the newly developed FCs can be attributed to several factors, including a higher filler-to-monomer ratio, type of fillers, and type of monomers used. According to the manufacturer's instructions, Revolution Formula 2, Filtek, and G-aenial contain 60%, 65%, and 69% filler content, respectively. In contrast, the newly developed FCs contain 50% filler content, which is lower than that of the control groups. It is postulated that the filler packing density in the control groups has improved more than in the newly developed FCs, leading to an increase in Vickers hardness values. Several studies have reported that the filler content had a notable impact on the mechanical properties [34]-[35]. Additionally, different types of fillers are also used in the control groups which might have contributed to better mechanical properties, as shown in Table 1.

Furthermore, the organic resin matrix present in each control group varies significantly. Revolution Formula 2 and Filtek employ a mixture of common dimethacrylate monomers, including Bis-GMA and TEGDMA, with Filtek incorporating an additional monomer known as procrylat. Procrylat is a hydrophobic and low-viscosity resin monomer, and it is postulated that its inclusion helps in preventing excessive water sorption during the 24-hour storage at 37 °C. On the other hand, G-aenial contains a mixture of UDMA, TEGDMA, and Bis-MEPP. Bis-MEPP is considered a rigid monomer with greater hydrophobicity compared to UDMA [36]. This variation in resin monomer composition compared to other materials most likely contributes to the enhanced Vickers hardness observed in the control groups compared to the newly developed FCs.

Flexural strength and modulus

The flexural strength and modulus values of the newly developed FCs and control groups are presented in Table 2. In line with the observations for Vickers hardness, the flexural strength and modulus values of the newly developed FCs exhibited an increase with higher ratios of UDMA, conforming to the sequence of 20:80 < 30:70 < 50:50 < 60:40 < 80:20 < 90:10.

For flexural strength, the newly developed FC at ratios of 50:50, 60:40, 80:20, and 90:10 (ranging from 79.61 - 102.14 MPa) passed the minimum requirement set by the International Organization for Standardization (ISO) 4049, which is 80 MPa. The newly developed FC ratio of 50:50 was comparable to the control group of G-aenial, while 60:40 was comparable to both the G-aenial and Revolution Formula 2 control groups. The values for 80:20 and 90:10 ratios were comparable to all control groups. In contrast, the flexural strength values of newly developed FCs at 20:80 and 30:70 showed a significant decrease compared to all control groups. The highest flexural strength was achieved by the newly developed FC at the ratio of 90:10, followed by Revolution Formula 2 and Filtek.

Figure 3 presents the flexural stress-strain graph of the newly developed FCs at all ratios and the control groups. The stress-strain curve showed both the newly developed FCs and the control groups are brittle in behavior.

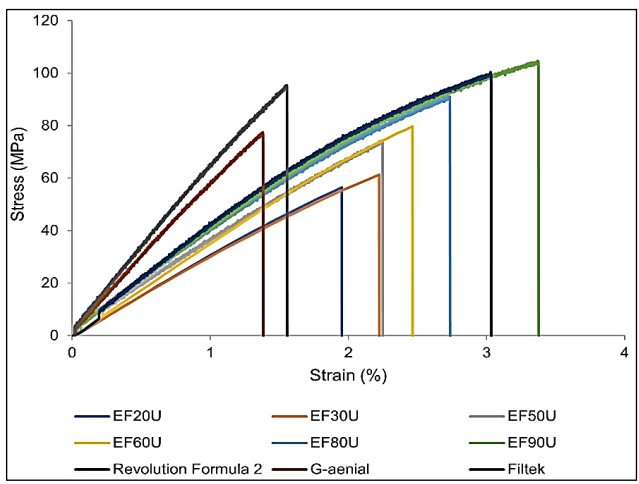

Figure 3: Stress-strain curve of the newly developed FCs and control groups

For flexural modulus, significant decreases were observed for the newly developed FCs when compared to all control groups, except at the 90:10 ratio, which showed a modulus value comparable to that of Revolution Formula 2.

Filtek had the highest flexural modulus, followed by G-aenial and Revolution Formula 2.

UDMA plays a vital role in determining the flexural strength and modulus of the newly developed FC, as a higher ratio of UDMA increases the flexural strength and modulus. The presence of urethane groups in UDMA facilitates the formation of hydrogen bonds. As the ratio of UDMA in the resin matrix increases relative to TEGDMA, more urethane linkages become available for hydrogen bonding interactions, thus enhancing the flexural strength and modulus of the newly developed FCs. A higher degree of conversion and a greater cross-linking density are postulated with an increased UDMA ratio, specifically at 80:20 and 90:10, resulting in a more rigid and densely packed polymer network, which contributes to increased flexural strength and modulus. Conversely, the higher ratio of TEGDMA, specifically at 20:80 and 30:70 within the resin matrix in the newly developed FCs decreased the flexural strength and modulus values. Such low flexural strength and modulus values of the newly developed FCs can also be explained with regard to primary cyclization, the high hydrophilicity of TEGDMA, and greater susceptibility of TEGDMA to polymerization shrinkage.

In this study, where the ratios of filler and monomers for the newly developed FCs were maintained at a consistent ratio of 50 wt%, we can reasonably infer that the interaction between the filler and resin matrix has been effectively established. Despite this, the control groups consistently demonstrated higher flexural strength and modulus values compared to the newly developed FCs. This disparity can be attributed to various factors, such as differences in filler-to-monomer ratio, types of fillers, and resin monomer compositions. The higher filler content in the control groups compared to the newly developed FCs enhanced the flexural strength and modulus. Additionally, the presence of resin monomers of procrylat in Filtek and Bis-MEPP in G-aenial which is hydrophobic in properties have resulted in less water sorption during the 24-hour incubation of samples in distilled water, thus giving the superior strength and modulus values compared to the newly developed FCs.

In summary, the knowledge and understanding of the mechanical properties of Vickers hardness, flexural strength, and modulus at different monomer ratios facilitate the selection and optimization of FCs. While ideal values for these properties remain undefined, especially for Vickers hardness and flexural modulus, prior research provides a range of values for comparison. Previous studies reported a range of Vickers hardness values for FCs, varying from 16 HV - 60 HV [37], 22 HV - 33 HV [38], to 35 HV - 50 HV [9]. For flexural strength, a minimum of 80 MPa was found to meet the requirement of ISO standards for polymer-based [25], while values of 90.5 MPa - 135.1 MPa [39] to 128.6 MPa - 162 MPa [40] were reported in previous studies. For flexural modulus, the values vary between 2.0 GPa - 8.6 GPa [37], 4.7 GPa - 7.6 GPa [39] and 7.51 GPa - 9.24 GPa [40]. Although not all values for newly

developed FCs corroborate with previous studies, this information serves as a valuable reference for future material improvement.

Conclusion

The study successfully incorporated silica and zirconia as fillers while varying the monomer ratio at 20:80, 30:70, 50:50, 60:40, 80:20, and 90:10. The variations in monomer ratios had a notable impact on the mechanical properties of the newly developed FCs, with all mechanical properties showing an increasing trend as the UDMA ratio increased. While most values exhibited a significant decrease compared to the control groups, the ratios of 80:20 and 90:10 for flexural strength and modulus were found to be comparable to those of the control groups. These newly developed FCs could be the next potential substitute for sustainable dental restorative materials, subjected to further investigations.

Contributions of Authors

The authors confirm the equal contribution in each part of this work. All authors reviewed and approved the final version of this work.

Funding

This work was supported by the Research University Grant from Universiti Sains Malaysia RUI (1001/PPSG/8012380).

Conflict of Interests

All authors declare that they have no conflicts of interest.

Acknowledgment

The first author received support from the Graduate Fellowship USM Scheme. The authors would like to acknowledge the facilities provided by the Biomaterials Research Laboratory of the School of Dental Sciences, Universiti Sains Malaysia Health Campus.

References

[1] K. Baroudi and J. C. Rodrigues, "Flowable resin composites: A systematic review and clinical considerations," *Journal of Clinical and Diagnostic Research*, vol. 9, no. 6, pp. 18–24, 2015.

[2] E. Habib, R. Wang, Y. Wang, M. Zhu, and X. X. Zhu, "Inorganic fillers for dental resin composites: present and future", *ACS Biomaterials Science and Engineering*, vol. 2, no. 1, pp. 1–11, 2016.

[3] N. S. C. Zulkifli, I. A. Rahman, D. Mohamad, and A. Husein, "A green sol-gel route for the synthesis of structurally controlled silica particles from rice husk for dental composite filler", *Ceramics International*, vol. 39, no. 4, pp. 4559–4567, 2013.

[4] M. Noushad, I. A. Rahman, A. Husein, and D. Mohamad, "Nanohybrid dental composite using silica from biomass waste", *Powder Technology*, vol. 299, pp. 19–25, 2016.

[5] S. S. L. Galvin, N. R. Ghani, N. H. Ismail, K. Singbal, T. Y. Noorani, and N. Mamat, "New experimental zirconia-reinforced rice husk nanohybrid composite and the outcome of its surface roughness and microhardness in comparison with commercialized nanofilled and microhybrid composite resins", *Contemporary Clinical Dentistry*, vol. 12, pp. 21–27, 2021.

[6] M. AL-Rawas, Y. Johari, D. Mohamad, M. F. Khamis, W. M. A. W. Ahmad, Z. Ariffin and A. Husein, "Water sorption, solubility, degree of conversion, and surface hardness and topography of flowable composite utilizing nano silica from rice husk," *Journal of Materials Research and Technology*, vol. 15, pp. 4173–4184, 2021.

[7] S. B. Sheng, R. Alawi, Y. Johari, N. A. Muttlib, M. H. Hussin, D. Mohamad, and M. I. Karobari, "Effects of fiber loading on mechanical properties of kenaf nanocellulose reinforced nanohybrid dental composite made of rice husk silica," *Journal of Functional Biomaterials*, vol. 14, no. 184, pp. 1–16, 2023.

[8] N. M. Yusoff, Y. Johari, I. A. Rahman, D. Mohamad, M. F. Khamis, and Z. Ariffin, "Hardness of flowable resin composite from rice husk," *Journal of Mechanical Engineering*, vol. 5, no. 2, pp. 181–190, 2018.

[9] N. M. Yusoff, Y. Johari, I. A. Rahman, D. Mohamad, M.F. Khamis, Z. Ariffin, and A. Husein, "Physical and mechanical properties of flowable composite incorporated with nanohybrid silica synthesised from rice husk," *Journal of Materials Research and Technology*, vol. 8, no. 3, pp. 2777–2785, 2019.

[10] N. Timpe, H. Fullriede, L. Borchers, M. Stiesch, P. Behrens, and H. Menzel, "Nanoporous silica nanoparticles with spherical and anisotropic shape as fillers in dental composite materials", *BioNanoMaterials*, vol. 15, no. 3–4, pp. 89–99, 2014.

[11] J. W. Kim, L. U. Kim, and C. K. Kim, "Size control of silica nanoparticles

and their surface treatment for fabrication of dental nanocomposites", *Biomacromolecules*, vol. 8, pp. 215–222, 2007.

[12] T. N. A. T. Rahim, D. Mohamad, A. R. Ismail, and H. M. Akil, "Synthesis of nanosilica fillers for experimental dental nanocomposites and their characterizations," *Journal of Physical Science*, vol. 22, no. 1, pp. 93–105, 2011.

[13] I. Porto, N. Honório, D. Amorim, A. Franco, L. Penteado, and A. Parolia, "Comparative radiopacity of six current adhesive systems," *Journal of Conservative Dentistry*, vol. 17, no. 1, pp. 65–69, 2014.

[14] R. B. Ermis, D. Yildirim, G. Yildiz, and O. Gormez, "Radiopacity evaluation of contemporary resin composites by digitization of images," *European Journal of Dentistry*, vol. 8, no. 3, pp. 342–347, 2014.

[15] N. M. Bahaa, D. I. El-Korashy, and D. I. Sherief, "The effect of adding ytterbium trifluoride on the radiopacity, compressive strength, setting time and bioactivity of biodentine : An in vitro study", *Brazilian Dental Science*, vol. 25, no. 3, pp. 1–11, 2022.

[16] C. Hu, J. Sun, C. Long, L. Wu, C. Zhou, and X. Zhang, "Synthesis of nano zirconium oxide and its application in dentistry," *Nanotechnology Reviews*, vol. 8, pp. 396–404, 2019.

[17] R. Li, C. Wang, S. Q. Ma, Z. H. Liu, C. C. Zhang, and Y. C. Sun, "High bonding strength between zirconia and composite resin based on combined surface treatment for dental restorations," *Journal of Applied Biomaterials & Functional Materials*, vol. 18, pp. 1–9, 2020.

[18] S. H. Dickens, J. W. Stansbury, K. M. Choi, and C. J. E. Floyd, "Photopolymerization kinetics of methacrylate dental resins," *Macromolecules*, vol. 36, no. 16, pp. 6043–6053, 2003.

[19] I. Sideridou, V. Tserki, and G. Papanastasiou, "Effect of chemical structure on degree of conversion in light-cured dimethacrylate-based dental resins", *Biomaterials*, vol. 23, no. 8, pp. 1819–1829, 2002.

[20] I. M. Barszczewska-Rybarek, M. W. Chrószcz, and G. Chladek, "Novel urethane-dimethacrylate monomers and compositions for use as matrices in dental restorative materials," *International Journal of Molecular Sciences*, vol. 21, no. 7, pp. 1–23, 2020.

[21] V. E. Gajewski, C. S. Pfeifer, N. R. Froes-Salgado, L. C. Boaro, and R. R. Braga, "Monomers used in resin composites : degree of conversion, mechanical properties and water sorption/solubility," *Brazilian Dental Journal*, vol. 23, no. 5, pp. 508–514, 2012.

[22] K. Fujita, N. Nishiyama, K. Nemoto, T. Okada, and T. Ikemi, "Effect of base monomer's refractive index on curing depth and polymerization conversion of photo-cured resin composites," *Dental Materials Journal*, vol. 24, no. 3, pp. 403–408, 2005.

[23] W. Li, K. Wang, Z. Wang, and B. Li, "Optimal resin monomer ratios for light-cured dental resins", *Heliyon*, vol. 8, no. e10554, pp. 1-7, 2022.

[24] S. O. Alsharif, H. M. Akil, N. A. A. El-Aziz, and Z. A. Ahmad, "Influence

of silica and zirconia particles as filler loading on the radiopacity of dental resin composites", *Advanced Materials Research*, vol. 620, pp. 345–349, 2013.
[25] International Standard of Organization (ISO) 4049 - Dentistry - Polymer-based filling, restorative and luting materials, 2000.
[26] L. Wang, P. D'Alpino, L. Lopes, and J. Pereira, "Mechanical properties of dental restorative materials: relative contribution of laboratory test," *Journal of Applied Oral Science*, vol. 11, no. 3, pp. 162–167, 2003.
[27] M. Albakry, "Insightful understanding of the role of the mechanical properties in defining the reliability of all-ceramic dental restorations : A review," *Journal of Biomaterials and Nanobiotechnology*, vol. 12, pp. 57–78, 2021.
[28] I. M. Barszczewska-Rybarek, "Structure-property relationships in dimethacrylate networks based on Bis-GMA, UDMA and TEGDMA", *Dental Materials*, vol. 25, pp. 1082–1089, 2009.
[29] M. T. Lemon, M. S. Jones, and J. W. Stansbury, "Hydrogen bonding interactions in methacrylate monomers and polymers," *Journal of Biomedical Materials Research Part A*, vol. 83, no. 3, pp. 734–746, 2007.
[30] A. Szczesio-Wlodarczyk, M. Domarecka, K. Kopacz, J. Sokolowski, and K. Bociong, "An evaluation of the properties of urethane dimethacrylate-based dental resins", *Materials*, vol. 14, no. 2727, pp. 1–17, 2021.
[31] I. M. Barszczewska-Rybarek, "A guide through the dental dimethacrylate polymer network structural characterization and interpretation of physico-mechanical properties", *Materials*, vol. 12, no. 4057, pp. 1–30, 2019.
[32] F. Goncalves, Y. Kawano, and R. R. Braga, "Influence of BisGMA, TEGDMA, and BisEMA contents on viscosity, conversion, and flexural strength of experimental resins and composites," *European Journal of Oral Sciences*, vol. 117, pp. 442–446, 2009.
[33] I. Sideridou, V. Tserki, and G. Papanastasiou, "Study of water sorption, solubility and modulus of elasticity of light-cured dimethacrylate-based dental resins", *Biomaterials*, vol. 24, no. 4, pp. 655–665, 2003.
[34] E. Habib, R. Wang, and X. X. Zhu, "Correlation of resin viscosity and monomer conversion to filler particle size in dental composites", *Dental Materials*, vol. 34, no. 10, pp. 1501–1508, 2018.
[35] H. A. Rodríguez, W. M. Kriven, and H. Casanova, "Development of mechanical properties in dental resin composite : Effect of filler size and filler aggregation state", *Materials Science & Engineering C*, vol. 101, pp. 274–282, 2019.
[36] W. Huang, L. Ren, Y. Cheng, M. Xu, W. Luo, D. Zhan, and H. Sano, "Evaluation of the color stability, water sorption, and solubility of current resin composites", *Materials*, vol. 15, no. 6710, pp. 1–14, 2022.
[37] 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*, vol. 28, no. 4, pp. 349–359, 2012.

[38] N. A. F. Azlisham, Y. Johari, D. Mohamad, M. F. Yhaya, and Z. Mahmood, "Degree of conversion and physicomechanical properties of newly developed flowable composite derived from rice husk using urethane dimethacrylate monomer," *Journal of Engineering in Medicine*, vol. 00, no. 0, pp. 1–9, 2023.

[39] N. Sumino, K. Tsubota, T. Takamizawa, K. Shiratsuchi, M. Miyazaki, and M. A. Latta, "Comparison of the wear and flexural characteristics of flowable resin composites for posterior lesions", *Acta Odontologica Scandinavica*, no. 71, pp. 820–827, 2013.

[40] A. Tsujimoto, M. Irie, E. C. N. Teixeira, C. A. Jurado, Y. Maruo, G. Nishigawa, T. Matsumoto, and F. Garcia-Godoy, "Relationships between flexural and bonding properties, marginal adaptation, and polymerization shrinkage in flowable composite restorations for dental application", *Polymers*, vol. 13, no. 2613, pp. 1–13, 2021.