

Research Article

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Effect of alkaline treatment time of fibers and microcrystalline cellulose addition on mechanical properties of unsaturated polyester composites reinforced by cantala fibers

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Abstract: The aim of this study is to investigate the effect of alkaline treatment and addition of microcrystalline cellulose (MCC) on the density, tensile strength, elastic modulus, impact strength, and Poisson's ratio of unsaturated polyester resin composites reinforced by cantala fibers (UPR-CFs). The fibers were immersed in alkaline solution of 6% NaOH for 0, 3, 6, 9, and 12 h. The composite had cantala fibers (CFs) and MCC volume fraction (v_f) of 30 and 5%, respectively. The results showed that the alkaline treatment for 6 h resulted in the highest density, tensile strength, and elastic modulus of the composites, while the highest Poisson's ratio was achieved in UPR-untreated CF composites. The addition of MCC filler also increased the density, tensile strength, and elastic modulus of the composite significantly. Both alkaline treatment and MCC addition did not significantly affect the impact strength. With the increase in the strength and modulus of elasticity, composites can be applied more widely to structures that bear higher loads.

Keywords: unsaturated polyester, cantala fibers, alkaline treatment, microcrystalline cellulose, Poisson's ratio

1 Introduction

The rapid development of science and technology has also increased the need for new materials, one of which is composite material. Composite is a material that consists of at least two components, namely, matrix and reinforcement, which has advantages such as light weight, good mechanical properties, durability, heat resistance, corrosion resistance, easy fabrication, and relatively low production costs [1]. Now composites are widely used in the industrial world, for example in the car, aircraft, and construction industries [2]. Increasing problems of environmental pollution, waste and the greenhouse effect have encouraged the industry to use environmentally friendly and renewable composite materials in producing products [3], namely, biocomposite. It is a composite material in which at least one component comes from nature, usually consisting of a polymer matrix with natural fibers as reinforcement and/or filler, such as flax, hemp, agave, kenaf, jute, and sisal [4]. There was also utilization of coconut fibers in ceramic matrix composite, such as concrete roads due to their higher toughness compared to other natural fibers and cheaper cost. The application of natural fibers reduced the thickness of concrete roads and lowered the use of more expensive materials [5]. The previous study also indicated that the addition of 2% coconut fibers enhanced the overall properties of concrete having silica-fume and super plasticizer content of 15 and 1%, respectively [6]. The addition of 50 mm length coconut fibers of 2% into the concrete containing 10% fly ash and 15% silica-fume showed higher overall properties than those without coconut fibers [7].

Natural fibers are fibers produced from plants, animals, or minerals [8], which have advantages over synthetic fibers, such as being abundant in nature, lightweight, non-corrosive, low density, good thermal properties, inexpensive, environmentally friendly, and renewable [9]. The most abundant

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natural fibers on earth are plant or vegetable fibers [10]. One of the plant fibers that can be implemented in biocomposites is agave fiber [11], where the agave plant species that is easily found in Indonesia is an *Agave cantala*. An *Agave cantala* plant has some characteristics. The color of its leaves is bluish-gray and their edges are slightly thorny, which through the extraction process will produce cantala fibers (CFs) that are strong and hard. Based on research results from the Industrial Research and Development Agency of the Yogyakarta Department of Industry, a CF has a cellulose content of around 64.23%, making it a potential reinforcing component in composite materials.

Some polymer matrices often used in natural fiber reinforced composites can be classified as thermoplastic and thermoset polymers [12]. Most thermoplastic polymers have non-polar properties, such as PE, PP, and PVC, while thermosets have polar properties, such as unsaturated polyester resins (UPR), epoxy, and phenol [13]. UPR is a synthetic resin with straight chains resulting from the reaction of glycol with difunctional acids, such as maleic acid and adipic acid. This resin has high mechanical properties, heat and chemical resistance, low price, and easy fabrication, and has been widely applied in the automotive and construction fields [14].

In natural fibers, the compound that most affects the mechanical properties of the composite is cellulose [15]. Cellulose is a sustainable, abundant, and naturally occurring polymer derived from biomass [16]. One of the problems in composite fabrication is the uneven distribution of natural fiber cellulose. Therefore, the cellulose particles of natural fiber must be reduced in the form of microcrystalline cellulose (MCC) [17]. MCC is pure cellulose isolated from alpha cellulose using mineral acids with the advantages of being renewable, inexpensive, non-toxic, good in mechanical properties, and biocompatible [18]. The addition of MCC increased the tensile and flexural strength and elastic modulus of UPR matrixed composites [19]. The most optimum addition of MCC was obtained by 5%. Therefore, MCC is often added to UPR as matrix for plant fiber composites.

The mechanical properties of a fiber-reinforced polymer composite are greatly influenced by the adhesion bond between their fibers and matrix. Natural fibers have hydrophilic properties that prevent adhesion bonds with hydrophobic polymer matrices. These properties can be improved by chemical treatment of the fiber surface [20], one of which is alkali [21,22]. Alkaline treatment uses a solution of distilled water and NaOH, with a variable concentration and immersion time, which serves to remove hemicellulose and lignin on the surface of natural fibers [22]. *Agave americana* fibers treated by alkali indicated significant reduction in hemicellulose, lignin, and other impurities [23].

One of the important variables in the alkaline treatment (NaOH) is immersion time because it affects the fiber surface in order to obtain the maximum mechanical properties of the fiber [24]. Asim *et al.* [25] have investigated the effect of alkaline immersion time on the mechanical properties of pineapple fiber with two variables, namely, 3 and 6% NaOH concentrations and immersion times of 3, 6, 9, and 12 h. The results showed that the highest mechanical properties of pineapple fiber were obtained within 6 h because it can remove hemicellulose and lignin effectively. In addition, the alkaline treatment also increases the surface area and roughness of the pineapple fiber so that it can create a strong adhesive bond with the thermoset or thermoplastic matrix.

The characteristics of the composite are also greatly influenced by the method of composite manufacture. In the last decade, the conventional method often used in industry is the hand lay-up and spray-up due to its easiness and simplicity, but there is one drawback, namely, that there is still a lot of trapped air [26]. One new method that eliminates air ingress is compression molding, using pressure on the top and bottom of the die. The advantages of this method are the small amount of residual material, high precision, environmental friendliness, and reduced air bubbles [27].

CF has the potential to be used as a reinforcement for UPR + MCC matrix composites. However, the effect of alkali treatment time on CF and MCC addition on physical and mechanical properties of unsaturated polyester resin composites reinforced by cantala fibers (UPR-CFs) composite fabricated using a compression molding process has not been investigated yet. This study investigates the properties of materials, including density, tensile strength, elastic modulus, impact strength, and the value of the composite Poisson's ratio. The fracture morphology of the specimen will be observed using scanning electron microscopy (SEM). By understanding the effect of the duration of the alkali treatment and the addition of MCC to the UPR-CFs composite, the utilization of these composites can be expanded to applications capable of withstanding greater loads, such as furniture, partition panels, building components, and vehicle body parts.

2 Materials and methods

2.1 Materials

CFs were obtained from *Agave cantala* plants, as shown in Figure 1(a), in Pengasih District, Kulon Progo Regency,



Figure 1: (a) Agave cantala plant and (b) CFs.

Special Province of Yogyakarta, Indonesia. Unsaturated polyester YUKALAC 157-BQTN EX of Ortho-Phthalic type was purchased from PT. Justus Kimiaraya, Semarang, Indonesia, which was used as the matrix. The MEKPO brand catalyst, being a mixture of methyl ethyl ketone peroxide diplasticizer of the phthalate type and having function to speed up the curing process was also obtained from PT. Justus Kimiaraya, Semarang. Aldrich-branded MCC in the form of white powder with a size of $20\ \mu\text{m}$, and a density of $1.56\ \text{g}\cdot\text{cm}^{-3}$, playing a role as an additional reinforcement in polymer composites, was purchased from Sigma-Aldrich PTE. Ltd, Singapore.

Distilled water functioned as a solvent for NaOH crystals in the alkaline fiber treatment. NaOH crystals with a content of 99% were purchased from MERCK, Jakarta, Indonesia. Acetic acid, also purchased from MERCK, served to remove alkaline properties and neutralize the pH of CF after alkaline immersion. Wax mirror glaze, purchased from PT. Indus Chemika Suplindo, Tangerang, Indonesia, was used as release agent.

2.2 Method

2.2.1 Fiber preparation

The cantala plant was cut by choosing good quality leaves, then the cantala leaf extraction process was carried out to get the fiber, then a drying process is carried out for 1–2 days until the CF dried, as indicated in Figure 1(b). The process of alkaline treatment used NaOH crystals mixed in distilled water, then the fibers were soaked for various times of 3, 6, 9 and 12 h with the composition of CF, distilled water, and NaOH crystals being 500 g, 2.35 L, and 150 g, respectively.

The CF was taken, rinsed, and then soaked in acetic acid with a concentration of 1%, then rinsed with water repeatedly until the pH of the water became neutral. The fibers were allowed to stand at room temperature for 24 h, then heated in an oven at 60°C for 10 h. If it was dry, the next process was to cut the fiber to a size of $\pm 10\ \text{mm}$ and put it in a container.

2.2.2 Composite manufacturing process

CFs with 30% volume fraction were put into a mold with a size of $170\ \text{mm} \times 110\ \text{mm} \times 10\ \text{mm}$, which had previously been coated with wax mirror glaze. Then, the matrix consisting of UPR resin, catalyst, and MCC with a volume fraction of 1 and 5%, respectively, was put into the mold until the mixture was evenly distributed. After that pressure was applied at $10\ \text{N}/\text{mm}^2$ for 12 h at room temperature. The composite was then put into the oven for curing for 2 h at 60°C . Scheme of the fiber preparation and composite manufacturing process are shown in Figure 2. The composites produced, as indicated in Figure 3, were then named as shown in Table 1.

2.2.3 Density test

Calculation of the actual density of composite materials with ASTM D792 standard is given in Eq. (1) [28].

$$\rho = \frac{m_a}{m_w + m_a} \times \rho_w, \quad (1)$$

where ρ , ρ_w , m_a , and m_w represent the density of the composite ($\text{kg}\cdot\text{m}^{-3}$), the density of water at room temperature ($\text{kg}\cdot\text{m}^{-3}$), the mass of the composite in air (kg), and the mass of the composite in water (kg), respectively. Based on the

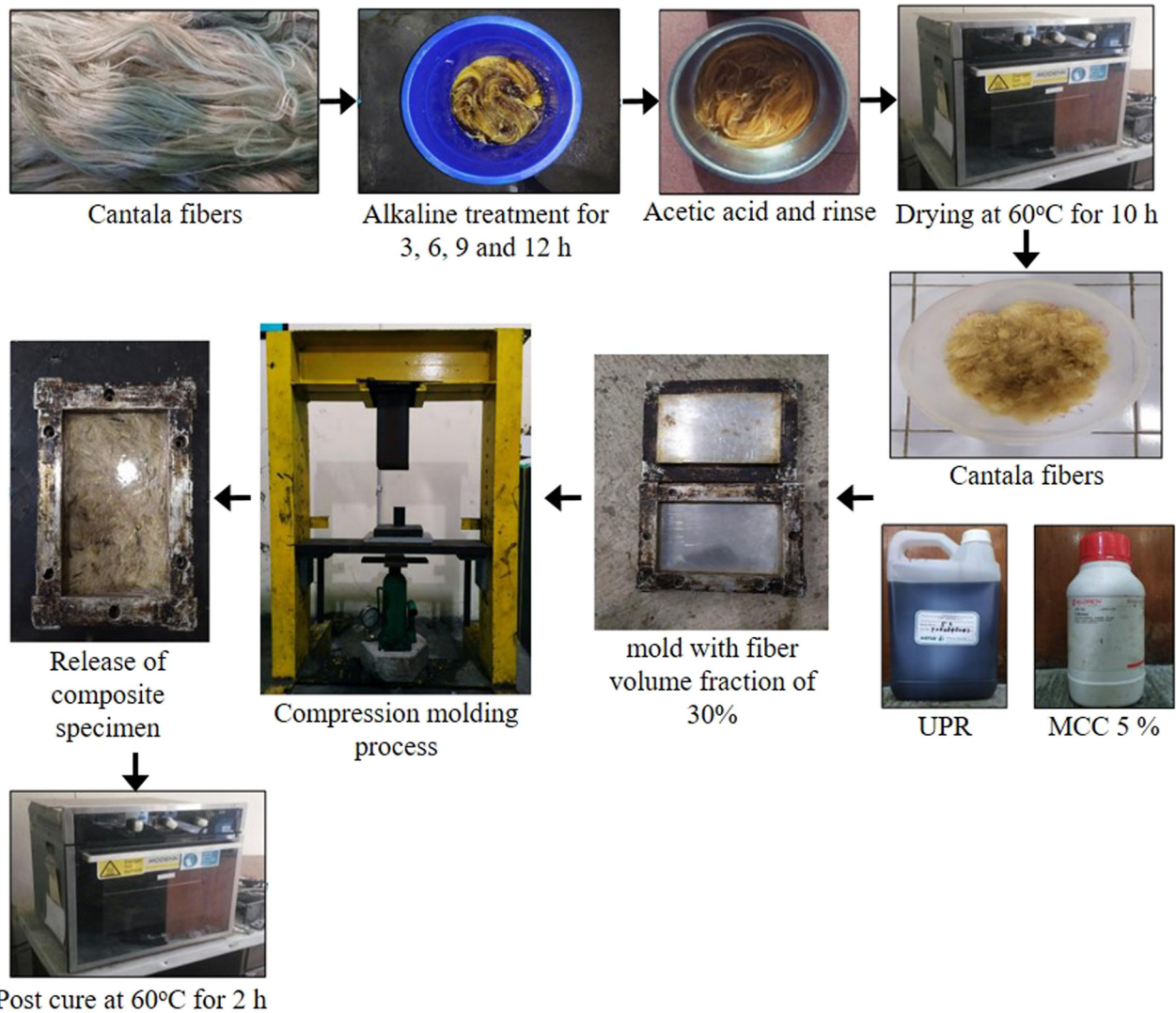


Figure 2: Fiber preparation and composite manufacturing process.

rule of mixture, the theoretical value of the density of polymer composites can be calculated by Eq. (2) [29].

$$\rho_c = v_f \cdot \rho_f + (1 - v_f) \cdot \rho_m, \quad (2)$$

where ρ_c , ρ_f , and ρ_m represent the density of the composite, fiber, and matrix ($\text{kg} \cdot \text{m}^{-3}$), respectively, and v_f represents the volume fraction of the fiber.

2.2.4 Tensile test

This test was carried out to determine the tensile strength and elastic modulus of the composite, carried out based on ASTM D638 standard using the JTM-UTS510 universal testing machine, made in People's Republic of China (Figure 4(a)), with an accuracy of 0.5%, crosshead speed of $50 \text{ mm} \cdot \text{min}^{-1}$,



Figure 3: Variation in composite specimens.

Table 1: Nomenclature of composites

Symbol of composites	Annotation
UPR	UPR matrixed composites without CF
UPR + MCC	UPR and MCC ($v_f = 5\%$) composites without CFs
AL0.MCC	UPR and MCC ($v_f = 5\%$) composites with untreated CFs
AL3.MCC	UPR and MCC ($v_f = 5\%$) composites with alkaline-treated CFs for 3 h
AL6.MCC	UPR and MCC ($v_f = 5\%$) composites with alkaline-treated CFs for 6 h
AL9.MCC	UPR and MCC ($v_f = 5\%$) composites with alkaline-treated CFs for 9 h
AL12.MCC	UPR and MCC ($v_f = 5\%$) composites with alkaline-treated CFs for 12 h

and load-cell capacity of 500 kg [30]. The specimen dimension is shown in Figure 4(b). As a comparison, micromechanical modeling was carried out using the Hirsch and Manera models to predict the value of tensile strength and elastic modulus, respectively [31], as shown in Eqs. (3) and (4).

$$\sigma_c = x \cdot (\sigma_m \cdot v_m + \sigma_f \cdot v_f) + (1 - x) \cdot \frac{\sigma_m \cdot \sigma_f}{\sigma_m \cdot v_f + \sigma_f \cdot v_m}, \quad (3)$$

$$E_c = v_f \left(\frac{16}{45} \cdot E_f + 2 \cdot E_m \right) + \frac{8}{9} \cdot E_m, \quad (4)$$

where σ_c , σ_m , and σ_f represent the strength of the composite, matrix, and fiber (MPa), respectively. v_m and x represent the volume fraction of the matrix and parameter of short fiber orientation, respectively. E_c , E_m , and E_f represent the elastic modulus of the composite, matrix, and fiber, respectively.

2.2.5 Poisson's ratio test

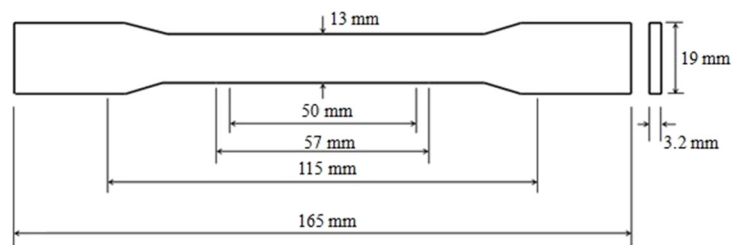
The Poisson's ratio test is carried out to determine the ratio of the lateral and axial deformation strain values which can be determined using Eq. (5) [32], based on ASTM D638 standard.

$$\nu = -\frac{\varepsilon_l}{\varepsilon_a}, \quad (5)$$

where ν , ε_l , and ε_a represent the Poisson's ratio, lateral strain (%), and axial strain (%), respectively. Poisson's ratio measurement used three devices to obtain data, namely, strain gauge, amplifier, and microcontroller, often referred to as data acquisition, as shown in Figure 5(a) and (b). Microcontroller is a tool used for data acquisition from



(a)



(b)

Figure 4: Tensile test of composites: (a) universal testing machine; and (b) specimen of tensile test based on ASTM D638.

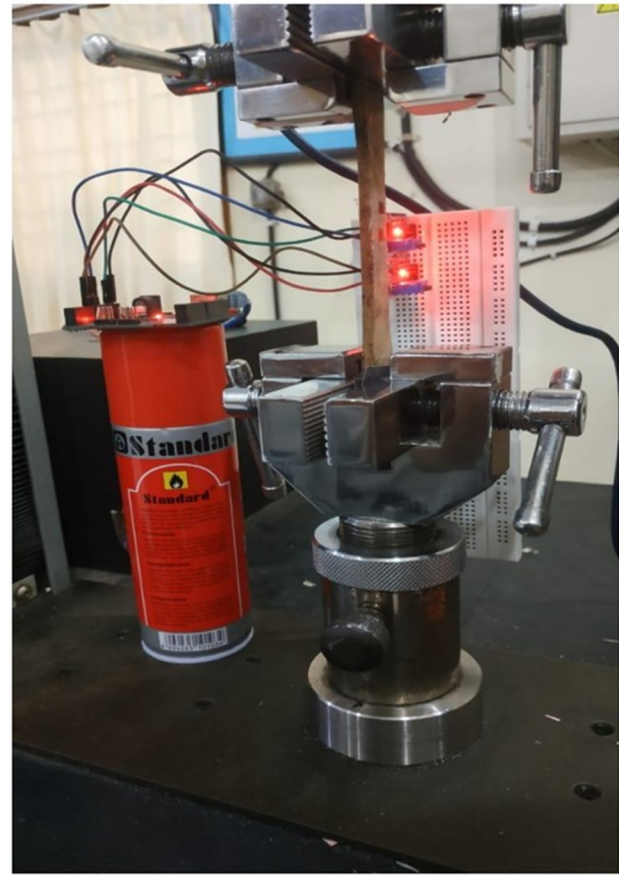
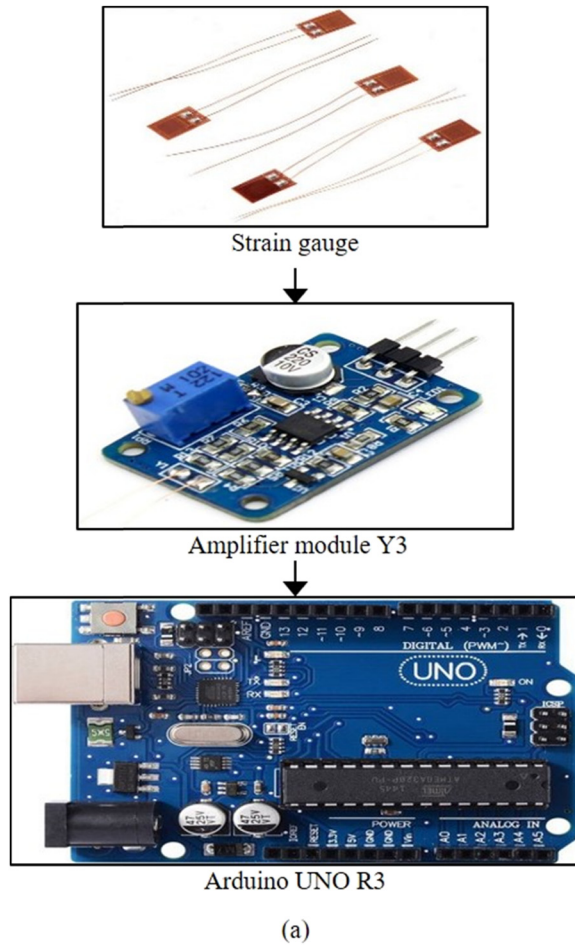


Figure 5: (a) Scheme of data acquisition for measuring Poisson's ratio; and (b) measurement of Poisson's ratio.

analog to digital. The Poisson's ratio results in data acquisition are still in the form of a voltage difference. The conversion from difference in electric stress to strain can be determined as follows:

$$\Delta E_a = \frac{\Delta E_{out}}{G}, \quad (6)$$

where ΔE_a , ΔE_{out} , and G represent the difference in the output voltage (V), the difference in the input voltage (V), and the amplifier gain, respectively. Meanwhile, the amount of strain can be determined based on Eq. (7).

$$\varepsilon = \frac{4 \times \Delta E_a}{GF \times E_{in}}, \quad (7)$$

where GF is the gauge factor.

2.2.6 Impact test

The amount of energy absorbed by the material when it receives an impact load was measured by Toyo Seiki

impact testing machine, as shown in Figure 6(a), made in Japan, where in the izod method, the specimen was placed standing [33]. In this case, the specimen refers to ASTM D5941, Figure 6(b). The impact strength is calculated from the potential energy of the pendulum before it is released minus the potential energy of the pendulum after it strikes the specimen, as shown in Eq. (8).

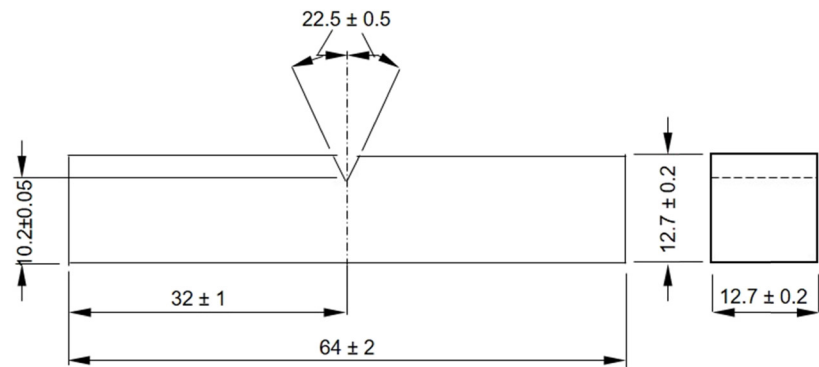
$$W = w \cdot R \cdot (\cos \beta - \cos \alpha'), \quad (8)$$

where W , w , R , β , and α' represent the total energy absorbed (J), the weight of the pendulum (N), the distance from the center of mass to the center of rotation of the pendulum (m), the angle of reflection of the swing arm (deg.), and the initial angle of the arm swing (deg.), respectively. The value of the impact strength of the specimen according to the thickness and width of the specimen is shown in Eq. (9).

$$\alpha = \frac{W}{h \times b} \times 10^3, \quad (9)$$



(a)



(b)

Figure 6: Izod impact: (a) testing machine; and (b) specimen dimension based on ASTM D5941.

where α , h , and b represent the value of the impact strength ($\text{J}\cdot\text{m}^{-2}$), the thickness of the specimen (m), and the width of the specimen (m), respectively.

2.3 Microscopic examination of UPR-CFs composite using SEM

The fracture morphology of UPR-CFs composites after tensile testing were examined using SEM to determine their fracture mechanism. The SEM equipment was FEI type Inspect-F50 (Figure 7), made in USA, with 10 kV voltage. Before examination, the specimens were gold-coated using sputtering machine and then laid on a silver-painted

holder. This SEM used low vacuum mode for non-conductive samples.

3 Results and discussion

3.1 Density of UPR-CF composites

Figure 8 indicates that the addition of MCC increases the density of UPR + MCC composites by 3.00% than the neat UPR. This is due to the higher density of MCC than that of UPR. The addition of MCC also reduces voids significantly. This case is in accordance with the study on the improvement of mechanical properties due to the addition of MCC to nylon 6 matrixed composites [34].

The effect of time variation in alkali treatment on CF on the theoretical and experimental density value of the UPR-CFs composite is also shown in Figure 8. The maximum density of UPR-CFs with alkaline-treated fibers is higher, 4.88 and 7.19% for both theoretical and experimental results, respectively, than the untreated one. The alkaline treatment partially eliminated the amorphous components of fibers, such as hemicellulose and lignin, so that the percentage of crystalline one, cellulose, was increased. The higher density of the crystalline component of the fibers than the amorphous ones causes an increase in the composite density [35]. The higher increase in experimental results than the theoretical ones is caused by the lower experimental density of the composite with untreated fibers than the theoretical one.

The average theoretical density values of the composites are higher than the experimental ones. This is caused by the assumption of the rule of mixture that there is no



Figure 7: SEM equipment.

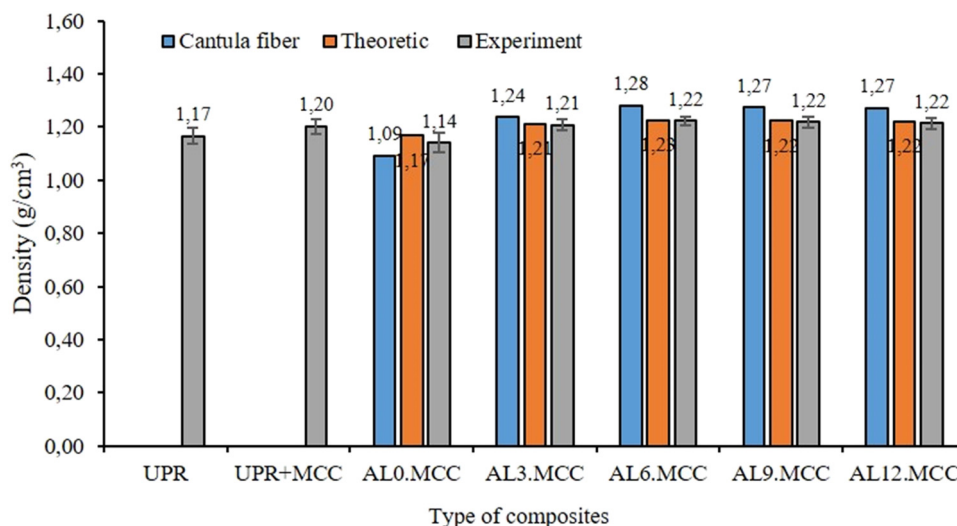


Figure 8: Theoretical and experimental density of UPR + MCC-CFs as a function of alkaline treatment time.

porosity in polymer composites [36]. The research results of the influence of time of alkaline treatment on the density value of CFs by Sakuri *et al.* [37] are used as a reference for the theoretical density value of UPR-CFs composite. Analysis of variance (ANOVA) done on experimental results indicates that the duration of alkaline treatment significantly influences the density of UPR-CFs composite. The highest density of composites due to alkaline treatment is achieved for 6 h alkaline treatment of CFs, for both theoretical and experimental results. Longer treatment leads to a decrease in composite density. It is due to the degradation of crystalline component of a fiber after the amorphous ones. The fiber also undergoes a polymorphic transformation from cellulose I to cellulose II which lead to the decrease in crystalline area of the fiber that in turn causes the decrease in composite density [38].

Cisneros-López *et al.* [39] investigated the effect of alkali treatment on the density value of CF reinforced linear low-density polyethylene composites. The result is 16.02% smaller than the UPR.MCC-CFs composite, because the addition of MCC increases the spread of cellulose with a wider micron size so that the density of the material will increase and the empty space will decrease. MCC also adds a filler component to the composite so that the density value will increase [35,40].

3.2 Tensile strength and elastic modulus of UPR-CF composites

The tensile strength value of the UPR-CFs composite can be seen in Figure 9(a). The theoretical tensile strength of the

composite is higher than the experimental one. This is due to the theoretical assumption that the fiber and matrix bond is perfect [36]. The theoretical calculation uses Hirsch's model because it can accurately predict the tensile strength of composites [31]. Sakuri *et al.* [17] studied the effect of alkali treatment time on the tensile strength of CFs, which used the micromechanical reference on the tensile strength of UPR-CFs. ANOVA done on experimental results reveals that the duration of alkaline treatment significantly influences the tensile strength of composites.

Figure 9(a) shows that the tensile strength of the UPR-CFs composite increased with the alkali treatment time. This is due to that the alkaline treatment of fiber can reduce the amorphous compounds, namely, hemicellulose, lignin, pectin, and other dirty substances. Reducing the amorphous area of the fiber increases the surface roughness of the fiber so that the interfacial bond between the fiber and the matrix becomes better [35]. Alkaline treatment of natural fibers also reduces hydrophilic properties in order to provide better interfacial bonds with hydrophobic matrices [41].

The most optimal tensile strength value of the UPR-CFs composite was obtained in the alkaline treatment for 6 h because it isolated cellulose effectively and removed elements such as lignin, hemicellulose, and impurities so that the tensile strength of the composite became better [37]. The maximum increase in the tensile strength due to the alkaline treatment was 8.75 and 22.17% for theoretical and experimental results, respectively. This was due to lower experimental tensile strength of composites with untreated fibers. The decrease in the tensile strength of the composites during the alkaline treatment for 9 and 12 h was

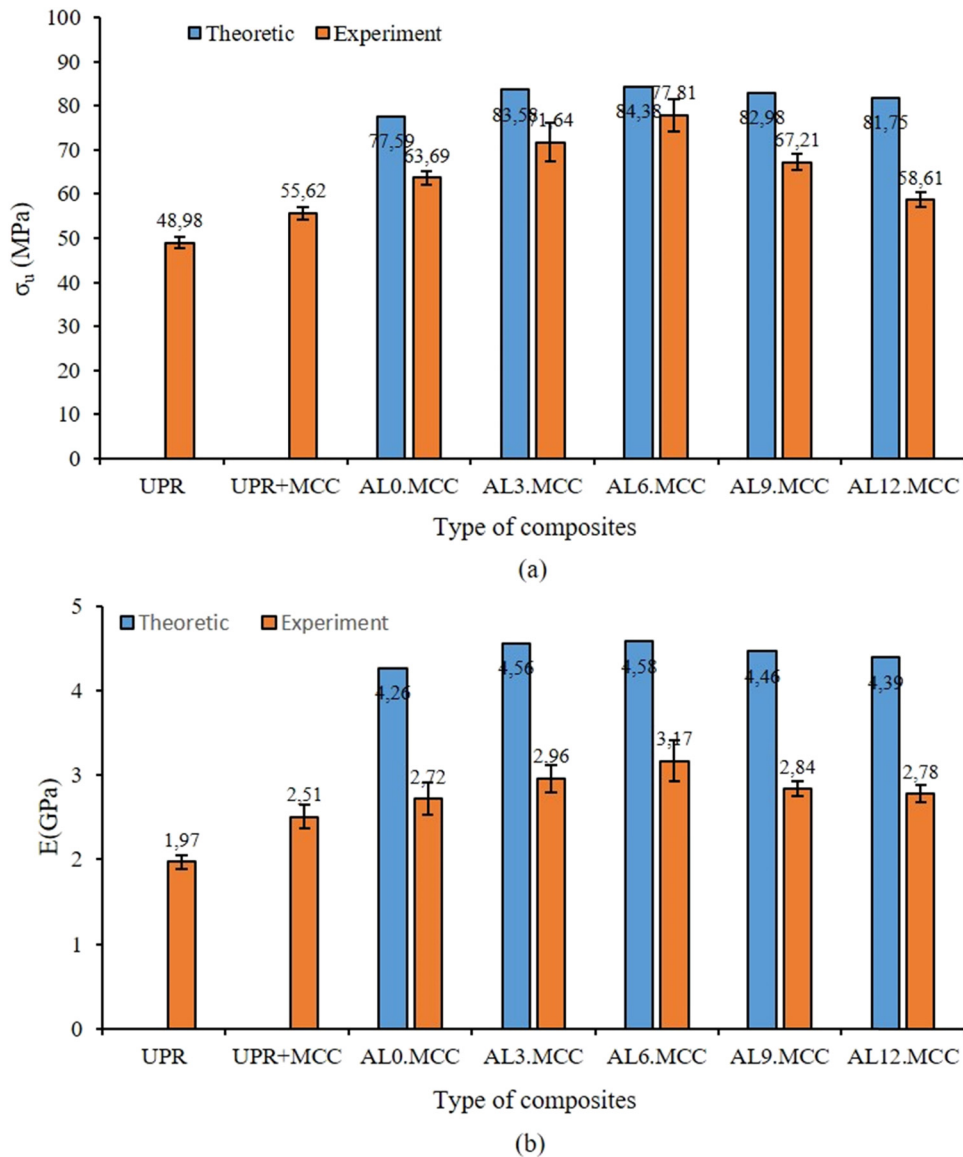


Figure 9: (a) Tensile strength; and (b) elastic modulus of UPR.MCC-CFs as a function of alkaline treatment time.

caused by the excessive degradation of lignin, hemicellulose, and cellulose elements [42].

The value of the composite tensile strength is also affected by the addition of MCC. In Figure 9(a), it can be seen that the tensile strength of the UPR + MCC composite is 13.57% higher than the neat UPR polymer because MCC is a micron-sized crystalline cellulose material. In addition, the tensile strength of the UPR-CFs composite with alkaline treatment for 6 h was 58.86% higher than the neat UPR caused by micron-sized MCCs which expanded the bond surface, thus increasing the compatibility between UPR and CFs. MCC size also increased the density between materials so that voids in the composite were reduced causing load transfer from matrix to fiber to be better. This was in accordance with the study of Sakuri *et al.* [37] related to

mechanical properties of CFs reinforced UPR subjected to MCC.

Nesimnasi *et al.* [43] compared the effect of alkali treatment time with a concentration of 5% on the tensile strength of the UPR-CF composite fabricated using the compression molding method but not using MCC, the highest tensile strength of the composite was 36.86 MPa. These results show that the UPR-CFs composite with the addition of MCC has a 52.6% higher tensile strength than without MCC. This is due to the addition of MCC increasing the spread of micron-sized crystalline cellulose, where cellulose is the component that has the highest strength in natural fibers. In addition, the addition of MCC also increases hydrogen bonding so as to improve the tensile strength of the composite [18,34].

The elastic modulus value of the UPR-CF composite with variations in alkali treatment time is shown in Figure 9(b). The theoretical value of the elastic modulus is higher than the experiment because the Manera rule of mixture model uses the assumption of perfect matrix and fiber bonds. The Manera method is a method for predicting the elastic modulus of composites with the condition that the matrix elastic modulus (E_m) is between 2 and 4 GPa [31,37].

The experimental elastic modulus obtained was analyzed statistically using ANOVA. It indicated that the time of alkaline treatment significantly influenced the values of elastic modulus. They increased due to the alkaline treatment, but after 6 h, there was a decrease. This was caused by an increase in the tensile strength curve compared to a decrease in the strain curve before 6 h treatment. The alkaline treatment removed hemicellulose, lignin, and other impurities thereby improving the adhesion bond between the fiber and matrix [42].

Alkaline treatment of CF for 9 and 12 h caused a decrease in the value of the elastic modulus. This was due to the prolonged alkaline treatment causing damage to the structure of hemicellulose, lignin, and cellulose in the CFs, thereby reducing the tensile strength of the composite. In addition, the long alkaline treatment increased the strain because the bond between the fiber and the matrix was very strong so that the CFs stretched longer together with the matrix. However, these fibers became brittle so that the load received by the UPR matrix was more dominant. As a result, the composite strain was more dominated by the matrix [44].

The elastic modulus of the UPR composite with the addition of MCC was 27.41% higher than that of neat UPR. This was due the addition of MCC filler increasing the stiffness so that the elastic modulus increased. These results were in accordance with the research of Kiziltas *et al.* [34], namely, the addition of MCC to the nylon 6 matrix resulted in an increase in the elastic modulus value of the composite. In addition, Mathew *et al.* [45] conducted the same study using polylactic acid (PLA), with the MCC addition, the elastic modulus was higher than that of neat PLA because the addition of MCC made the composite strain value decrease [46].

The value of the modulus of elasticity in the UPR + MCC-CFs composite was higher than that of UPR-sugarcane waste composite [47]. This happened because the micron-sized MCC increased the wider distribution of reinforcing filler which made the tenacity value of the composite to decrease so that the value of the elastic modulus of the composite increased [34]. The components of cantala and bagasse fiber have almost the same components, *i.e.*, CF contains cellulose, hemicellulose, and lignin, respectively, by 65, 12, and 9.9%, while bagasse fiber contains 55.2, 16.8, and 25.3%, respectively [48].

3.3 Impact strength of UPR-CFs composite

The impact strength of the UPR.MCC-CFs composite can be seen in Figure 10. Analysis of variance revealed that the

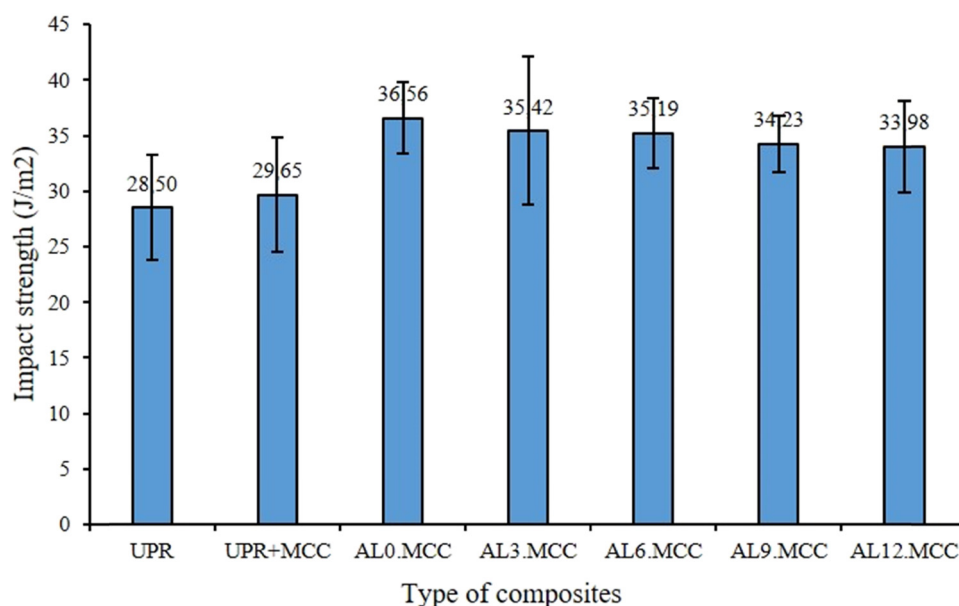


Figure 10: Impact strength of UPR + MCC-CFs composite as a function of alkaline treatment time.

duration of alkaline treatment did not significantly influence the strength of composites. However, the average impact strength achieved its maximum value, $36.56 \text{ kJ}\cdot\text{m}^{-2}$, in the composite without alkaline treatment (AL0.MCC). This was due to a poor fiber and matrix interface bonding in the impact fracture which forms an irregular fracture direction which made the composite able to withstand shock loads and absorb more energy. In contrast, UPR-CF composites with alkaline treatment having good fiber–matrix interface bonding produced straight and fast impact fractures because the fiber and matrix withstood loads together resulting in lower impact strength [49]. The highest decrease in impact strength was 7.05% for 12 h of alkaline treatment. These results were in accordance with the existing research, namely, longer alkaline treatment tended to decrease the impact strength of the composites [50].

Previous studies also had proven that the impact strength of the UPR-arena fiber composites decreased with increasing duration of alkaline treatment. This was caused by the loss of the arena fiber structure, namely, lignin, hemicellulose, and cellulose so that the energy absorbed by the composite was reduced which resulted in a decrease in impact strength [50].

The addition of MCC to the UPR-CFs composite increased the impact strength up to 4.02%. This was due to the addition of MCC filler with the right concentration to make MCC dispersion in the UPR perfect without agglomeration so that the load transfer was held together between the matrix and MCC. Neat UPR had an impact strength of $28.50 \text{ kJ}\cdot\text{m}^{-2}$, while the impact strength of the UPR + MCC-CFs composite was $29.65 \text{ kJ}\cdot\text{m}^{-2}$. However, the addition of MCC with an excess concentration reduced the impact strength because the mobility of the polymer matrix decreased so that it could not disperse MCC completely which resulted in the not-optimal bonds between filler and matrix [34].

The most optimal UPR-CFs composite with the addition of MCC was in a correlation with the study by Rahman *et al.* [46] who obtained an impact strength of the UPR-arena fiber composite without the addition of MCC of $32.1 \text{ kJ}\cdot\text{m}^{-2}$. The increase in impact strength was due to MCC increasing the structure in the form of micron-sized crystalline cellulose so that it can absorb more energy [17].

3.4 Poisson's ratio value of UPR-CFs composite

The Poisson's ratio value of the UPR-CFs composite with variations in the alkaline treatment time can be seen in Table 2. It shows that the longer treatment time caused the Poisson's ratio value of the composites to decrease. This

Table 2: Poisson's ratio of UPR.MCC-CF composite as a function of alkaline treatment time

Composite specimens	ε_x	ε_y	ν_{xy}
UPR	0.0054	0.0128	0.421
UPR.MCC	0.0051	0.0127	0.401
AL0.MCC	0.0045	0.0119	0.378
AL3.MCC	0.0047	0.0126	0.373
AL6.MCC	0.0048	0.0130	0.369
AL9.MCC	0.0042	0.0113	0.371
AL12.MCC	0.0041	0.0110	0.372

was due to the longer alkaline treatment of the fibers which caused the amorphous areas to disappear so that the composite strain value decreased and the Poisson's ratio value also declined. The amorphous region is the first region to bear loads before the crystalline region of natural fibers. The UPR-CFs composite with the highest Poisson's ratio was the fiber composite without alkaline treatment (AL0.MCC) because the fiber still had a lot of amorphous regions so that it could withstand more loads resulting in increased strain values [51,52]. The alkaline treatment for 6 h decreased the Poisson's ratio of composites by around 2.38% compared to the untreated ones. This minimum Poisson's ratio was achieved due to the elimination of amorphous components of the fibers, such as hemicellulose and lignin. The dominance of cellulose led to the higher stiffness of the composite causing lower Poisson's ratio obtained.

The Poisson's ratio of neat UPR was 4.99% higher than the UPR + MCC composite. This was due to the nature of the MCC material which had high stiffness, making the composites have low strain values in both the lateral (x) and axial (y) directions [34]. In addition, thermoset polymers have strong crosslinks and covalent bonds so that the Poisson's ratio tends to be high. UPR-natural fiber polymer composites have a lower Poisson's ratio than the neat UPR composites due to the addition of fiber filler and many crosslink molecules so that the stiffness properties increase [53,54].

3.5 Microscopic observation of UPR-CFs composite

Microscopic observation of the UPR + MCC-CFs composite was carried out on fractures of the composite tensile test specimen observed using SEM, as shown in Figure 11(a). The cross section shows the quality of the fiber-matrix bond, porosity, fiber breakage, and fiber pullout [52].

The fracture of the tensile test specimen on the neat UPR composite, which is shown in Figure 11(a), still has

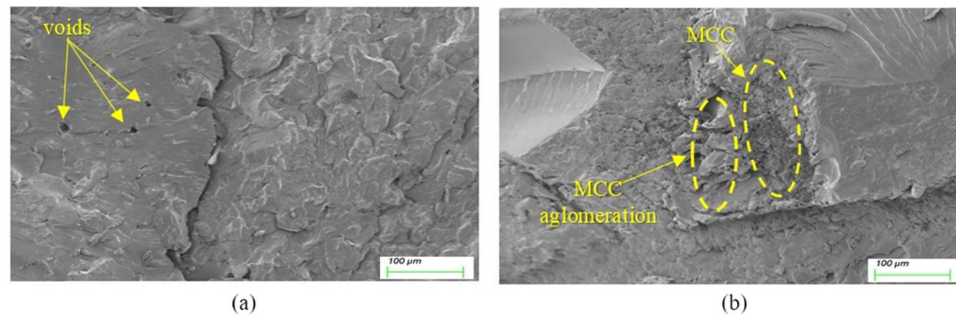


Figure 11: SEM image of: (a) neat UPR composite fracture; and (b) UPR + MCC composite fracture.

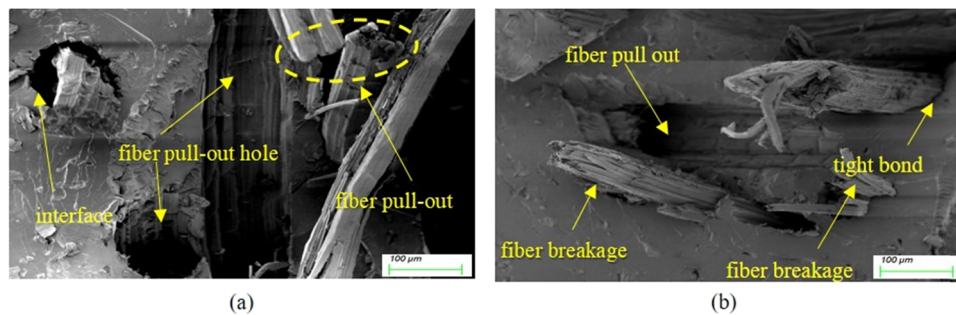


Figure 12: SEM image of UPR + MCC-CFs fracture: (a) without alkali treatment; and (b) with alkaline treatment for 6 h.

many voids and the shape of the fracture is flat and smooth caused by the absence of fibers so that it cannot withstand excessive loads and indicates high brittle properties. For comparison, Figure 11(b) shows that some of the MCC grains bond well with the UPR matrix so that the tensile strength of the composite increases. The fracture of the UPR + MCC composite occurs because there are still many interfaces between the UPR and MCC matrices so that the bond is not optimal. In addition, the composite fracture also has a little agglomeration caused by the inhomogeneous MCC distribution [34,35,55].

Figure 12(a) shows that in the UPR + MCC-CFs composite without alkaline treatment (AL0.MCC), the fiber surface is still smooth and then there is an interfacial gap and it is dominated by pull-out fiber. The interface was caused by the fiber still containing lignin, hemicellulose, and other impurities, as well as differences in the hydrophilic and hydrophobic properties of the matrix which made the mechanical interlocking between the fiber and the matrix poor so that the composite experienced fiber pull-out [56,57]. Fracture with fiber pull-out on the composite indicates that its strength is lower than the one with fiber breakage [44].

Figure 12(b) indicates that in the UPR + MCC-CFs composite with alkaline treatment for 6 h, there are some fiber breakage and little fiber pull-outs. The tight bonds between

fibers and matrix in the composite are due to the elimination of hemicellulose and lignin. In addition, alkaline treatment increases the roughness of the fiber so that the bond between natural fibers and the thermoset matrix becomes more optimal when the matrix transfers the load to the fiber so that the tensile strength of the composite becomes higher [56,58].

The results of these microscopic observations were confirmed by Sakuri *et al.*'s observations regarding the morphology of CFs with variations in the time of alkaline treatment [17]. The result is that fiber without alkaline treatment still has a smooth surface and contains a lot of lignin, hemicellulose, and impurities, while CF with alkaline treatment for 6 h can remove lignin, hemicellulose, and other impurities effectively which makes the surface area of the fiber. The CF gets wider and coarser so it forms a better bond with the matrix.

MCC on the UPR-CFs composite makes the composite surface smoother because of its micron-sized particles. A smooth surface indicates that the material has intermolecular cross-links. In addition, MCC also reduces voids or porosity so that the strength of the polymer matrix composite increases. The addition of MCC to the composite increases the strength of the composite because the bond between MCC and the matrix is getting better. This is because in MCC there are only crystalline regions, namely, pure cellulose compounds [59,60].

4 Conclusion

From the results and discussion of the research, following conclusions can be drawn:

- Theoretically, alkaline treatment of the CFs significantly increased the values of density, tensile strength, and elastic modulus of the CF-reinforced unsaturated polyester composites of 4.88, 8.75, and 7.51%, respectively, but experimentally, it significantly raised them 7.19, 22.17, and 16.50%, respectively. The alkaline treatment decreased the value of Poisson's ratio by 2.38% and did not change the impact strength of UPR-CFs composite significantly. However, longer treatment reduced the tensile strength and modulus due to the excessive elimination of hemicellulose and lignin, and the degradation of cellulose.
- The addition of MCC to the UPR-CF composites significantly increased the values of density, tensile strength, and elastic modulus of 3.00, 13.57, and 27.41%, respectively. It decreased the value of Poisson's ratio by 4.75% and did not change the impact strength value significantly.
- Fracture of composites reinforced with CF without alkali treatment was dominated by fiber pull-out, while composites with alkali treatment indicated a lot of fiber breakage. This indicated that the alkaline treatment enhanced the interfacial strength between unsaturated polyester matrix and CF surface.

The increase in strength and modulus of elasticity due to the alkali treatment and the addition of MCC allows for the widespread use of these composites for structures with higher load bearing.

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