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Influence of Water Immersion on Flexural and Impact Strength of Microcrystalline Cellulose-added Unsaturated Polyester-matriced Composites Strengthened by Untreated, Fumigated and Alkalized Cantala Fibers

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Abstract:The aims of this study is to investigate the effect of immersion on flexural and impact strength of UPRs-CFs composites with addition of MCC. Composites were made with 30% v_f of CFs using compression molding process. The specimens were soaked into 3 types of fluids: fresh water (represented by distilled water), sea water, and acidic solution. Each variation of immersion involved 4 types of composites: UPRs-MCC, UPRs-UCFs, UPRs-FCFs, and UPRs-ACFs composites. The immersion process took 60 days. The highest mass and dimensional changes were obtained for composites soaked in fresh water, followed by acidic solution, and sea water. UPRs-ACFs composites had the lowest water absorption rate, followed by UPRs-FCFs and UPRs-UCFs composites in all fluid types. Both flexural and impact strength decreased after immersion compared to control specimens. The 60-days-immersion accelerated degradation of specimen components, microscopic crack growth, and deterioration of the interfacial area between matrix and fibers.

Keywords: different fluid immersion; mechanical properties; fumigation treatment; alkaline treatment; UPRs; cantala fibers; microcrystalline cellulose

1. Introduction

One of the materials that are widely used in industry, such as cars, planes and construction, is a composite[1),2)], which is a combination of two or more materials that produces a new material with superior characteristics from its constituents[3),4)]. Current environmental issues encourage the development of environmentally friendly materials, including biocomposites, which at least one of its constituent components comes from natural materials [5)]. Most biocomposites consist of a polymer matrix with natural fiber reinforcement such as flax, hemp, agave, kenaf[6)], areca nut[7)], groundnut shell[8)], and jute fibers. In comparison with synthetic fibers, including glass[9)] and carbon fibers[10)], natural fibers are cheaper, have low density, are easily extracted, renewable, widely available, and environmentally friendly[11)].

The agave plant is known as a source of fiber[12)]. The agave species that mostly grow in Indonesia is Agave cantala[13)]. Given its great potential, its function needs to be improved, not only as a traditional product, but also as a composite reinforcement[14)]. Hydrophilic natural fibers have low compatibility with synthetic polymer matrices having hydrophobic nature. Broadly, this deficiency is overcome by chemical treatment to eliminate some hemicellulose and lignin content, so that the fiber surface becomes clean, rougher and has better adhesiveness[15)]. Besides the commonly used alkaline treatment[13)], other treatment alternatives are sodium-bicarbonate treatment[16)] and fumigation[11)] using smoke. The smoke resulted from burning coconut shell contains compounds of carbonyl, phenol and acetic acid[17)]. In fumigation, there is a process of withdrawing water and depositing chemical compounds from the smoke, thereby changing the chemical and physical properties of the fiber surface. Carbonyl compounds contribute to the shift in the color of product surface from brownish yellow to dark brown, while phenols act as antimicrobials and antioxidants, causing the product to be durable[18)].

Besides thermoplastics, polymers that are often used as

matrices include thermosets, such as polyurethane (PU), unsaturated polyester resins (UPRs), phenols, polymides and epoxy. UPRs are synthetic resins having straight chains produced from the reaction of glycol with difunctional acids such as adipic and maleic acid. In addition to having high strength, UPRs are cheap and easy to manufacture[19].

The component of natural fibers having the highest strength is cellulose. It is a polymer that naturally resulted from biomass, which is abundant and renewable[20]. The lack of dispersion of natural fiber cellulose in the composites was overcome by using micro-crystalline celullose (MMC)[21]. MMC is pure cellulose extracted from alpha cellulose using mineral acids. It is renewable, non-toxic, and relatively cheap. Also, it has good mechanical properties and high biocompatibility[22]. Further purification of cellulose results in nano-cellulose[23].

Composites reinforced by natural fibers tend to absorb water in humid environments or when immersed in liquids[24]. Research by Nosbi et al.[25] showed that there are significant differences between kenaf fiber composites immersed in different environmental conditions, where the composites soaked in fresh water showed the highest liquid absorption rate, followed by acid solution, while those immersed in sea water had the lowest absorption values. The tensile strength of kenaf fibers changed with increasing immersion time, which indicated fiber degradation. Microstructure investigation of dyed kenaf fiber showed micro-gap development and increased surface roughness of the fibers.

The utilization of the UPRSs+MCC-cantala fiber composites has a potency to be increased for both indoor and outdoor use. However, the influence of water absorption on these composites on their mechanical characteristics, especially flexural and impact strength is unknown. This study used distilled water to represent fresh water, sea water and acid solution while the fibers used were untreated, alkaline-treated and fumigated.

2. Methods

This study consisted of four main steps: fiber treatment, composites fabrication and immersion, followed by mechanical testing. Water absorption rate, and mass and dimension change of the composites were measured daily until 60 days. All activities are shown in a flowchart (Fig. 1).

2.1 Materials

Fibers were obtained from the cantala plantation in Pengasih District, Kulon Progo Regency, Special Region of Yogyakarta Province, as shown in Fig. 2. The leaves of cantala plants are cut, then extracted to obtain the fibers. The drying is done for 1-2 days until the fibers become dry.

The average density of untreated cantala fibers is 1.056 ± 0.0145 g/cm^3[3),26]. The average single fiber tensile strength and elastic modulus of untreated cantala fiber are 245.87 ± 26.75 MPa and 4.96 ± 0.13 GPa, respectively. Its interfacial shear strength (IFSS) and critical length with UPRs and MCC matrix are 2.49 ± 0.02 MPa and 6.37 ± 0.98 mm, respectively[27].

UPRs used was Yukalac 157-BQTN EX, an ortho-phthalic resin type, having a density of 1.12 g/cm^3 and insoluble in water, purchased from Justus Kimiaraya Company. The catalyst, methyl ethyl ketone (MEKPO), was also bought from the same company. The MCC powder having a density of 0.6 g/cm^3 at 25^{o}C was purchased from Sigma-Aldrich Pte Ltd.

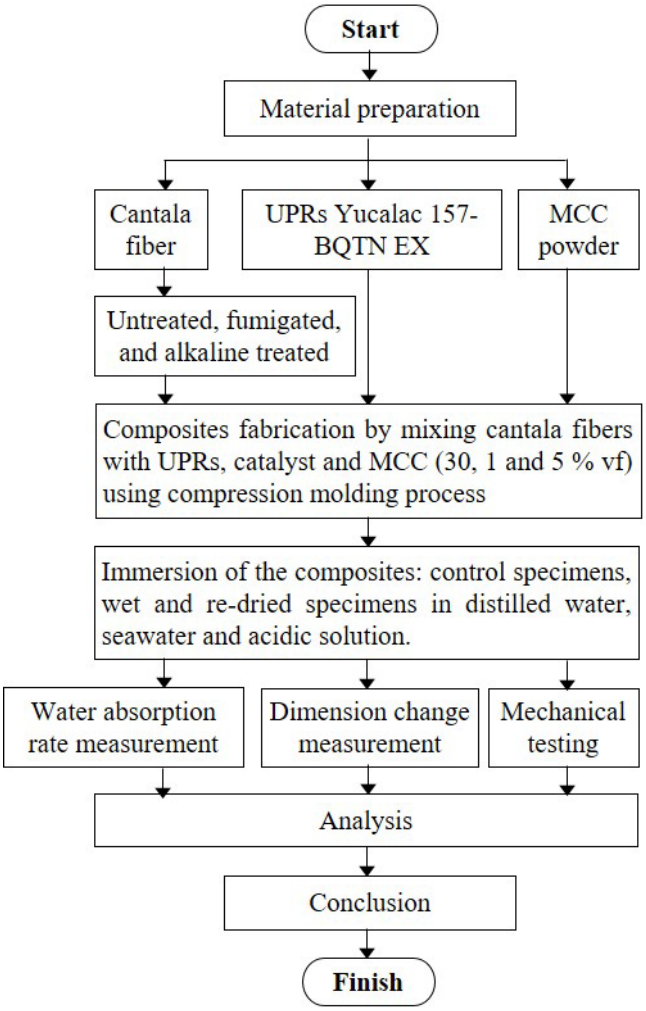
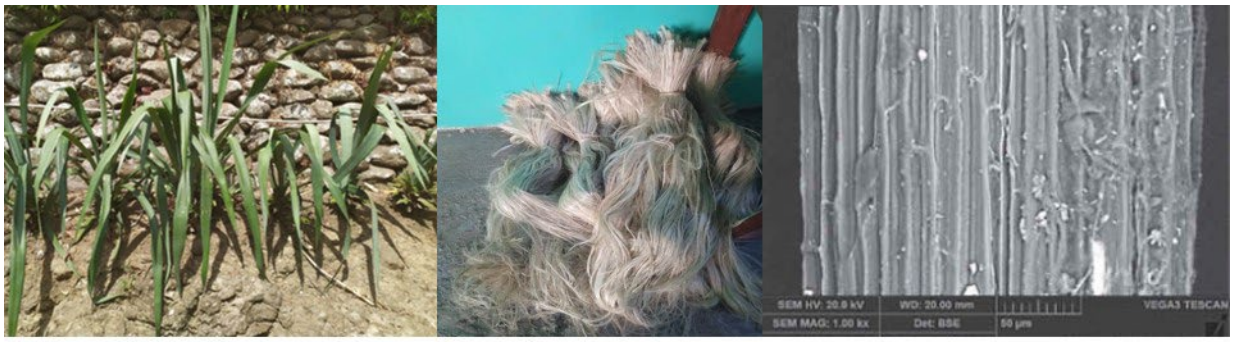


Fig. 1: Research flowchart

Acetic acid of 99% was added by distilled water to obtain acidic solution with pH = 3. The solution was then used as soaking media of the composites.

2.2 Fiber treatment

Some fibers were untreated, treated by fumigation and the other by alkaline immersion. The fumigation process was carried out for 10 h using smoke from burning coconut fibers. The temperature on the fiber is controlled between 40 - 60^{o}C and every 1 hour the fiber is turned



(a) (b) (c)

Fig.2: (a) Agave cantala plants; (b) Cantala fibers; and (c) Surface morphology of a cantala fiber observed using SEM[26)]

over so that the fumigation process is evenly distributed. The cantala fibers were then cut to a 10 mm-sized length and put into a plastic container. The alkaline immersion of cantala fibers was carried out for 6 h with 6% sodium hydroxide solution. Furthermore, the cantala fiber was rinsed and soaked in 1% acetic acid solution, then rinsed using distilled water. The drying process was carried out without exposure to direct sunlight. The cantala fibers were then also cut into 10 mm length.

The fumigation of cantala fibers for 10 h increased the average single fiber tensile strength, elastic modulus, IFSS and critical length became 373.72 ± 28.56 MPa, 7.89 ± 0.11 GPa, 3.39 ± 0.03 MPa, and 8.46 ± 0.78 mm[27)]. The alkaline treatment of cantala fibers with 6% NaOH for 6 h increased the average IFSS became 3.67 MPa[28)].

2.3 Fabrication of composites

The cantala fibers of 30 % v_f was mixed with UPRS matrix, catalyst and MCC with 64, 1 and 5 % volume fraction, respectively, then poured into a 170 x 110 x 10 mm-sized-die of a compression molding machine. The pressure of 10 kg/cm^2 was applied to the dies with a duration of 12 hours. The composites formed then put in the oven for 12 hours with a temperature of 50^{o}C. The MCC addition of 5 % v_f was based on the study of Sakuri and co-workers[29)].

2.4 Immersion of the composites

The neat specimens of UPRS+MCC (UPRs-MCC), UPRS+MCC-cantala fibers without treatment (UPRs-UCFs), with fumigation treatment (UPRs-FCFs) and alkaline treatment (UPRs-ACFs) were divided into three groups, namely control, wet and re-dried specimens. Control specimens was specimens without immersion. Some specimens were immersed for 60 days in 3 types of liquid: distilled water, sea water and acidic solution, having pH = 7, 8 and 3, respectively. Some specimens were then directly weighed and measured in dimensions, named as wet specimens. The others were dried by placing it in an oven for 24 h with 50^{o}C temperature then weighed and measured its dimensions, named as re-dried specimens.

The weight and thickness of test specimens were measured using a scale and a digital caliper having 0.001 g and 0.01 mm accuracy, respectively. The measurements were carried out every day at the same time for 60 days.

2.5 Water absorption rate

The rate of water absorption can be calculated using Equation (1) for room temperature and atmospheric pressure[30)].

$$M_m = \left(\frac{M_2 - M_1}{M_1}\right).100\% \quad (1)$$

in which M_m, M_1 and M_2 represent water absorption rate, initial and final mass of the specimen. The relationship between diffusion coefficient and water absorption rate can be represented by Equation (2)[30)].

$$D = \pi\left(\frac{h}{4M_m}\right)^2\left(\frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}}\right)^2 \quad (2)$$

in which D, t_1 and t_2 are diffusion coefficient, initial and final time, respectively. The data was taken every day in the same time up to 60 days.

2.6 Dimension change

Percentage of dimension change can be calculated using Equation (3)[31)]. Slope of dimension change also can be determined based on Equation (4)[31)].

$$h_{max} = \left(\frac{h_2 - h_1}{h_1}\right).100\% \quad (3)$$

$$S = \frac{h_2 - h_1}{\sqrt{t_2} - \sqrt{t_1}} \quad (4)$$

in which h_{max}, S, h_1 and h_2 represent percentage and slope of dimension change, initial and final thickness of the specimen, respectively. The value is averaged from 3 specimens measured in 3 different parts of each specimen.

2.7 Mechanical testings

The mechanical properties tested were flexural and impact strength. The three points bending and izod impact testing were done according to ASTM D790 and D5941, respectively. While the wet specimen or wet specimen is a specimen that before being tested, the surface of the specimen is first wiped with a dry cloth.

2.8 Morphological observation on composite surface

Before and after fluid submersion, the composite surface was examined using a Carl Zeiss® type Evo 10 scanning electron microscope (SEM) instruments with voltage and frequency of 100-250 V and 50-60 Hz, respectively. Before SEM examination, the nonmetal samples must be gold coated using a plasma sputtering equipment at 2 mbar pressure for 1 min.

3. Results and Discussion

3.1 Water absorption rate

In immersion tests, specimens in all types of fluid absorb liquid. This can be seen from the increase in mass and thickness of the specimen. During the 60-day testing process the specimens absorb water to a saturation point, that is, there is no increase in mass and thickness in the specimen. This case is consistent with Fickian's law when the specimen reaches an equilibrium point after the starting point[32]. As shown in Fig. 3, the relationship curve between the rate of water absorption (% M_m) and the root of time ($t^{1/2}$) in each fluid shows the saturation point after several days of immersion. The curve data is from data calculated based on Equation (1).

Fig. 3 shows that for UPRs specimens with the addition of MCC the percentage of fluid absorption is the smallest compared to composite specimens with cantala fibers. This is due to the uniformity of elements in the UPRs-MCC specimens compared to other composites such as UPRs-cantala fibers composite specimens, avoiding the existence of fiber-matrix interfaces then causing the specimens to be more hydro-phobic. These results are consistent with the research of Najafi and co-workers[33] which finds that hydrophilic natural fibers contribute significantly in the ability of composites to absorb fluid. The amount of fiber used as a matrix reinforcement is directly proportional to the ability of the composite to absorb fluid. However, in UPRs-MCC specimens, fluid absorption still occurs, as indicated in Fig. 3. This is as a result of the presence of some bubbles in the UPRs matrix and the existence of UPRs-MCC interface which affects the ability of the fluid to move into the specimen. The bubbles form when UPRs, catalyst, and sometimes MCC, are mixed together, as occurred in the research of Sakuri and co-workers[29].

In the UPRs-cantala fiber specimen without fiber treatment, the highest fluid absorption occurs in all types of fluid. The fumigation and alkaline treatment of the fiber reduce water absorption rate due to the removal of hemicellulose which absorbs more water compared to cellulose. This is appropriate to the previous study by Ariawan and co-workers[34] on the reduced water absorption rates associated with removal of hemicellulose due to the alkaline treatment of kenaf fibers. This reduction also occurs in specimens with fumigated fibers, because fumigation treatment can reduce the hemicellulose content in the fiber[12].

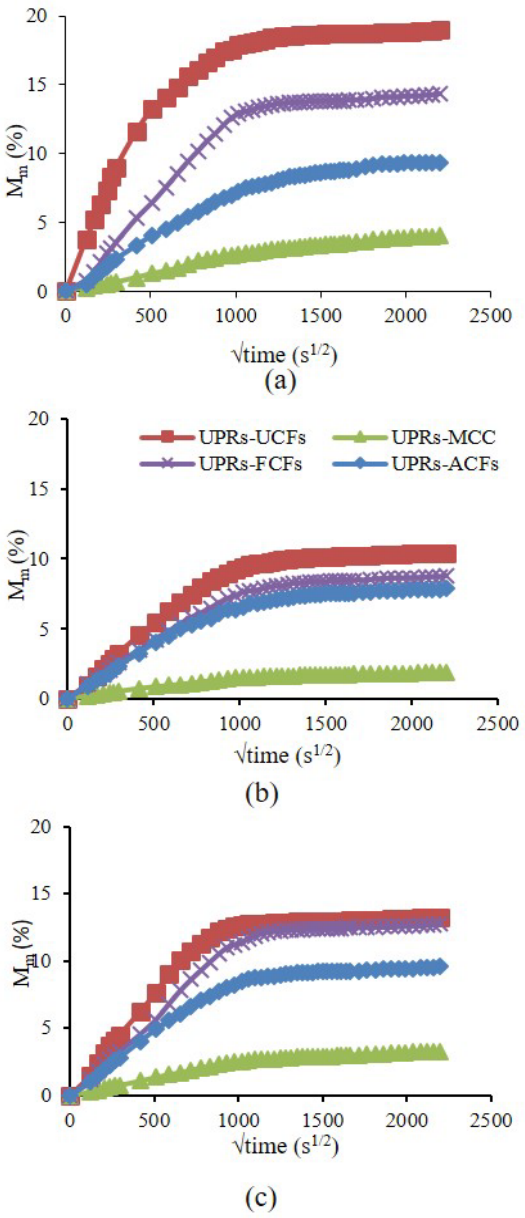


Fig.3: Percentage of water absorption of specimens for different type of liquids: (a) fresh water; (b) sea water; and (c) acidic solution

3.2 Diffusion coefficient (D)

The diffusion coefficient of fluid, D, determined from Equation (2), is shown in Table 1. It reveals that the highest D is obtained for UPRs-cantala fiber composite specimen without fiber treatment followed by specimens with fumigation and alkaline treatment, and specimens of UPRs-MCC. It indicates that the value of D is directly proportional with M_m. In the study of Sreekumar and co-workers[35], the entering of moving fluid molecules into the fiber is followed by the entry of fluid into the voids in the composite during the immersion process. Table 1 reveals that D value in sea water immersion has the lowest values. It is consistent with the research of Nosbi and co-workers[24] explaining that the presence of ionic salts in seawater blocks the diffusion path after prolonged

exposure thereby retarding the absorption process. The D value of the acid solution is in the middle position, which is lower than the specimen soaked in distilled water, and higher than in sea water.

Table 1. Maximum value of absorbed water M_m and diffusion coefficient D

Fluid type	Fresh water			
Specimen	UPRs-MCC	UPRs-UCFs	UPRs-FCFs	UPRs-ACFs
M_m (%)	9.47	21.24	14.53	13.2
D (x 10^{-3} mm^2/s)	4.01	18.98	14.33	9.79
Fluid type	Sea water			
Specimen	UPRs-MCC	UPRs-UCFs	UPRs-FCFs	UPRs-ACFs
M_m (%)	5.92	16.44	10.72	11.2
D (x 10^{-3} mm^2/s)	1.93	10.54	8.751	7.87
Fluid type	Acidic solution			
Specimen	UPRs-MCC	UPRs-UCFs	UPRs-FCFs	UPRs-ACFs
M_m (%)	8.77	18.34	14.16	12.2
D (x 10^{-3} mm^2/s)	3.54	13.46	12.8	9.58

3.3 Dimension change

The change of dimension in immersion testing is expressed by percentage change in dimensions (h_{max}) calculated using Equation (3), as shown in Fig. 4. The curve indicates that the dimensional change has increased with immersion time. Near the starting point, the dimensional change in all types of fluids increases rapidly, until after several weeks it reaches the saturation point of immersion. The slope of the change in dimensions is indicated in Table 2, as results of calculation using Equation (4).

Table 2. Gradient of dimension change S and maximum dimension change h_{max} due to specimen submersion in fresh water, sea water, and acidic solution

Fluid type	Fresh water			
Specimen	UPRs-MCC	UPRs-UCFs	UPRs-FCFs	UPRs-ACFs
S (%/h)	0.008	0.018	0.013	0.013
h_{max} (%)	4.239	9.212	7.218	7.091
Fluid type	Sea water			
Specimen	UPRs-MCC	UPRs-UCFs	UPRs-FCFs	UPRs-ACFs
S (%/h)	0.005	0.014	0.012	0.011
h_{max} (%)	2.984	6.556	6.483	6.228
Fluid type	Acidic solution			
Specimen	UPRs-MCC	UPRs-UCFs	UPRs-FCFs	UPRs-ACFs
S (%/h)	0.006	0.016	0.012	0.013
h_{max} (%)	3.442	8.058	6.527	6.273

In Figure 4 and Table 2, both the value of h_{max} and S show that the sea water immersion has the lowest value, meanwhile, the immersion in distilled water showed the highest ones. This fact occurs due to the blocking the diffusion pathway by the presence of ionic salts in sea water after long-term exposure. It is correlated to the study of Nosbi and co-workers[24]. The study of Akil and co-workers[30] also reveals that large molecules in seawater and acidic solutions reduce the diffusion rate into the composite matrix resulting in lower liquid absorption. The absorption process in distilled water is higher due its purity and lower molecules.

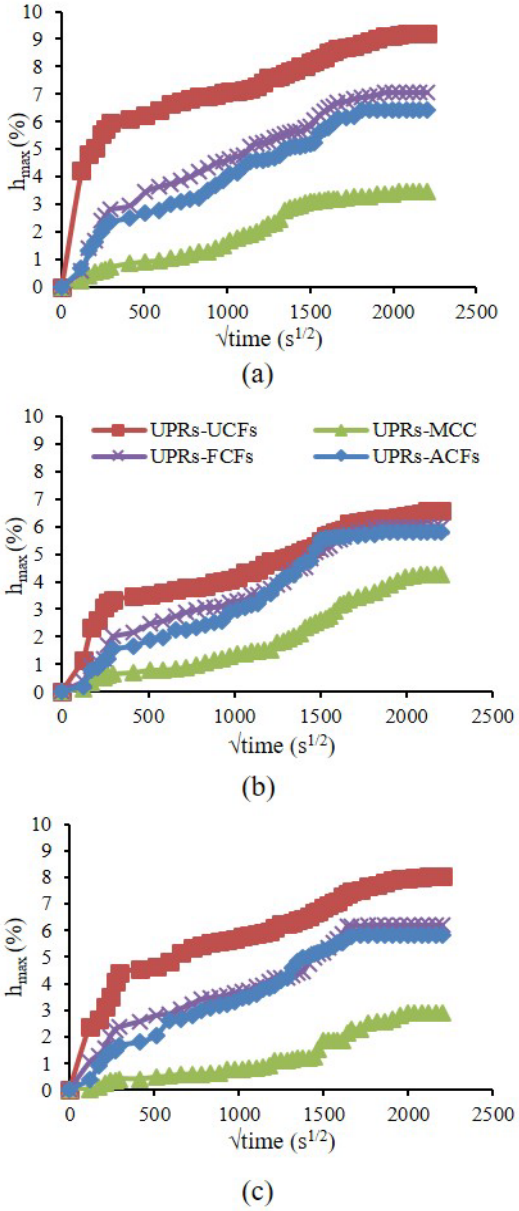


Fig.4: Dimension change of specimens in different type of liquids: (a) fresh water; (b) seawater; and (d) acidic solution

3.4 Impact strength

The impact strength obtained for some specimens is shown in Fig. 5 as histograms. It indicates that testing

done on wet specimens has a better impact strength value than on re-dried specimens. This is due to the existence of water as plastisizing agents which fill the gaps of matrix-fiber interfacial bonds and enhance their flexibility. It is also corresponds to the study of Alamri and Low[36] that the water absorption in the specimens caused fibers swelling which can increase the the impact strength caused by the hydrogen bonds formation between fibers and water molecules so that the dipole interaction between fiber and matrix molecules decreases, which further increases the flexibility of the polymer chain which causes the enhancement of impact strength.

Fig. 5 indicates that there is a reduction of the impact strength of each type of immersed specimen compared to the control specimen. This fact is due to the long immersion time so that the fibers absorb more liquid content after the immersion process and also result in higher fiber degradation. This is correlated to the study of Akil and co-workers[30] that the absorption of liquid after exposure to an aqueous environment causes a decline in the interfacial bond between the fibers and matrix.

The UPRs-cantala fibers specimens with alkaline and fumigation-treated fibers have lower impact strength compared to specimens with untreated fibers during the immersion process. It is due to the alkalization and fumigation treatment which make the fibers surface rough then enhance the fibers-matrix bonds. They also have lower impact strength than control specimens. This agrees with the study of Kabir and co-workers[37] which found that the lower impact strength revealed that the decreased energy absorbed by the fiber is caused by stronger interfacial bonds after alkaline treatment of the fibers. It also conforms with the research of Asfar and co-workers[38] in which a composites having strong interfacial bonds when get a sudden load will experience inelastic deformation which bring on direct loading across its cross-section without making a spread of loading. Meanwhile, in composites having weak interfacial bonds, a sudden load causes the spread of cracks which further enhanced the energy absorption capacity and ultimately raises the impact strength[39].

The influence of immersion with different fluids on the impact strength was also indicated in Fig. 5. It reveals that the largest value of impact strength is occurred in fresh water, followed by acidic solution, and sea water. This sequence is in compliance with the value of water absorption M_m. The impact test results in acidic solutions that are always in the middle are conformed with its diffusion coefficient (D) where after the immersion process in an acidic solution has a nearly neutral pH (around pH 7). This case may be due to dissolved lignin that may have resulted in alkaline ions in the solution, which leads to raise the pH value, as studied by Nosbi and co-workers[25]

3.5 Flexural strength and modulus

The flexural strength and modulus of UPRs-cantala fibers are indicated in Fig. 6. (a) and (b), respectively. The flexural strength of control specimens, wet and re-dried specimens of each different immersion fluid after 60 days are shown in Fig. 6. (a). It reveals that the value of flexural strength decreases in all immersing specimens compared to the control ones. Cantala fibers in the composites tend to absorb more water which causes faster degradation of the specimens, resulting in weak interfacial bonds between the fibers and matrix. Water molecules play role as plasticizing agents and reduce the structural rigidness

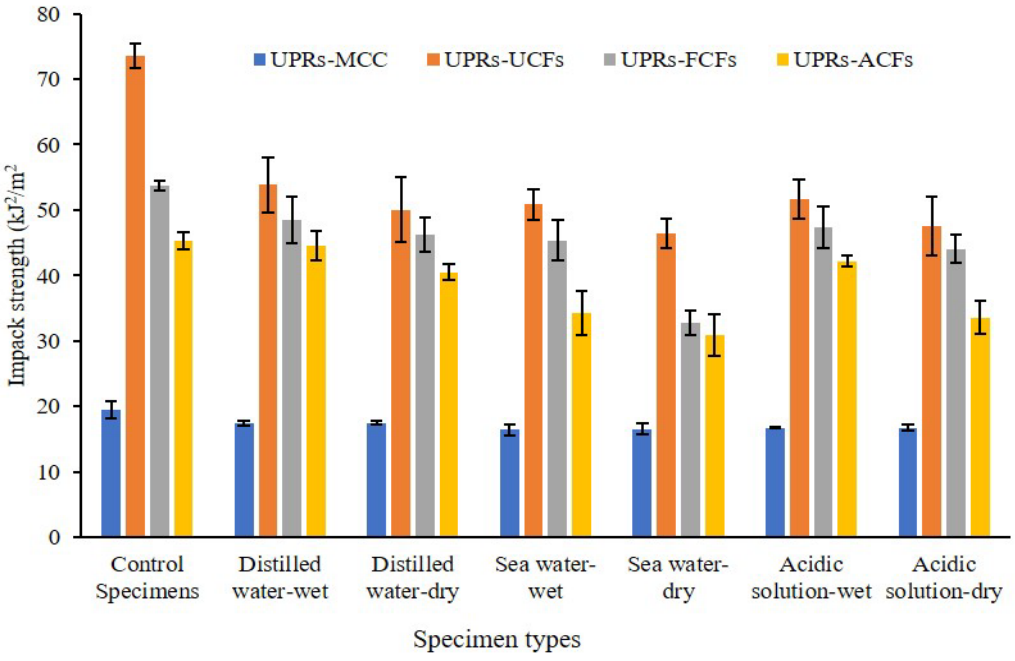


Fig. 5: Impact strength of composites before and after soaked in the different fluids

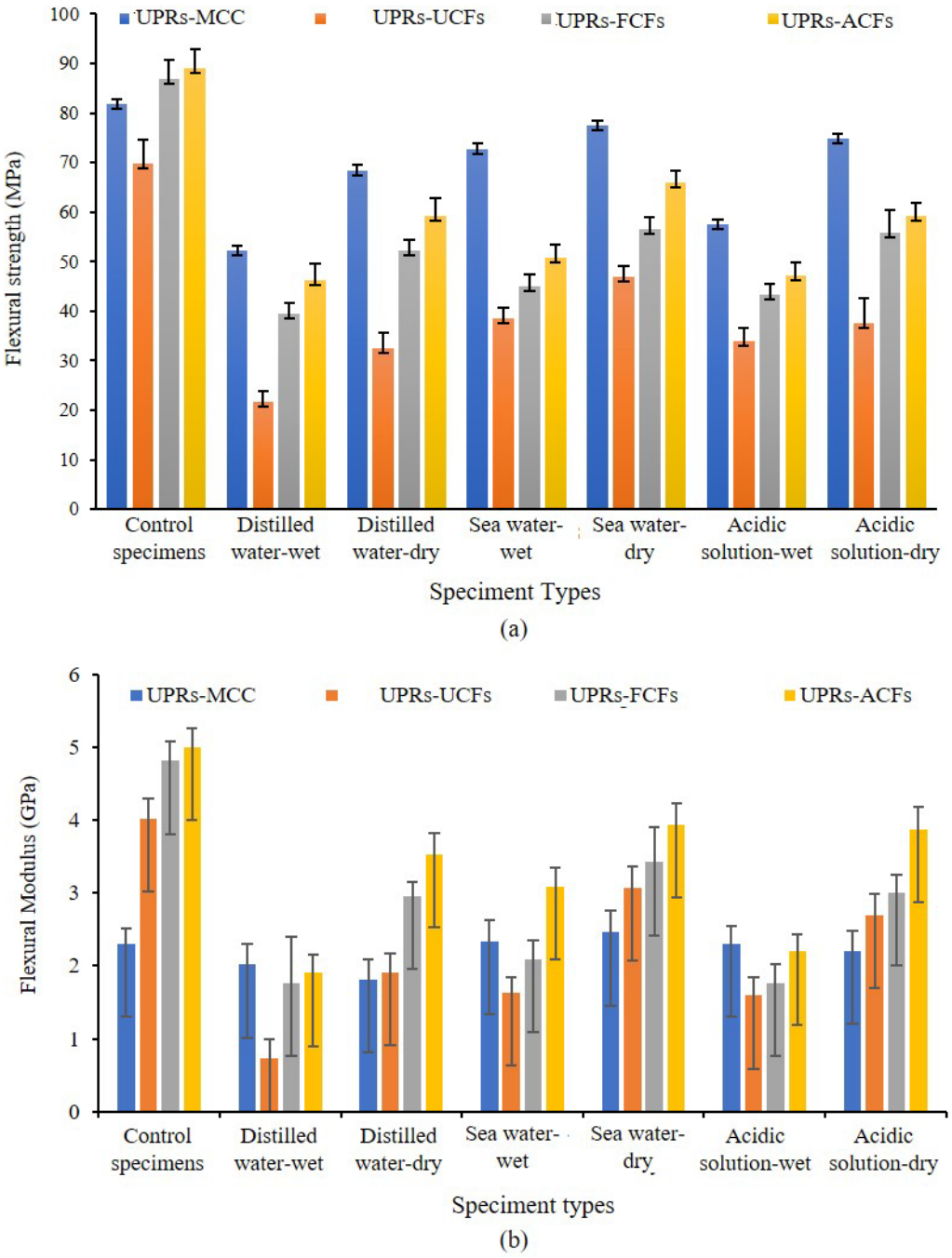


Fig. 6: (a) Flexural strength and (b) flexural modulus of composites before and after soaked in the different fluids

of cellulose, as studied by Dhakal and co-workers[40]. This tendency can be caused by the weak interfacial bond between the fiber and matrix caused by water absorption after immersion[30].

The flexural testing of immersed specimens indicates that specimens with sea water immersion has the highest flexural strength and modulus, followed by the specimens immersed in acidic solution and distilled water, respectively. These results are related to the magnitude of M_m and D in Table 1, which are higher and tend to provide higher degradation so that bring on decreasing flexural strength and elastic modulus. This is due to the water molecules acting as plastisizing agent in which sea water penetrates the composites in lowest quantity compared to acidic solution and distilled water. This case is correlated with study of Akil and co-workers[30]. Meanwhile, the specimens with re-drying process shows a higher flexural strength than the wet ones. Nonetheless, compared to control specimens, the re-dried specimens have lower flexural strength. Previous studies suggest that the re-drying process could annihilate the plasticizing effect of the elements in the composite without producing the effects of failure and microscopic crack growth as well as the interfacial area of the fibers and matrix[40].

The elastic modulus curve, shown in Fig. 6. (b), indicates that fumigation and alkali treatments can increase the elastic modulus in re-dried and wet specimens after immersion. This is owing to the removal of amorphous components such as hemicellulose, lignin, and impurities, thereby increasing the interfacial bonding between the matrix and fibers. In the study of Sakuri and co-workers[27], fumigation treatment can enhance fiber strength, fiber-matrix adhesion, and the tensile strength of composites. Alkaline treatment can raise the crystallinity of the natural fibers leading to an increase in tensile strength[41]. The alkaline treatment removes wax, dirt and hydroxyl groups that adhere to hemicellulose so that increases the surface area of fiber-matrix contact[34] and the fiber surface energy[42]. However, immersion in all fluids results in degradation of the UPRs-cantala fiber composites, as well as in specimens treated with fumigated and alkaline fibers, compared to the control specimen.

3.6 Morphological examination on composite surface

The surface morphology of composites of UPRs-MCC, UPRs-UCFs, UPRs-FCFs, and UPRs-ACFs, using SEM, are indicated in Fig. 7. (a), (b), (c) and (d), for the control conditions, respectively. Morphological surface examined using SEM are shown in Fig. 7. (a) for the UPRs-MCC specimen. There are no visible signs of damage or cracks on the specimen surface due to absorption of the liquid. This fact is suited to the study of of Najafi and co-workers[33] that the number of fibers used as matrix reinforcement is directly proportional to the ability of the composite to absorb fluid. Because UPRs-MCC specimens contain only UPRs and MCC, they cannot absorb more fluid.

In the control specimens of UPRs-cantala fibers indicated in Fig. 7. (b), (c), and (d), there are little gaps. More gaps between the fibers and matrix existing indicated in Fig. 8 (a), (b) and (c) for UPRs-cantala fibers composites after immersion in fresh water, sea water, and acidic solution, respectively. Gaps and cracks tend to be more due to fiber swelling occurring on the surface of the UPRs-cantala fiber composite without fiber treatment, followed with fiber fumigation and alkaline treatment, when compared to control specimens. During the immersion process, the cantala fibers absorb moisture and cause fiber swelling. It is possible to increase a large stress on the interface bonds and result in the breaking of the interfacial bonds between the matrix and fiber then cause cracks in the matrix adjacent to the fiber interface. The emergence of cracks on the specimen surface can affect fluid absorption, for fresh water, sea water, or acidic solution.

The gaps and cracks that occurred in the specimen caused by the release of the interfacial bonds between the matrix and fibers that originated from swelling of the fibers tended to be more common in specimens immersing in distilled water, as shown in Fig. 8 (a). This fact is related

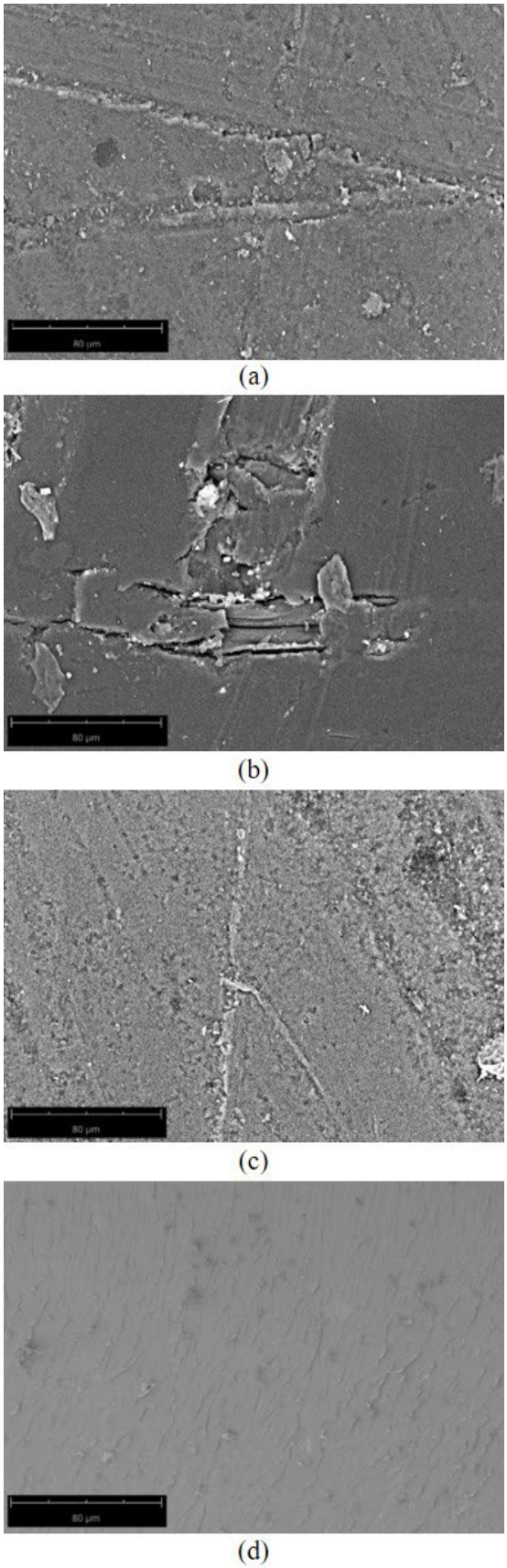


Fig.7: Surface of control specimens of: (a) UPRs-MCC composite; (b) UPRs-UCFs composite; (c) UPRs-FCFs composite; and (d) UPRs-ACFs composite

to the data in Table 1, the value of fluid absorption (% M_m) and the diffusion coefficient D in fresh water which has the highest value compared to other immersing fluids. In fresh water, the absorption process is high due to the high purity and uniformity of the water molecules so that it is easier to fit into the composite. This fact is supported by the study of Nosbi and co-workers[24], in accordance with the hydrolysis mechanism of cellulose in natural fibers.

The more effective it is to absorb the fluid, the more easily composite experience the swelling process of the fibers, thereby increasing the presence of cracks between the matrix and fibers. Swelling of the fibers results in radial stresses around the fibers and causes microscopic cracks near the interface area[35].

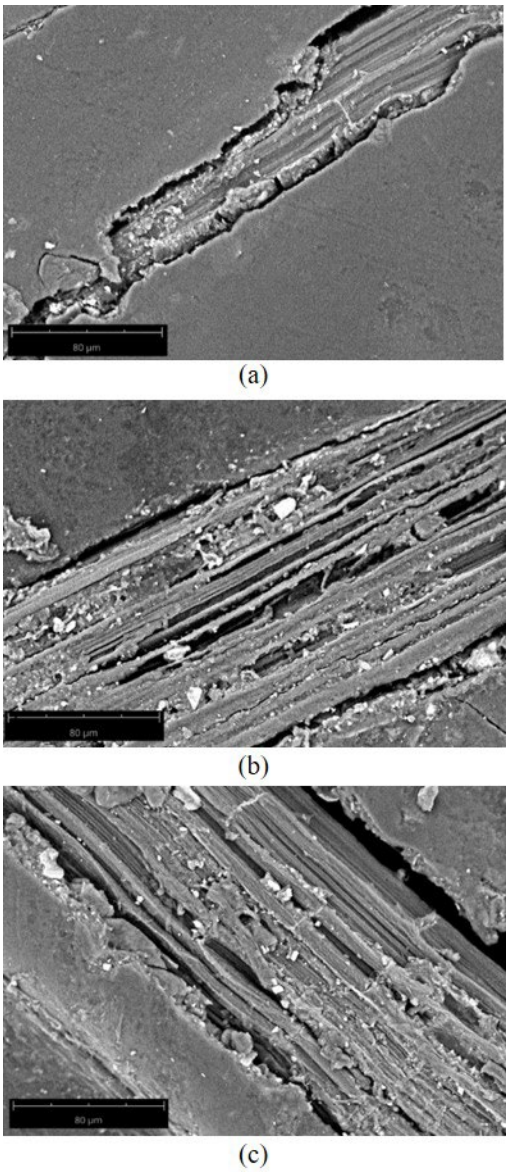


Fig. 8: Surface of UPRs-UCFs composite immersed in: (a) fresh water; (b) sea water; and (c) acidic solution

4. Conclusion

UPRs-cantala fibers composites with the addition of MCC increased in mass and dimension along with the duration of immersion process. They absorb fresh water the most highly compared to other immersion fluids with M_m and D average values of 14.61 % and 11.77 x 10^{-3} mm^2/s, respectively. UPRs–UCFs composites absorb fluid the most highly compared to other specimens in all types of immersion fluids. The fiber swelling occurs on the surface of UPRs-CFs composites with addition of MCC after immersion process caused by the fluid absorption. After immersion, UPRs-CFs composites experience a decrease in impact strength, and flexural strength and modulus when compared to control specimens. The wet specimens have better impact strength than the re-dried specimen. However, their flexural test results have lower than those of re-dried specimens. Fumigation and alkaline treatment on UPRs-CFs composites with addition of MCC reduce the water absorption rate and impact strength but increase the flexural strength, as well as the flexural modulus both in the control specimen and after the immersion process. The water absorption and average impact strength of the UPRs-FCFs fiber composite specimens are higher than specimens with alkaline-treated CFs. However, the flexural strength and modulus of UPRs-FCFs composite specimens are lower than specimens with alkaline-treated CFs.

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Nomenclature

UPRs-MCC	Unsaturated polyester and MCC composites
UPRs-UCFs	UPRs + MCC matriced composite reinforced by untreated cantala fibers
UPRs-FCFs	UPRs + MCC matriced composite reinforced by fumigated cantala fibers
UPRs-ACFs	UPRs + MCC matriced composite reinforced by alkalized cantala fibers
M_m	water absorption rate (%)
M_1	initial mass of the specimen (g)
M_2	final mass of the specimen (g)
D	diffusion coefficient
t_1	initial time (s)
t_2	final time (s)
h_{max}	maximum dimension change (%)
h_1	initial thickness of the specimens (mm)
h_2	final thickness of the specimens (mm)
S	slope of dimension change

References

1) W.D. Callister, and D.G. Rethwisch, "Materials science and engineering : an Introduction," 10th ed. New Jersey: John Wiley & Sons, Inc., 2018.
2) J. Holbery, and D. Houston, "Natural-fibre-reinforced polymer composites in automotive applications," *J. Miner.*, **58** (*11*) 80–86 (2006), doi: 10.1007/s11837-006-0234-2.
3) R.F. Gibson, "Principles of Composite Material," 4th ed. Boca Raton: CRC Press, 2016.
4) S. Pilla, "Engineering Applications of Bioplastics and Biocomposites - An Overview," in "Handbook of Bioplastics and Biocomposites Engineering Applications," 1st ed., S. Pilla, Ed. Scrivener & Wiley, 2011.
5) M. Soleimani, L. Tabil, S. Panigrahi, X. Li, and B. Crerar, "Compression Molding of Flax Fiber-Polypropylene Composites: Factors Affecting Selected Properties," in "CSBE 2006 Conference," 2006. doi: 10.13031/2013.22102.
6) M.H. Palmiyanto, E. Surojo, D. Ariawan, and F. Imaduddin, "E-glass/kenaf fibre reinforced thermoset composites fiiled with MCC and immersion in a different fluid," *Sci. Rep.*, **12** (2022). doi: /10.1038/s41598-022-24506-w.
7) A. Mahyudin, S. Arief, H. Abral, Emriadi, M. Muldarisnur, and M.P. Artika, "Mechanical Properties and Biodegradability of Areca Nut Fiber-reinforced Polymer Blend Composites," *Evergreen*, **7** (*3*), 366–372 (2020). doi: 10.5109/4068618.
8) S.P. Dwivedi, M. Maurya, N.K. Maurya, A.K. Srivastava, S. Sharma, and A. Saxena, "Utilization of Groundnut Shell as Reinforcement in Development of Aluminum Based Composite to Reduce Environment Pollution: a review," *Evergreen*, **7** (*1*) 15–25 (2020). doi: 10.5109/2740937.
9) S. Ray, "Effect of Control Parameters on Erosion Wear Performance of Glass-Epoxy Composites Filled with Waste Marble Powder," *Evergreen*, **9**, (*1*) 23–32 (2022). doi: 10.5109/4774213.
10) M.K. Gupta, V. Singhal, and N.S. Rajput, "Applications and Challenges of Carbon-fibres reinforced Composites: A Review," *Evergreen*, **9** (*3*) 682–693 (2022). doi: 10.5109/4843099.
11) M.B. Palungan, R. Soenoko, Y.S. Irawan, and A. Purnowidodo, "The Effect of Fumigation toward the Engagement Ability of King Pineapple Leaf Fibre (Agave cantala roxb) with Epoxy Matrix," *ARPN J. Eng. Appl. Sci.*, **11** (*13*) 8532–8537 (2016).
12) M.B. Palungan and R. Soenoko, "The Effect of Fumigation Treatment towards Agave Cantala Roxb Fibre Strength and Morphology," *J. Eng. Sci. Technol.*, **12** (*5*) 1399–1414 (2017).
13) M. Asim, M. Jawaid, K. Abdan, and M.R. Ishak, "Effect of Alkali and Silane Treatments on Mechanical and Fibre-matrix Bond Strength of Kenaf and Pineapple Leaf Fibres," *J. Bionic Eng.*, **13** (*3*) 426–435 (2016). doi: 10.1016/S1672-6529(16)60315-3.
14) S. Sangthong, T. Pongprayoon, and N. Yanumet, "Mechanical property improvement of unsaturated polyester composite reinforced with admicellar-treated sisal fibers," *Compos. Part A Appl. Sci. Manuf.*, **40** (*6–7*) 687–694 (2009). doi: 10.1016/j.compositesa.2008.12.004.
15) G. Goud and R.N. Rao, "Effect of fibre content and alkali treatment on mechanical properties of Roystonea regia-reinforced epoxy partially biodegradable composites," *Bull. Mater. Sci.*, **34** (*7*) 1575–1581 (2011). doi: 10.1007/s12034-011-0361-4.
16) W.P. Raharjo, R. Soenoko, A. Purnowidodo, and A. Choiron, "Characterization of Sodium-Bicarbonate-Treated Zalacca Fibers as Composite Reinforcements," *Evergreen*, **6** (*1*) 29–38 (2019). doi: 10.5109/2321001.
17) L.Y. Mwaikambo and M.P. Ansell, "Chemical modification of hemp, sisal, jute, and kapok fibers by alkalization," *J. Appl. Polym. Sci.*, **84** (*12*) 2222–2234 (2002). doi: 10.1002/app.10460.
18) Z.N. Azwa, B.F. Yousif, A.C. Manalo, and W. Karunasena, "A review on the degradability of polymeric composites based on natural fibres," *Mater. Des.*, **47** 424–442 (2013). doi: 10.1016/j.matdes.2012.11.025.
19) A. Gharbi, R.B. Hassen, and S. Boufi, "Composite materials from unsaturated polyester resin and olive nuts residue: The effect of silane treatment," *Ind. Crops Prod.*, **62** 491–498 (2014). doi: 10.1016/j.indcrop.2014.09.012.
20) L.K. Kian, M. Jawaid, H. Ariffin, and O.Y. Alothman, "Isolation and characterization of microcrystalline cellulose from roselle fibers," *Int. J. Biol. Macromol.*, **103** 931–940 (2017). doi: 10.1016/j.ijbiomac.2017.05.135.
21) B.Y. Alashwal, M.S. Bala, A. Gupta, S. Sharma, and P. Mishra, "Improved properties of keratin-based bioplastic film blended with microcrystalline cellulose: A comparative analysis," *J. King Saud Univ. - Sci.*, **32** (*1*) 853–857 (2020). doi: 10.1016/j.jksus.2019.03.006.
22) D. Trache, M.H. Hussin, C. Tan, H. Chuin, S. Sabar, M.R.N. Fazita, O.F.A Taiwo, T.M. Hassan, and M.K.M. Haafiz, "Microcrystalline cellulose: Isolation, characterization and bio-composites application — A review," *Int. J. Biol. Macromol.*, **93** 789–804 (2016). doi: 10.1016/j.ijbiomac.2016.09.056.
23) D.F. Smaradhana, D. Ariawan, and R. Alnursyah, "A Progress on Nanocellulose as Binders for Loose Natural Fibres," *Evergreen*, **7** (*3*) 436–443 (2020). doi: 10.5109/4068624.
24) N. Nosbi, H.M. Akil, Z.A. Mohd Ishak, and A. Abu Bakar, "Behavior of Kenaf Fiber After Immersion in Several Water Conditions," *Bioresources*, **1** (*2*) 950–

960 (2011). doi: 10.15376/BIORES.6.2.950-960.

25) N. Nosbi, H.M. Akil, Z.A. Mohd Ishak, and A. Abu Bakar, "Degradation of compressive properties of pultruded kenaf fiber reinforced composites after immersion in various solutions," *Mater. Des*., **31** (*10*) 4960–4964 (2010). doi: 10.1016/j.matdes.2010.04.037.
26) W.W. Raharjo, R. Soenoko, Y.S. Irawan, and A. Suprapto, "The Influence of Chemical Treatments on Cantala Fiber Properties and Interfacial Bonding of Cantala Fiber/Recycled High Density Polyethylene (rHDPE)," *J. Nat. Fibers*, **15** (*1*) 98–111 (2017). doi: 10.1080/15440478.2017.1321512.
27) S. Sakuri, E. Surojo, D. Ariawan, and A.R. Prabowo, "Experimental investigation on mechanical characteristics of composite reinforced cantala fiber (CF) subjected to microcrystalline cellulose and fumigation treatments," *Compos. Commun.*, **21** (*March*) 100419 (2020). doi: 10.1016/j.coco.2020.100419.
28) S. Sakuri, E. Surojo, and D. Ariawan, "Thermogravimetry and Interfacial Characterization of Alkaline Treated Cantala Fiber/Microcrystalline Cellulose-Composite," *Procedia Struct. Integr.*, **27** 85–92 (2020). doi: 10.1016/j.prostr.2020.07.012.
29) S. Sakuri, D. Ariawan, and E. Surojo, "Mechanical properties of microcrystalline cellulose filled unsaturated polyester blend composite with various treatment," in *AIP Conference Proceedings* **2097**, 2019. 1–6. doi: 10.1063/1.5098240.
30) H.M. Akil, L.W. Cheng, Z.A. Mohd Ishak, A. Abu Bakar, and M.A. Abd Rahman, "Water absorption study on pultruded jute fibre reinforced unsaturated polyester composites," *Compos. Sci. Technol.*, **69** (*11–12*) 1942–1948 (2009). doi: 10.1016/j.compscitech.2009.04.014.
31) K.B. Adhikary, S. Pang, and M.P. Staiger, "Dimensional stability and mechanical behaviour of wood-plastic composites based on recycled and virgin high-density polyethylene (HDPE)," *Compos. Part B Eng.*, **39** (*5*) 807–815 (2008). doi: 10.1016/j.compositesb.2007.10.005.
32) Z.A. Mohd Ishak, A. Ariffin, and R. Senawi, "Effects of hygrothermal aging and a silane coupling agent on the tensile properties of injection molded short glass fiber reinforced poly(butylene terephthalate) composites," *Eur. Polym. J.*, **37** (*8*) 1635–1647 (2001). doi: 10.1016/S0014-3057(01)00033-7.
33) S.K. Najafi, M. Tajvidi, and M. Chaharmahli, "Long-term water uptake behavior of lignocellulosic-high density polyethylene composites," *Journal of Applied Polymer Science*, **102** (*4*) 3907–3911 (2006). doi: 10.1002/app.24172.
34) D. Ariawan, Z.A.M. Ishak, M.S. Salim, R.M. Taib, M.Z.A. Thirmizir, and H. Pauzi, "The Effect of Alkalization on the Mechanical and Water Absorption Properties of Nonwoven Kenaf Fiber/ Unsaturated-Polyester Composites Produced by Resin-Transfer Molding," *Polym. Compos.*, **16** (*2*) 101–113 (2017). doi: 10.1002/pc.
35) P.A. Sreekumar, S.P. Thomas, J.M. Saiter, K. Joseph, G. Unnikrishnan, and S. Thomas, "Effect of fiber surface modification on the mechanical and water absorption characteristics of sisal/polyester composites fabricated by resin transfer molding," *Compos. Part A Appl. Sci. Manuf.*, **40** (*11*) 1777–1784 (2009). doi: 10.1016/j.compositesa.2009.08.013.
36) H. Alamri and I.M. Low, "Effect of water absorption on the mechanical properties of n-SiC filled recycled cellulose fibre reinforced epoxy eco-nanocomposites," *Polym. Test.*, **31** (*6*) 810–818 (2012). doi: 10.1016/j.polymertesting.2012.06.001.
37) M.M. Kabir, H.Wang, K.T. Lau, and F. Cardona, "Composites : Part B Tensile properties of chemically treated hemp fibres as reinforcement for composites," *Compos. Part B*, **53** 362–368 (2013). doi: 10.1016/j.compositesb.2013.05.048.
38) M.F. Asfar, D. Ariawan, and J. Triyono, "Effect of immersion on the impact properties of zalacca midrib fiber/rHDPE by compression molding," *AIP Conf. Proc.*, **2097** (*April*), 2019. doi: 10.1063/1.5098201.
39) D. Ray, B.K. Sarkar, A.K. Rana, and N.R. Bose, "Effect of alkali treated jute fibres on composite properties," *Bull. Mater. Sci.*, **24** (*2*) 129–135 (2001). doi: 10.1007/BF02710089.
40) H.N. Dhakal, Z.Y. Zhang, and M.O.W. Richardson, "Effect of water absorption on the mechanical properties of hemp fibre reinforced unsaturated polyester composites," *Compos. Sci. Technol*., **67**, (*7–8*) 1674–1683 (2007). doi: 10.1016/j.compscitech.2006.06.019.
41) D. Ariawan, M.S. Salim, R. Mat Taib, M.Z.A. Thirmizir, and Z.A.M. Ishak, "Durability of alkali and heat-treated kenaf fiber/unsaturated polyester composite fabricated by resin transfer molding under natural weathering exposure," *Adv. Polym. Technol.*, **37** 1420–1434 (2018). doi: 10.1002/adv.21801.
42) D. Ariawan, Z.A. Mohd Ishak, M.S. Salim, R. Mat Taib, and M.Z. Ahmad Thirmizir, "Wettability and interfacial characterization of alkaline treated kenaf fiber-unsaturated polyester composites fabricated by resin transfer molding," *Polymer Composites*, **38** (*3*) 507–515 (2015). doi: 10.1002/pc.23609.