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Original article

Properties of nanocellulose and zirconia alumina on polymethylmethacrylate dental composite

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ABSTRACT

Background: *Polymethylmethacrylate (PMMA) is one of the synthetic polymers generally used for temporary jacket crown restorations because of its good translucency, making its aesthetic value higher, but its mechanical properties, such as hardness and flexural strength are lower than composite resins. Hence, adding zirconia and cellulose filler is necessary to enhance its mechanical properties.* ***Purpose:*** *This is an experimental laboratory study to make nanocomposites with PMMA as a matrix with crystalline nanocellulose, zirconia, and alumina added as fillers.* ***Methods:*** *The crystalline nanocellulose filler was synthesized by acid hydrolysis. Zirconia and alumina were synthesized using the sol-gel technique and then characterized by transmission electron microscope and X-ray diffraction. The Micro Vickers hardness test and three-point bending tested mechanical properties. The analysis was carried out with a one-way analysis of variance, followed by a post hoc Tuckey's test with a* $P < 0.05$ *taken as statistically significant.* ***Results:*** *The Micro Vickers hardness test showed the highest hardness in the group with a ratio of PMMA and zirconia-alumina filler of 50%: 2%: 48% (12.73 VHN). The results of the three-point bending test showed that the highest flexural strength was found in the control group (19.4 MPa).* ***Conclusion:*** *The addition of crystalline nanocellulose, zirconia, and alumina increase the hardness of the nanocomposite, while the flexural strength was lower than PMMA without filler addition.*

Keywords: *alumina; crystalline nanocellulose; mechanical properties; PMMA; zirconia*
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INTRODUCTION

The development of science and technology in dentistry improves the quality of life in patients with teeth and mouth problems. Damage to the calcified tooth structure and supporting tissues by noxious stimuli can cause pulp and peri-radicular tissue changes. Noxious stimuli can be physical, chemical, or bacterial that can produce reversible or irreversible changes, depending on the duration, intensity, and pathogenesis, and the ability of the host to resist them and repair tissue damage.[1] Caries is a microbiological infectious disease of the teeth that causes local changes and destruction of the hard tissues of the teeth. Tooth structure loss can be repaired with restoration procedures. The materials most commonly used to restore dental caries are metals, ceramics, polymers, and composites. The use of metal to restore caries has been abandoned because of aesthetic and biocompatibility issues. Therefore, many have started to switch to composite restorative materials. Nanocomposite has been widely developed for restorative materials today.[1,2]

Composites are physical mixtures of metals, ceramics, and/or polymers to obtain the desired mechanical properties of each mixed material. The mixture commonly found in dental composites is a mixture of ceramic and polymer matrix. Today, polymethylmethacrylate (PMMA) and bisphenol A-glycidyl methacrylate are the polymers widely used as composite materials. Mixing these materials is intended to obtain the desired mechanical properties of each material.[1,3] Composites are currently limited to restoring class III, IV, V, and class I cavities if aesthetics are required. Restoration materials should have good

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hardness and flexural strength like enamel and ideally have good antibacterial properties to prevent secondary caries.[4–6]

Composite materials currently being developed are often used as restorations in the anterior and posterior teeth. Current products in the market have low flexural strength and hardness, which is not good enough for patients with parafunctional habits such as bruxism. The lack of strength to resist composite fractures in high-pressure areas, such as in the case of extensive preparations, including the cusp, requires reinforcing material since the composites must withstand cavity restorations under high stresses.[7] Hardness and flexural strength can be reduced due to consuming various types of food, especially acidic foods, and frequent mouthwash usage.[8]

The raw materials for manufacturing composite fillers are generally imported from other countries, even though Indonesia's abundant natural resources provide good quality raw materials for manufacturing dental materials. Zirconia can be used as filler material for PMMA matrix because it is chemically stable, increases fracture toughness, has good dimensional stability, a modulus of elasticity and flexural strength similar to steel, and good biocompatibility. One of the main reasons zirconia is used as a raw material for dental materials, and especially restoration materials, is because its tooth-like color increases its aesthetic value. In addition to zirconia, alumina is often used as a composite filler because it has good wear resistance, optimal hardness, good thermal conductivity, and sufficient rigidity.[8,9]

Currently, nanocellulose is a new material being developed as a raw material for restoration materials in dentistry. Nanocellulose is a natural material with unique characteristics and is synthesized from cellulose (obtained from plants, animals, and bacteria). This material has received much attention for its use as a biomedical material because of its mechanical, chemical, and biological properties (biocompatibility, biodegradability, and low toxicity).

Nanocellulose can be developed as a raw material for biomedical materials, especially for filler composites, because of its properties. The nanocellulose is extracted from palm kernel cake by converting large units (cm) to small units (nm) using chemical aids such as acid hydrolysis, which is commonly used to remove the amorphous part and extract the crystalline form of pure cellulose, which is essential because it can fill the void in composites fused with alumina-toughened zirconia (ZrO_2-Al_2O_3).[10]

Based on the facts stated above, this study aims to develop a nanocomposite based on PMMA, crystalline nanocellulose, and ZrO_2-Al_2O_3 filler as a dental restoration material and analyze its morphological and mechanical properties. Since the resulting products must comply with the American National Standards Institute and the American Dental Association (ADA) standards, the flexural strength and hardness of the material must be analyzed.

MATERIALS AND METHODS

Palm kernel cake was used as a precursor for the synthesis of crystalline nanocellulose mixed with demineralized water (DM), nitric acid 3.5%, sodium nitrite, sodium hydroxide, sodium sulfite, sodium hypochlorite, and sulfuric acid (H_2SO_4) 45%. All chemicals were from the brand Sigma Aldrich^{TM}.

Zirconium dioxide (ZrO_2) powder was synthesized by mixing zirconium chloride ($ZrCl_4$), calcium chloride hydrate as a stabilizer, ethanol 90%, and DM. The $ZrCl_4$ precursor (4.66 g) was mixed with 200 ml of DM and then stirred using a magnetic stirrer for 15 minutes to obtain a 0.1m $ZrCl_4$ solution. Calcium chloride hydrate stabilizer 0.234 g was added to the precursor solution, mixed with a magnetic stirrer for 15 minutes until homogeneous, and added to approximately 8% of the total molarity of the precursor. The sample underwent an aging and drying process in an oven with a temperature of 120°C for 24 hours until the solvent evaporated and obtained xerogel results. After grinding, the sample was transferred to a combustion boat, then calcined in a furnace starting from room temperature to 900°C and maintained at that temperature for two hours to form metal oxide particles. The temperature was then lowered. The particles were ground with a mortar and pestle until smooth to get smaller particles. The calcined sample was dissolved in 50 ml of ethanol and then homogenized with an ultrasonic homogenizer with an amplitude of 80 for 30 minutes to produce nanometer-sized particles. The samples were dried in an oven for 24 hours to obtain ZrO_2 in the form of calcium partially stabilized zirconia (Ca-PSZ) particles.

Aluminum nitrate hydrate ($Al(NO_3)_3.9H_2O$) as a precursor (11.25 g) was dissolved in 50 ml of DM, and 0.1M ammonium carbonate solution was added as a pH controller to obtain a pH of 8–9 while stirring, using a magnetic stirrer for 45 minutes. Then aging was done using an ultrasonic bath for three hours without heat until two layers were formed. The top layer was clear, while the bottom layer was a gel precipitate. The precipitate was then filtered using a Buchner funnel while rinsing with aqua DM until a neutral pH was attained and filtered using filter paper. The filtered residue was then dried in an oven at 100°C for one day. After the sample was dry and formed a white solid, the next step was calcination at 550°C for one hour. The calcined results were then ground into powder using a mortar and pestle to obtain aluminum oxide (Al_2O_3) powder.

The nanocomposite was synthesized by mixing PMMA matrix, cellulose nanocrystalline, and ZrO_2-Al_2O_3 powder with three different compositions: (I) PMMA without filler, (II) PMMA 50% with cellulose nanocrystalline 1% and ZrO_2-Al_2O_3 powder 49%, and (III) PMMA 50% with cellulose nanocrystalline 2% and ZrO_2-Al_2O_3 powder 48%. Specimens were made according to ADA specification no. 27.[11] The Micro Vickers hardness test specimen was of Ø 6 mm x 3 mm thickness. It was tested with LECO-Japan M-400-H1 exposed to 100g force for 15 seconds in

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the Physical Metallurgy Laboratory, Institut Teknologi Bandung. The specimen for the three-point bending was a beam of 2 mm x 2 mm x 25 mm, exposed to 1 kN force and a crosshead speed of 1mm/min using the Shimadzu Autograph AGS-5kNx in the Faculty of Dentistry, Maranatha Christian University. Materials for making specimens were PMMA resin without filler and its monomer, ZrO_2-Al_2O_3 filler, and crystalline nanocellulose.

Transmission electron microscopy (TEM) was conducted using a Hitachi HT7700 (25000X magnification). X-ray diffraction (XRD) with 0–90° angle was performed using a Bruker-D8 Advance. Both characterizations were conducted at the Center of Advanced Science, Institut Teknologi Bandung.

RESULTS

The average hardness values in test group I were 9.61 ± 1.64 VHN in test group I, 11.34 ± 0.81 VHN in test group II, and 12.73 ± 0.6 VHN in test group III. The data for the Micro Vickers hardness were tested for normality with the Shapiro–Wilk test. The *P* value of < 0.05 confirmed that the data distribution was not normal. The data for the Micro Vickers hardness test was then tested for homogeneity. It showed a *P* value of 0.756 (> 0.05), confirming that the data was homogeneous. Since the data distribution was not normal, the Kruskal-Wallis non-parametric test was used for statistical analysis. The results show that the *P* value (Asymp. Sig.) was 0.004 (< 0.05), confirming that there was a significant difference in Micro Vickers hardness between the test groups. The Mann–Whitney non-parametric test was performed between two groups to check which differences between groups were significant. The test between groups I and II resulted in a *P* value (Asymp. Sig. 2-tailed) of 0.016 (< 0.05), confirming that there was a significant difference in the hardness between these groups. The test between groups II and III resulted in a *P* value (Asymp. Sig. 2-tailed) of 0.028 (< 0.05), establishing that there was a significant difference in hardness between groups II and III. The test between groups I and III produced a *P* value (Asymp. Sig. 2-tailed) of 0.009, confirming a significant difference in hardness between groups I and III. Table 1 shows the Micro Vickers hardness test results.

The three-point bending test was carried out on 15 specimens which were divided into three groups, namely group I (PMMA without the addition of filler), group II (PMMA with the addition of crystalline nanocellulose, zirconia and alumina fillers in a ratio of 50:1:49), group III (PMMA with the addition of crystalline nanocellulose, zirconia and alumina fillers in a ratio of 50:2:48) Table 2 shows the results of the three-point bending test.

Table 1. Micro Vickers hardness test results (VHN)

Sample	Group		
	I	II	III
1	10.03	12.3	13.06
2	10.26	11.8	13.6
3	10.03	11.8	12.1
4	9.73	10.76	12.9
5	8.03	10.06	12.0
Average	9.61	11.34	12.73

Table 2. Three-point bending test result (MPa)

Sample	Group		
	I	II	III
1	18.17	8.56	10.48
2	20.94	8.55	10.20
3	18.64	7.24	10.60
4	20.30	9.67	9.33
5	18.95	6.77	8.36
Average	19.4	8.15	9.79

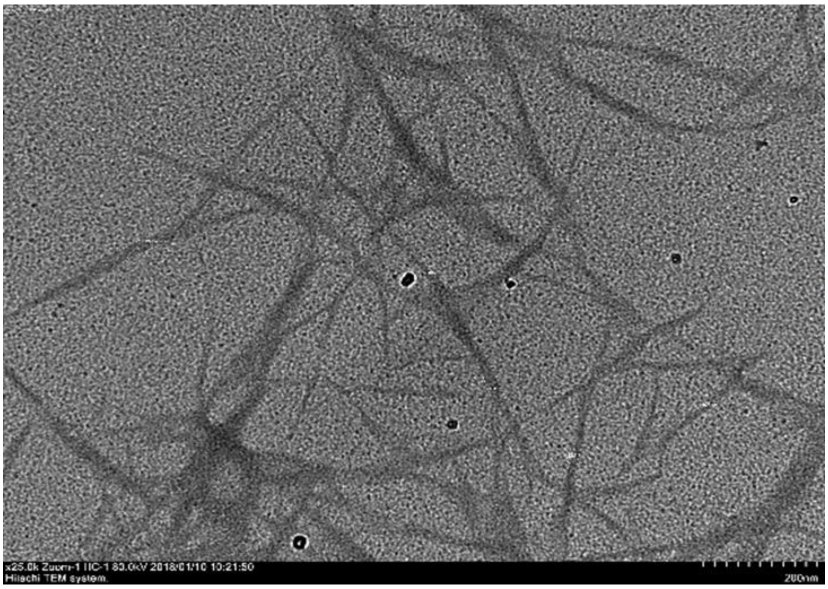

Figure 1. Results of TEM characterization of crystalline nanocellulose showing fiber-shaped particles (25000X magnification).

The data contained in Table 1 were tested for normality with the Shapiro–Wilk test. The *P* value (sig.) was > 0.05 in all groups, confirming that the data distribution was normal. The homogeneity test showed a *P* value (sig.) of 0.742 (> 0.05), establishing that the data was homogeneous. Since the data were normal and homogeneous, a one-way analysis of variance was used for statistical analysis. The result of the *P* value (sig.) was 0.000 ($P < 0.05$), indicating there was a significant difference in the flexural strength values between the test groups, and the null hypothesis was successfully rejected. Tukey's post hoc test then processed the data to find which groups had a significant difference in value. Tukey's post hoc test results showed that group I (nanocellulose content of 0.00%) significantly differed in flexural strength values compared with groups II and III.

Crystalline nanocellulose was characterized using TEM to measure its particle size, while zirconia and alumina were characterized using XRD to analyze their crystal structure. The results of the characterization of crystalline nanocellulose in the form of a gel that had previously been prepared using isopropyl ethanol using TEM can be seen in Figure 1. The results of XRD characterization of zirconia and alumina show peaks describing the diffractogram's crystallinity in Figures 2 and 3, respectively.

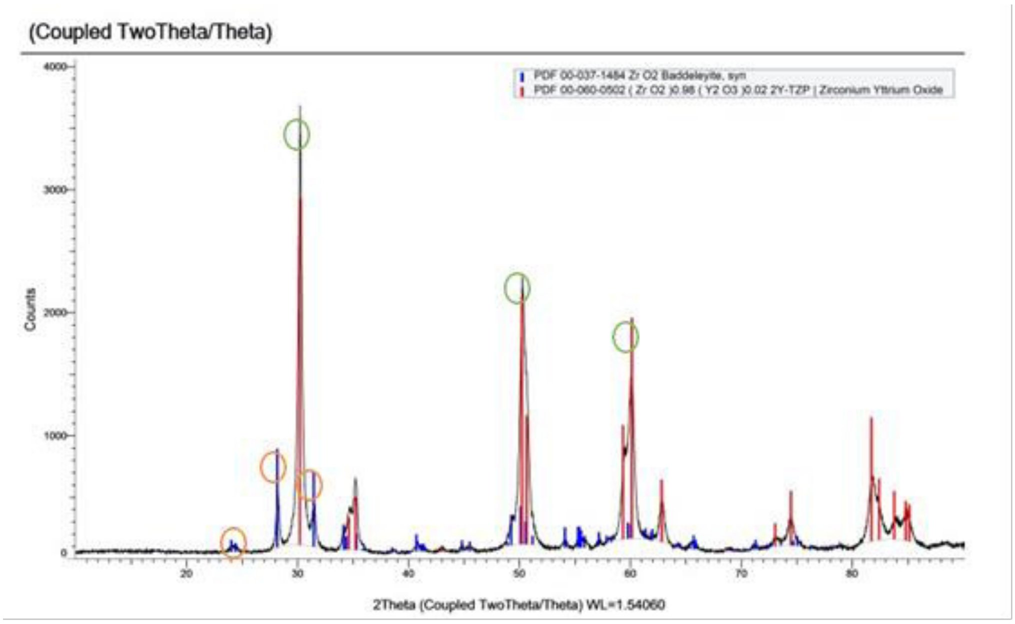

Figure 2. XRD Characterization results of zirconia powder with a calcination temperature of 900°C.

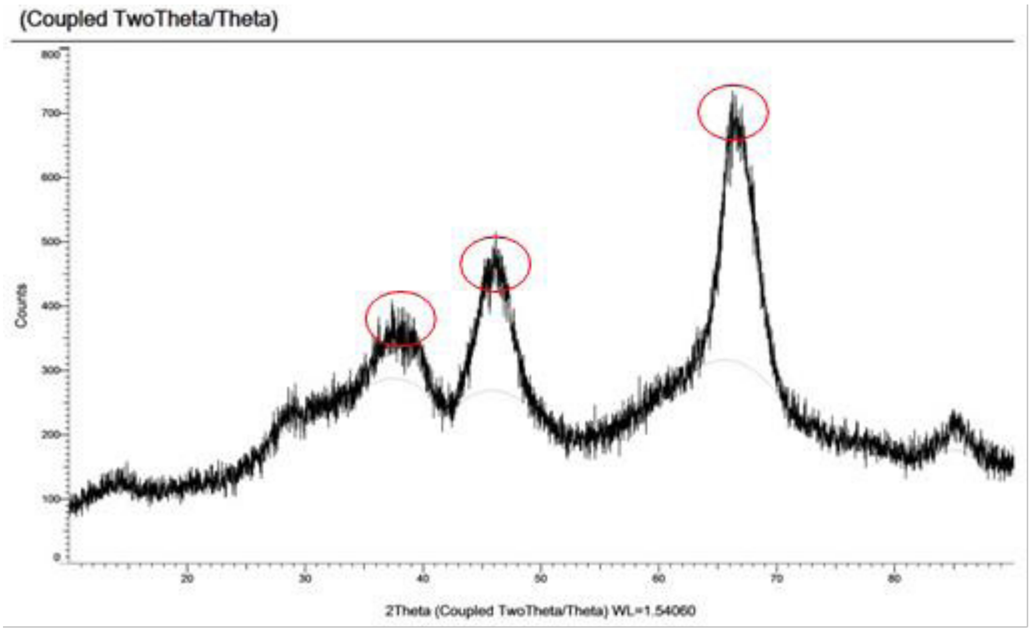

Figure 3. XRD characterization results of alumina powder with a calcination temperature of 550°C.

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DISCUSSION

Polymethylmethacrylate is often used as a resin matrix in dentistry because it has sufficient strength, good translucency, malleability, and resistance to microbial colonization. It is odorless, rigid, tasteless, and non-irritating to tissues. However, it is brittle compared to other composite resins. However, this deficiency can be overcome by adding filler.[1,12] The fillers used in this study were zirconia, alumina, and crystalline nanocellulose. Zirconia was synthesized with $ZrCl_4$ as a precursor and alumina with $Al(NO_3)_3.9H_2O$ as a precursor. Zirconia and alumina were obtained using the sol-gel technique. Crystalline nanocellulose in this study was synthesized using the acid hydrolysis method using H_2SO_4. The temperature was lower, and the processing time was faster to dissolve lignin and hemicellulose in the precursor to obtain crystalline nanocellulose. In this study, palm kernel cake was used. as a precursor. Palm kernel cake is often used as a source of crystalline nanocellulose because it has high levels of hemicellulose, cellulose, and lignin, making it suitable cellulose-forming biomass.

Palm kernel cake has a cellulose content of 30%, so it is produced quite a lot in the palm oil industry as a residual extraction material. Due to its abundant availability, palm kernel cake was used as animal feed before being widely used as cellulose-forming biomass. Palm kernel cake is environmentally friendly because it utilizes waste from palm oil extraction.[13]

The synthesized filler was characterized to see the particle size and morphology. The results of the TEM characterization of crystalline nanocellulose showed that the crystalline nanocellulose produced using the acid hydrolysis method had particle diameter sizes ranging from 6.80 nm to 25 nm and were in the form of fibers. Nanocellulose in fiber form generally has a diameter of 2–20 nm and a length of 100–600 nm. The acid hydrolysis process at 45°C typically produces crystalline nanocellulose in spherical form, but it does not rule out the possibility of forming nanocellulose as fiber.

The shape of this fiber depends on the transfer of stress. If the fiber is long, the modulus of elasticity will increase, so the stress transfer will be better.[14,15] The distribution of nanocellulose seen in the TEM in this study was not homogeneous and experienced agglomeration. Agglomeration may be caused by a preparation error during characterization when mixing isopropyl ethanol.[16]

The results of XRD characterization of the alumina filler contained one crystallite phase, namely, alumina with a metastable phase, identified from alumina powder calcined at a temperature of 550°C. A metastable condition is one in which a material has critical stability. External influences, such as humidity or specific temperature changes, can disrupt the stability causing the material to fall to a lower energy level. When a peak is narrower and sharper, the degree of crystallinity of the material is higher. A higher degree of crystallinity increases the mechanical properties of the material. The calcination temperature affects The crystallite structure and phases formed in a material. In this study, zirconia was calcined at 900°C for two hours so that two crystallite phases were identified, namely tetragonal and monoclinic zirconia, where the tetragonal phase was more dominant than the monoclinic. The tetragonal structure has advantages over the monoclinic and cubic structures because it has a relatively high resistance to cracking.[17–19]

Metastable alumina is the purest form of alumina and has high porosity and surface area. Due to its acidic and basic properties, it is often used as a catalyst, absorbent material, catalyst support, filler, or polymer composite component with reasonably good mechanical properties.[20]

Based on their composition, there were three groups of nanocomposite specimens: I, II, and III. A comparison of the filler composition, ranging from 50–85% of the matrix, was selected.[21] In this study, the ratio of the matrix and filler was 50%:50%. This ratio was chosen because it is easier to mix, even though theoretically, the more the filler, the better the mechanical strength. Adding more filler than the matrix makes mixing difficult, mainly if manual mixing and agglomeration are performed. The statistical analysis between test groups I and II showed that the P value was 0.016 (< 0.05), indicating a significant difference in hardness between groups I and II. Similarly, the results of the statistical analysis conducted between groups II and III resulted in a P value of 0.028 (< 0.05), indicating a significant difference in hardness between groups II and III.

Thus, adding zirconia, alumina, and crystalline nanocellulose fillers can increase the hardness of PMMA because crystalline nanocellulose acts as a binding material between PMMA and zirconia and alumina fillers. Crystalline nanocellulose has a tensile strength of 7.5–7.7 GPa (greater than steel), and the modulus of elasticity is 150 GPa. Nanocellulose is often used as a reinforcing agent because it has good mechanical strength and fibers that can transfer stress. The PMMA hardness increases with the amount of crystalline nanocellulose added as filler compared with the test group without filler.[22,23] Table 1 shows the three-point bending test results. The average results of the test were 19.4 MPa in group I, 8.15 MPa in group II, and 9.79 MPa in group III. The average value was the highest in group I compared with the other groups that were given filler. This is because adding zirconia, alumina, and crystalline nanocellulose fillers in test groups II and III makes the compound more brittle, lowering its flexural strength as compared with group I not given a filler. This is due to the poor mixing process, whereby crystalline nanocellulose is not bonded and mixed homogeneously. In test groups II and III, there was an increase in flexural strength because cellulose is a natural polymer that is fibrous, strong, has good mechanical properties, and is often used as a reinforcing agent.

Cellulose has a tensile strength of 7.5—7.7 GPa, greater than steel, and a modulus of elasticity of 150 GPa,

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demonstrating that mechanical strength will increase the more crystalline nanocellulose is added to a material.[24] The hardness value of PMMA is around 20 VHN, and the flexural strength is 70 MPa.[25] The hardness value of enamel is 300 VHN, and the flexural strength is 85 MPa, whereas the hardness value of dentin is 60 VHN, and the flexural strength is 15 MPa.

The average hardness value was only 12.73 VHN in test group III, and the mean flexural strength was 9.79 MPa, lower than expected. This could be due to the poor polymerization process because of the presence of impurities in the specimen. Moreover, the heating was done over a stove and hot water. Mixing the polymer, matrix, and monomer manually resulted in a less homogeneous and more porous compound due to air trapping. Further, a coupling agent was not used.

In this study, nanocomposite has been successfully synthesized from crystalline nanocellulose, ZrO_2, and Al_2O_3. TEM results of crystalline nanocellulose showed that the fiber generally has a diameter of 2–20 nm and a length of 100–600 nm. XRD results show that ZrO_2 has tetragonal and monoclinic forms. These phases have good resistance to cracking. Alumina has a metastable phase, showing the material has critical stability. The addition of crystalline nanocellulose, zirconia, and alumina reduces the flexural strength but increases the hardness of the PMMA matrix nanocomposite. The best hardness was achieved in Group III (12.73 ± 0.6 VHN), which shows an improvement compared with PMMA without filler addition (9.61 ± 1.64 VHN).

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