Research Article

Mochamad Asrofi*, Rizki Setyobudi, R. A. Ilyas*, M. L. Sanyang, A. O. Adegbenjo, I. Idris, Senthil Muthu Kumar Thiagamani, C. D. Midhun Dominic, V. F. Knight, M. N. F. Norrrahim*, L. Rajeshkumar, and M. R. M. Asyraf

Influence of ultrasonication time on the various properties of alkaline-treated mango seed waste filler reinforced PVA biocomposite

https://doi.org/10.1515/rams-2023-0137
received April 25, 2023; accepted October 13, 2023

Abstract: The usage of biodegradable materials is gaining mounting applications owing to the environmental problems created by petroleum-based synthetic materials. Agro-waste materials are finding more scope as reinforcement materials in polymer composites, mainly because of their cost-effectiveness and availability. The main aim of this research work is to utilize agro-waste material in the form of mango seed waste (MSW) as a reinforcing material with polyvinyl alcohol (PVA) to form biocomposite. The biocomposites were fabricated using the solution casting technique. The research work focuses on the influence of varying ultrasonication periods (0–20 min at 5-min intervals, *i.e.*, 0, 5, 10, 15, and 20 min) on the properties of PVA/MSW biocomposites. The fabricated biocomposites were subjected to tensile test, fracture morphology, moisture absorption (MA), Fourier transform infrared spectroscopy, and thermogravimetric analysis. The results revealed that the material with the highest tensile strength was 3.95 MPa obtained with the 20-min ultrasonication sample. This value is 49% higher than that obtained in the sample without ultrasonication. The scanning electron microscopy analysis shows that the 20-min ultrasonication sample indicated a good distribution of MSW fillers in the PVA contrasted with the rest of the samples. The thermal stability of the samples treated with ultrasonication was found to be significantly higher than that of the untreated samples, whereas the MA value decreased with increasing ultrasonication time. It can be observed from the results that

*** Corresponding author: Mochamad Asrofi,** Department of Mechanical Engineering, University of Jember, Kampus Tegalboto, Jember 68121, East Java, Indonesia, e-mail: asrofi.teknik@unej.ac.id
*** Corresponding author: R. A. Ilyas,** Faculty of Chemical and Energy Engineering, Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia; Centre for Advanced Composite Materials (CACM), Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia; Institute of Tropical Forest and Forest Products, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia; Centre of Excellence for Biomass Utilization, Universiti Malaysia Perlis, 02600, Arau, Perlis, Malaysia, e-mail: ahmadilyas@utm.my
*** Corresponding author: M. N. F. Norrrahim,** Research Centre for Chemical Defence, Universiti Pertahanan Nasional Malaysia, Kem Perdana Sungai Besi, Kuala Lumpur, 57000, Malaysia, e-mail: faiz@upnm.edu.my
Rizki Setyobudi: Department of Mechanical Engineering, University of Jember, Kampus Tegalboto, Jember 68121, East Java, Indonesia, e-mail: rizkisetyo14@gmail.com
M. L. Sanyang: Directorate of Research and Consultancy, University of The Gambia, Post Office Road, P.O Box 3530, Kanifing, Gambia; Department of Environmental Sciences, School of Agriculture and Environmental Science, University of The Gambia, P.O Box 3530, Brikama Campus, West Coast Region, Gambia, e-mail: m.sanyang@utg.edu.gm
A. O. Adegbenjo: Department of Mechanical Engineering Science, University of Johannesburg, Johannesburg, South Africa; Mechanical Engineering Department, Adeseun Ogundoyin Polytechnic, Eruwa, Oyo State, Nigeria, e-mail: waleeleect@gmail.com
I. Idris: School of Chemical Engineering, College of Engineering, Universiti Teknologi MARA (UiTM), 40000, Shah Alam, Selangor, Malaysia, e-mail: iyliaidris@uitm.edu.my
Senthil Muthu Kumar Thiagamani: Department of Mechanical Engineering, Kalasalingam Academy of Research and Education, Srivilliputhur, India, e-mail: tsmkumar@klu.ac.in
C. D. Midhun Dominic: Department of Chemistry, Sacred Heart College (Autonomous), Thevara, Kochi, Kerala, India, e-mail: midhundominic@shcollege.ac.in
V. F. Knight: Research Centre for Chemical Defence, Universiti Pertahanan Nasional Malaysia, Kem Perdana Sungai Besi, Kuala Lumpur, 57000, Malaysia, e-mail: victor.feizal@upnm.edu.my
L. Rajeshkumar: Centre for Materials and Manufacturing Technology, KPR Institute of Engineering and Technology, Coimbatore, 641407, Tamilnadu, India; Department of Mechanical Engineering, KPR Institute of Engineering and Technology, Coimbatore, 641407, Tamilnadu, India, e-mail: lrkln27@gmail.com
M. R. M. Asyraf: Centre for Advanced Composite Materials (CACM), Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia; Engineering Design Research Group (EDRG), Faculty of Mechanical Engineering, Universiti Teknologi Malaysia, 81310, Johor Bahru, Johor, Malaysia

biocomposites can be a potential alternative to the synthetic materials used in packaging applications.

Keywords: biocomposite, polyvinyl alcohol, mango seed waste filler, ultrasonication effect, mechanical properties, thermal stability

1 Introduction

Plastics are commonly utilized in daily life in the form of polyolefins (polyethylene and polypropylene) because they are strong, lightweight, and stable, but they are not easily broken down by microorganisms in the environment, causing huge environmental problems [1,2]. Efforts are being made to reduce this problem through several approaches, such as recycling, plastic waste treatment, and the development of biodegradable plastic [3–6]. Accordingly, it was discovered that the focus on creating novel polymer systems and materials based on polymers that had an ecological advantage was expanding in the field of polymer science and technology. As a result, numerous studies have used polymer composites with the addition of natural fillers to improve the performance of the matrix and to be more environmentally friendly [7–12].

Biodegradable plastics are generally produced with renewable raw materials like polylactic acid, polyvinyl alcohol (PVA), and starch. PVA is soluble in water and falls under the category of synthetic polymers. The reasons for choosing PVA material are based on its nontoxic and water-soluble properties. Despite being synthetic, these polymers are known as biodegradable materials. Apart from being used in medical applications, PVA is also widely used in food packaging applications because it has a hydrogel characteristic that makes it suitable for the formation of bioplastic films [13–15].

However, PVA material alone is not sufficient for making biodegradable plastics. This is because mechanical characteristics are poor and moisture absorption (MA) is significant [16,17]. The addition of natural fiber can improve these weaknesses, as reported by previous studies [16,18–21]. Many researchers have attempted to infuse various waste fillers into polymer matrices to improve the performance characteristics of the ensuing biocomposites [22,23]. Waste fillers obtained from brewed coffee [24–26], coconut shell [27], tamarind nut [28–30], banana peels [31], rice starch [32], sapodilla seeds [33], fish scales [34], corn husk [35], *etc.* were utilized for this purpose.

One of the most widely grown fruits is the mango (*Mangifera indica* L.), which is native to tropical and subtropical areas, including India, Malaysia, Sri Lanka, and Thailand. Mango seeds are readily available as a byproduct of mango fruit consumption and can be obtained in large quantities. The mango seed is currently not used for any commercial applications; instead, it is thrown away as garbage, contributing to pollution [36]. Utilizing mango seed waste (MSW) as a reinforcing filler promotes sustainable practices by reusing agricultural waste material, reducing environmental impact, and contributing to waste management.

Hence, MSW filler powder was chosen as a suitable filler and reinforcement in PVA matrix-based biocomposite because it has enough cellulose content that is quite competitive among other natural fibers [37]. Unfortunately, the addition of natural fiber alone in the matrix is less effective at improving mechanical properties [38] because these fibers have a few downsides despite their many advantages, including poor wettability, incompatibility with some polymeric matrices, and significant MA [39–43]. Researchers recommended particular physical and chemical techniques for fiber surface modification to address this issue. Natural fiber composites frequently use alkali treatment, one of the easiest and most efficient surface modification procedures [26,44]. Furthermore, ultrasonication of the biocomposite materials helps to maximize their mechanical, thermal, and moisture resistance properties [45–47].

Previous researchers conducted a study on plastic-based yarn bean starch reinforced with water hyacinth fibers to evaluate the mechanical properties of the biocomposites by varying the duration of vibration in the ultrasonic bath during their fabrication. After ultrasonic treatment, they asserted that the mechanical attributes improved [48]. According to a comparable study, the gelatinization process' ultrasonication might disperse the matrix's fibers [49]. The main working principle of the ultrasound device is to provide sound waves through the water medium, resulting in the breakdown of fiber agglomeration in the matrix [48,49].

Several previous studies used a long time duration of ultrasonication, resulting in expensive production costs. Therefore, this research provides a new perspective on the use of short time duration of ultrasonication with low power ultrasonication to enhance the traits of environmentally friendly biocomposites plastic. The current work stems from the use of PVA and alkaline-treated MSW filler-based composites, which were characterized by tensile test, scanning electron microscopy (SEM), MA, Fourier transform infrared spectroscopy (FT-IR), thermogravimetric analysis (TGA) and differential thermal analysis.

2 Materials and methods

2.1 Materials used

MSW was procured locally from mango sellers in Tanjung Market, Jember, Indonesia. PVA (under Chuang Chun

Petrochemical merk) with a molecular weight and density of 30,000 and 1.080 $g \cdot cm^{-3}$, respectively, was obtained from UD. Aneka Kimia store Jember, Indonesia. Several chemicals for the filler extraction process, such as glycerin (content 99.7%; moisture 0.3%), sodium hydroxide (NaOH) with purity 98%, and sodium hypochlorite (NaClO), were purchased from the Makmur Sejati Chemical Shop, Jember, Indonesia. The distilled water was provided at the Laboratory of Material Testing, University of Jember.

2.2 Isolation of MSW powder fillers

MSW was washed under running water five times to remove dirt. It was dried in the sun for 3 days until a constant weight was obtained. It was crushed using a grinder for 10 min. MSW was obtained in the form of fine powder. Then, the MSW filler was treated by alkalization using a 10% NaOH solution at a temperature of 80°C and stirring constantly at 500 rpm for 2 h. The purpose was to remove lignin and waxy substances. Equation (1) can be used to calculate the quantity of removed contents as a result of the reaction between the MSW powder and alkali solution [20].

$$\text{Filler–OH} + \text{NaOH} \rightarrow \text{Filler–O–Na}^{+} + \text{H}_2\text{O}. \quad (1)$$

The MSW powder was then washed in water to obtain a pH neutral. Then, the alkalized MSW powder was dried in the sun for one day. The dried MSW fillers were prepared for the bleaching process. The aim of the bleaching process was to remove residual lignin and impurities in the filler, as evidenced by changing the color of the filler powders to a whiter color. The bleaching process was conducted by using a 5% NaClO solution at a temperature of 60°C and a speed of 500 rpm for a time period of 4 h. After the bleaching treatment, it was dried in a hot air oven for 60 min at a temperature of 40°C. Then, it was crushed for 15 min using a grinder to obtain a finer filler powder.

2.3 Fabrication of biocomposite

A flow chart depicting the fabrication of the biocomposites is presented in Figure 1. The fabrication of biocomposites in this study was done *via* the solution casting method by mixing 10 g of PVA and 3 ml of glycerin in 100 ml of distilled water. They were stirred and heated at 500 rpm and 70°C for 1 h, respectively. Then, the mixture was added by 1 g of MSW filler gradually at a temperature of 70°C and 700 rpm for 10 min until it became a homogeneous gel texture. Thereafter, it was poured into a rectangular glass mold (120 mm × 60 mm). Then, the ultrasonication treatment was applied to the biocomposites using BAKU BK-2400 (Guangzhou Hanker Electronics Technology, Guangdong, China) ultrasonic bath machine with various time

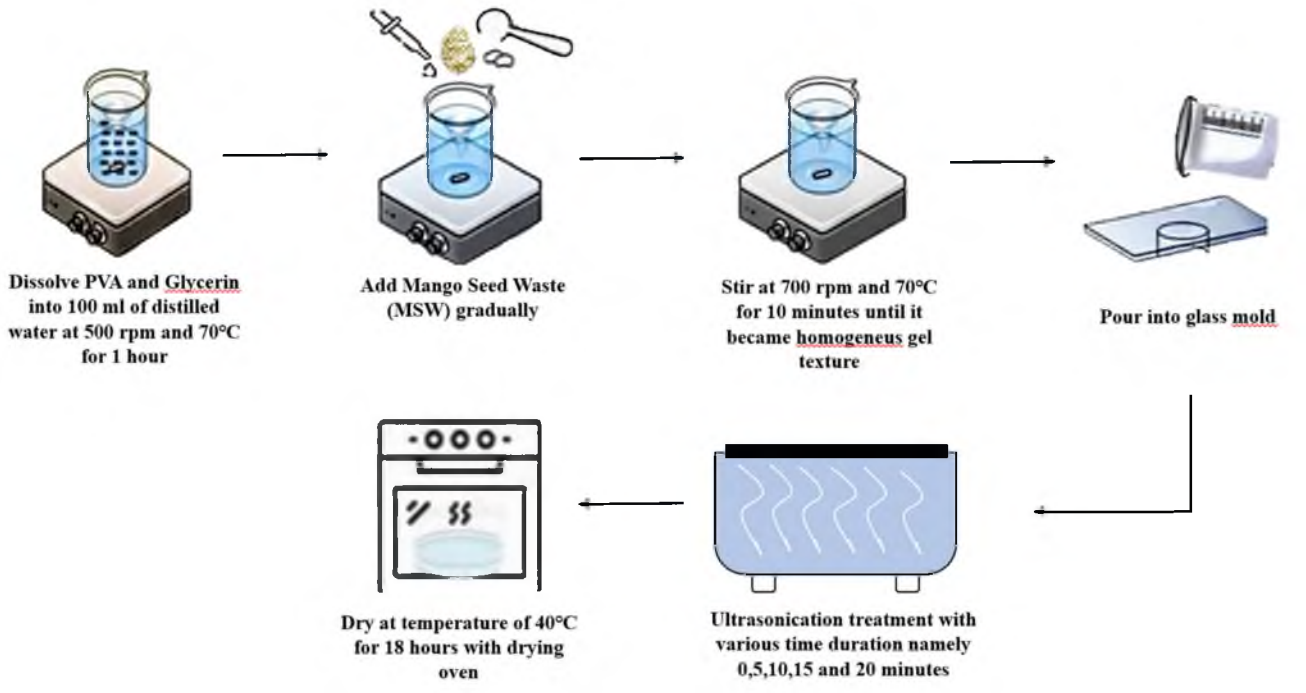

Figure 1: Schematic diagram of production biocomposite.

duration, namely 0, 5, 10, 15, and 20 min. In all the cases of ultrasonication, five replications were made. For the testing of mechanical properties in each case, five identical samples were tested, and the average values were recorded. After that, it dried at a temperature of 40°C for 18 h in a drying oven. Subsequently, the biocomposite was released from its mold and stored in a rectangular chamber with 45–55% relative humidity.

2.4 Characterization of biocomposites

2.4.1 Tensile test

Biocomposite tensile test specimens were manufactured and cut in accordance with the ASTM D882-02 standard. Thereafter, tensile tests were performed on the specimens using the HT-2328 Universal Testing Machine (Hung Ta Instrument Corporation, Thailand). For the testing of mechanical properties in each case, five number of identical samples were tested and the average values were recorded. The test was done at a temperature and tensile speed of 25°C and 10 mm·min^{-1}, respectively.

2.4.2 SEM

The morphological features of the biocomposites were observed using SEM analysis. TM3030Plus tabletop microscope (Hitachi High-Tech Corporation, Japan) machine model was used to examine the fracture morphology after a tensile test with an operation voltage of 15 kV. The test was conducted at room temperature. The magnification of the biocomposite fracture image was taken at 2,000×. SEM observations were carried out three times for each variation.

2.4.3 FT-IR

FT-IR spectrometer (Frontier, Perkin Elmer equipment, USA) was used to analyze the PVA and its biocomposite films' FT-IR spectra. The scanning was carried out in between 4,000 and 600 cm^{-1} wavelength at a resolution of 4 cm^{-1}.

2.4.4 Thermal analysis

This experiment followed a similar process to the one that was more fully detailed in a prior work [17]. The PVA and biocomposite films' thermal behavior, including TGA and derivative thermogravimety (DTG), was measured using the DTG-60 apparatus from Shimadzu in Japan. The heating rate of the samples was maintained at 20°C·min^{-1}, while the flow rate of nitrogen was kept at 50 mL·min^{-1}. The TGA curve of all biocomposite samples was measured using a thermal analysis instrument, with a nitrogen flow rate of 50 mL·min^{-1} and a heating rate of 10°C·min^{-1} within 25–475°C. The weight of each sample was between 5 and 10 mg.

2.4.5 MA

In accordance with the ASTM D570, the MA test was conducted [50]. Rectangular PVA and biocomposite film samples measuring 20 × 15 mm^2 area were dried in an oven (Model: KLAZ Universal Oven) till the weight was saturated. A closed chamber containing a saturated NaCl solution was utilized to achieve a relative humidity of 75%. After that, the dried samples were put in the enclosed chamber for 7 h at a temperature of 27°C, and they were weighed every 30 min using a digital balance (Keknko KK-LAB) with an accuracy of 0.001 g. The MA was calculated using Eq. (2) as follows [51–53]:

$$\mathrm{MA}(\%) = \frac{W_t - W_0}{W_0} \times 100\%, \tag{2}$$

where W_0 is the dry weight and W_t is the weight of biocomposite after test.

3 Results and discussions

3.1 Tensile strength

Three mechanical features of the tensile test findings were examined in this study: tensile strength, modulus of elasticity, and elongation (Figure 2). The maximum load received by the biocomposite was the maximum tensile strength that occurred during the test. In Figure 2c, the trendline for the lowest tensile strength was 2.32 MPa in the pure PVA sample. This tensile strength result of the biocomposites varies along with the variation in vibration duration during ultrasonication. The tensile strength of biocomposite with 0 min ultrasonication is 2.65 MPa, which is 14.22% higher than that of pure PVA film. A similar phenomenon occurs in biocomposite with 5 min ultrasonication for 2.61 MPa. This could be attributed to the porosity and voids present at the interface of matrix and mango

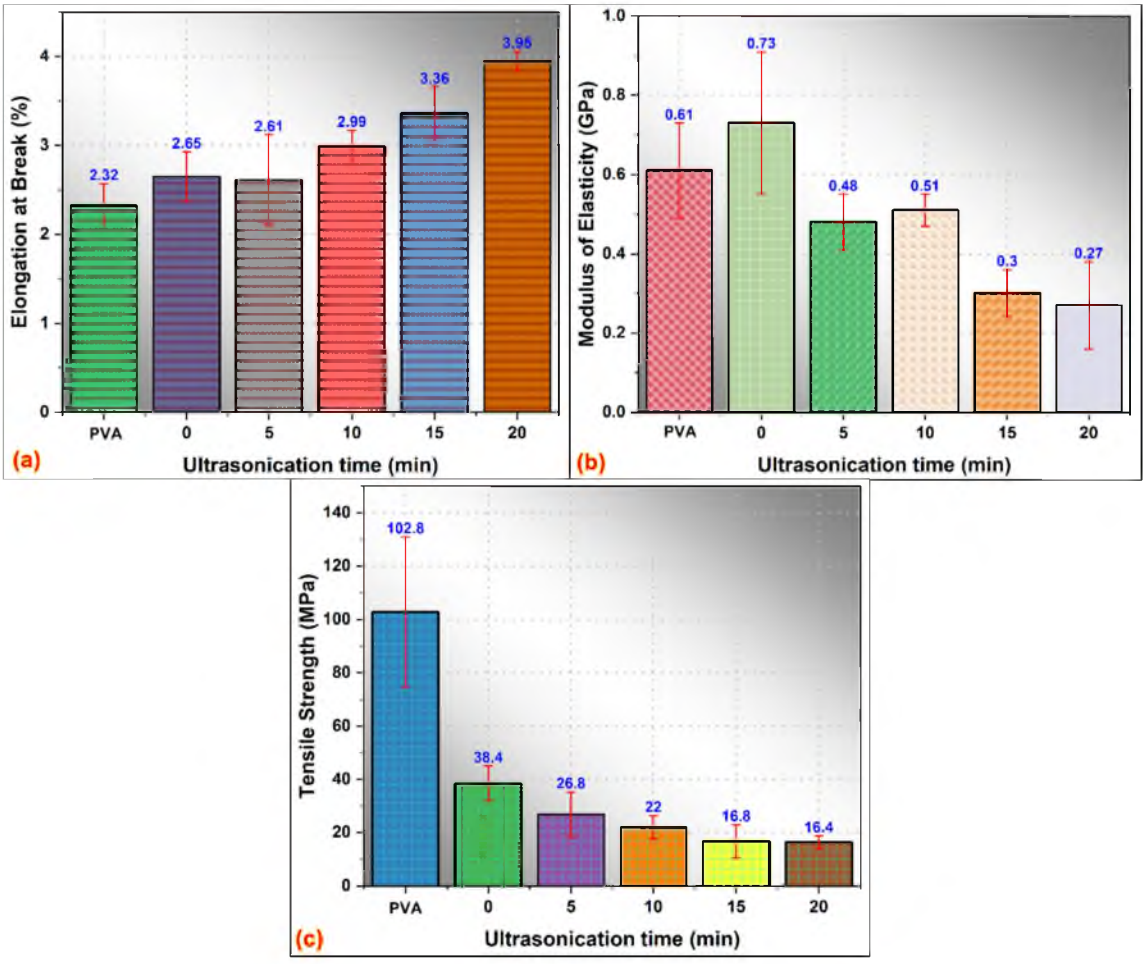

Figure 2: Tensile characteristics of biocomposite samples: (a) modulus of elasticity, (b) elongation at break, and (c) tensile strength.

seed filler reinforcement [14,54]. Poor performance results from porosity and gaps because the reinforcement's matrix and reinforcement cannot transmit loads effectively [55].

Biocomposite samples with 10, 15, and 20 min ultrasonication also showed an increase in the tensile strength of 2.99, 3.36, and 3.95 MPa, respectively. These results were significantly higher when compared to previous research conducted on biocomposites with water hyacinth fiber reinforcement exposed to different ultrasonication durations [48]. Tensile strength is growing due to the matrix and filler's strong adhesive bonding, lack of porosity, small gap, and compact structure [48,49]. This phenomenon was supported by SEM observation, which showed good interaction between matrix and filler (Figure 3d–f).

Unfortunately, the resultant modulus of elasticity did not rise along with the tensile strength. This is due to the difference in PVA hydrophilicity conditions before and after testing [18]. The highest modulus of elasticity (Figure 2a) was 0.73 MPa found in biocomposite 0 min ultrasonication. It indicates that this sample was inelastic (brittle). Meanwhile, the modulus of elasticity of the 20-min ultrasonication sample was 0.71 MPa, which means that this bioplastic property was also brittle. The brittle nature was influenced by the ultrasonication effect, which produces a compact structure [45,48].

The percentage value of elongation at break became higher as the ultrasonication duration increased (Figure 2b). It was indicated in another study that the addition of the ultrasonication effect causes the change in the fracture length of the biocomposite sample to be short [54]. As a result, the biocomposite sample becomes more brittle than the pure PVA film. This is in line with the results of previous studies [54,56].

3.2 SEM analysis

The SEM observations on the fracture surface of all biocomposite tests can be seen in Figure 3. Figure 3a depicts the fractured surface of a pure PVA film after the tensile

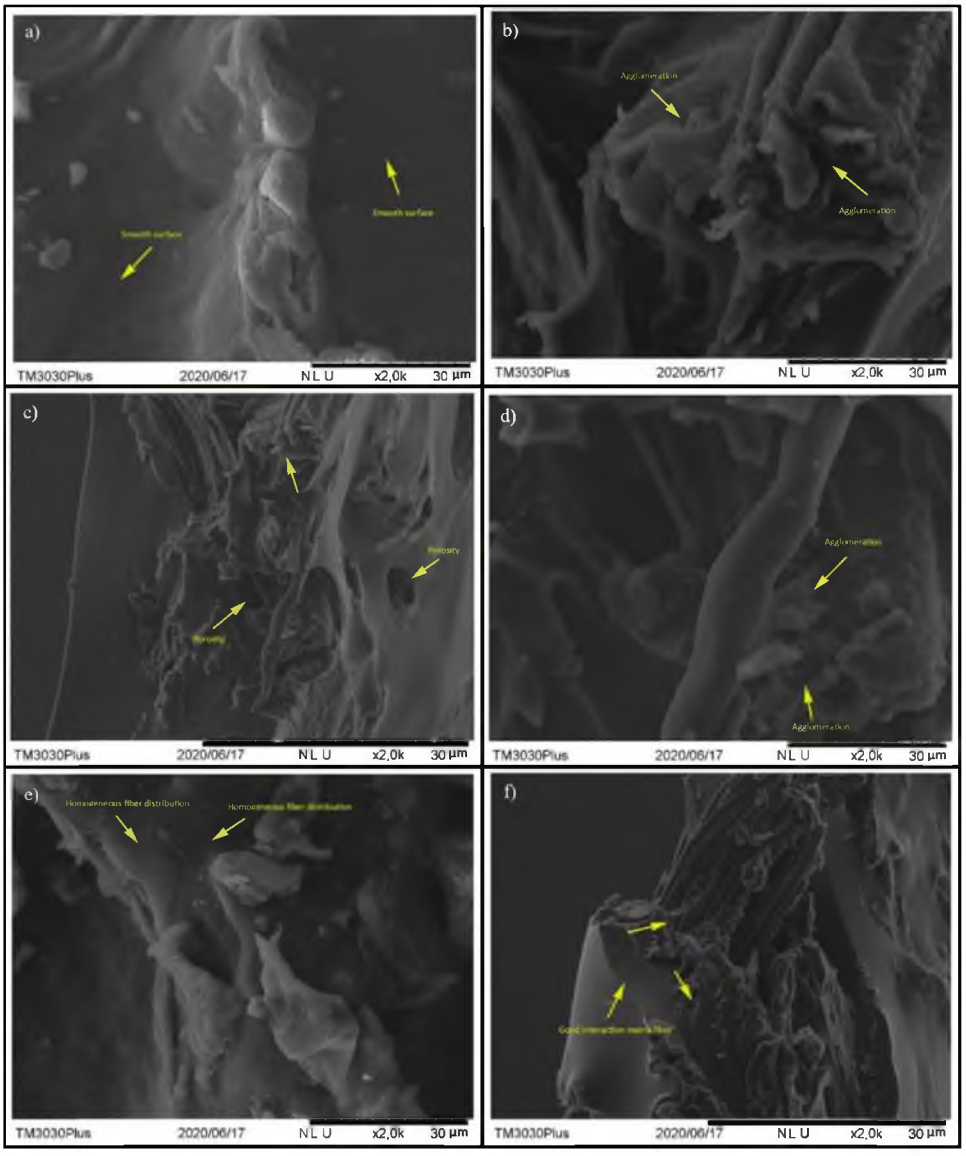

Figure 3: Fracture morphology of biocomposite by SEM observation: (a) PVA, morphology after a time duration of ultrasonication in minutes; (b) 0 min; (c) 5 min; (d) 10 min; (e) 15 min; and (f) 20 min.

test. It can be seen in the figure that the fracture of this film shows a smooth surface. This is consistent with previous studies, which showed that PVA without filler added had fine fractures [57]. Another appearance has been shown in Figure 3b and c, namely 0 and 5 min ultrasonication. In this figure, biocomposite fractures show agglomeration and porosity at several points. The biocomposite's tensile strength is reduced as a result. This demonstrates that adding ultrasonication for 0 or 5 min, respectively, is insufficient to dissolve filler clumping in the PVA matrix. A similar scene is demonstrated in Figure 3d (10 min ultrasonication). In this biocomposite fracture, the fillers were not evenly distributed within the PVA matrix. Previous studies also reported the same phenomenon, according to which agglomeration, voids, unevenly distributed fillers, and the mechanical characteristics of biocomposites were decreased because of gaps between the filler and matrix [45,48].

Meanwhile, at 15 and 20 min ultrasonication, Figure 3e and f displays multiple images on the biocomposite's fracture surface. The biocomposite samples with 15 and 20 min of ultrasonication displayed acceptable structure, homogeneous filler distribution, and strong matrix–filler bonding. [49]. This is presumably because the ultrasonic bath's kinetic energy improves the link between the mango seed filler and PVA matrix, which causes the uniform dispersion of the filler throughout the PVA matrix and dissolves filler clusters [48,58].

This observation is supported by previous studies showing that the addition of ultrasonication during the gelatin phase of the sample helps to spread evenly in the matrix. The biocomposite's tensile strength therefore increases [58].

3.3 MA

In general, natural fiber composites are moisture-prone [39,51], and this PVA-based biocomposite is not an exception. Whereas, one of the factors to consider in selecting a packaging material is its moisture resistance [16,18,59–61]. Therefore, in this study, PVA was combined with natural fibers to overcome this problem. Figure 4 illustrates how ultrasonication time affects how much moisture PVA and MSW-based biocomposites absorb. The pure PVA film has a maximum MA value of 19.1% after 7 h when compared to the biocomposite film. This value is the highest compared to biocomposites. This is due to the hydrophilic nature of PVA, as reported by previous researchers [57,62–65]. Different phenomena were shown by biocomposites with ultrasonication of 0, 5, 10, 15, and 20 min. At the 7th hour, the MA values of the biocomposite samples were 17.2, 15.8, 13.3, 12.8, and 11.2%, respectively. From the 1st hour until the 7th hour, conditions showed an increasing trend of MA in each biocomposite sample. This indicates that water or moisture molecules were able to penetrate into the biocomposite system through the surface. This case is similar to that reported by previous studies [57,58].

The lowest MA value was found in the biocomposite sample with ultrasonication for 20 min (11.2%). In general, the addition of ultrasonication helps to spread the MSW fillers evenly within the PVA matrix [45]. In addition, the ultrasonic kinetic energy of the ultrasonic bath was able to improve the bond between the matrix and the filler. This can be supported by the SEM imaging in Figure 3e and f. The good adhesion bonding between matrix and filler prevents moisture from entering the biocomposite system through the surface [58]. This phenomenon is similar to previous studies that reported that strong hydrogen bonding interactions between matrix and filler will reduce the number of free OH groups, causing a decrease in MA [49].

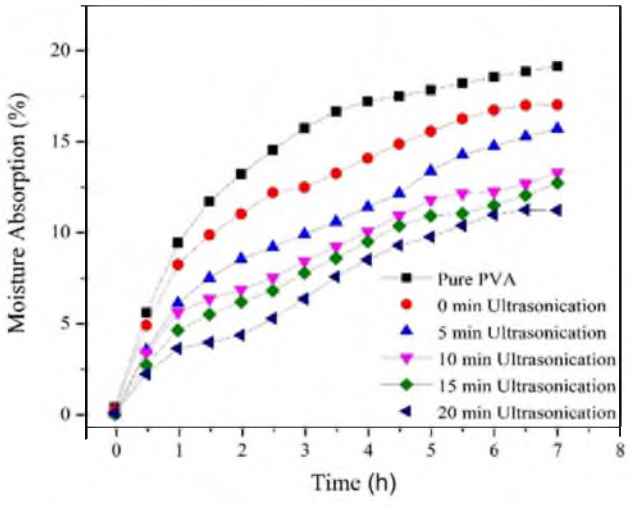

Figure 4: MA of all biocomposite samples.

3.4 Thermal analysis

Figure 5a and b shows the TGA and DTG curves for each pure PVA film and its biocomposite film with a variation of the ultrasonication for 0, 5, 10, 15, and 20 min. Figure 5a reveals that both the PVA film and biocomposite film undergo three stages of decomposition, as determined by TGA analysis. The initial decomposition of pure PVA takes place at a temperature range of 75–110°C, resulting in a mass loss of 0.06%/min due to the release of water molecules associated with the polymer [20].

Between 250 and 350°C, the most significant degradation took place, resulting in a dramatic weight loss of approximately 1.5%/min due to the breakdown of both the PVA and MSW fiber structures [66]. In this temperature range, the matrix and filler structures are damaged by high temperatures. Further decomposition and degradation occurred between 350 and 475°C, resulting in a mass loss of 0.15%/min, with the final product being ash [66,67].

Meanwhile, the DTG curve (Figure 5b) clarifies the temperature point shift from the TGA curve. In the DTG curve, the addition of ultrasonication duration shows a temperature shift towards a higher direction. In the temperature range of 250–350°C, the biocomposite with ultrasonication of 20 min has a temperature point of 311°C. In comparison to other biocomposites at 0 min (266°C), 5 min (293°C), 10 min (302°C), and 15 min (306°C), the temperature yield was greater (253°C). The higher thermal stability of MSW fillers increased the decomposition temperature compared to pure PVA [14,66]. The biocomposite film's thermal stability increased by 53°C when the ultrasonication time was extended to 20 min (compared to pure PVA), and a reduced mass loss of 1.5%/min as depicted in the graph with the maximum residual content. The reason for this trend could be due to the dissolvation of agglomerates by the ultrasonication process, where a more uniform distribution of the filler particles took place. The better interfacial

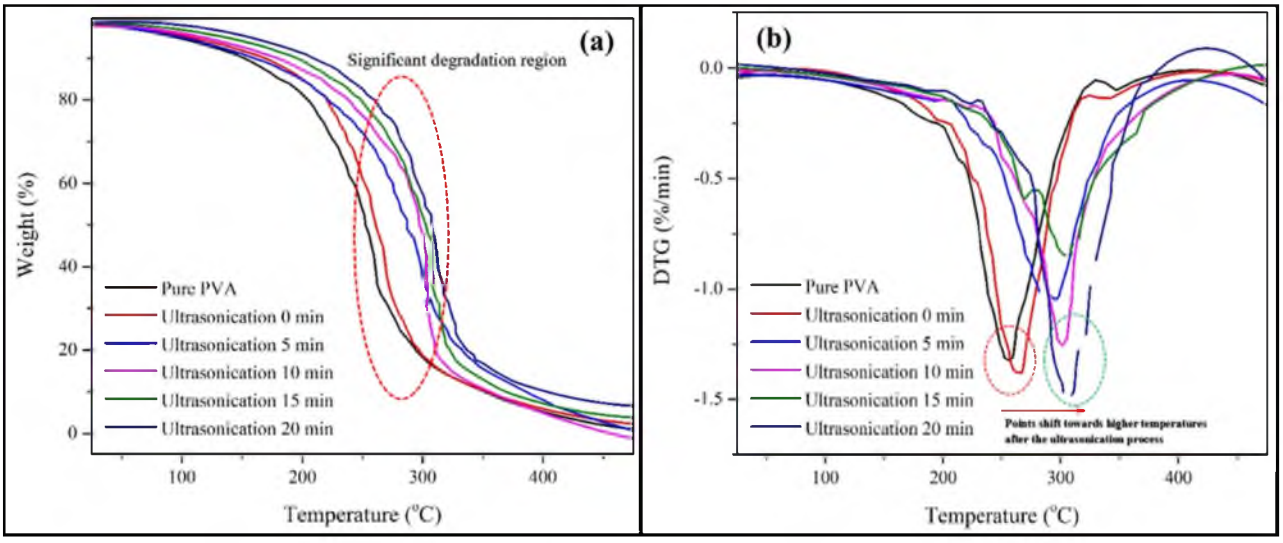

Figure 5: Thermal stability of mango seed fiber/PVA biocomposite samples: (a) TGA and (b) DTG.

connection between the filler and matrix as a result of improved dispersion can increase thermal stability. The resistance of the composite to heat degradation can be increased by preventing the onset and spread of microcracks or voids through a strong interfacial connection.

Due to its high decomposition temperature, MSW filler has greater thermal stability than PVA, which is attributed to the presence of amide groups. Moreover, the strong intermolecular hydrogen bonds formed between the hydroxyl groups of PVA and the amide groups of MSW fillers in the biocomposite contribute to increased thermal stability and mechanical strength [14]. The final residue produced by the biocomposite after thermal analysis is charcoal [68]. Based on this, it can be inferred that the reduction in mass loss observed with increasing ultrasonication duration is primarily due to the thermal stability properties of the MSW fillers present in the biocomposite [14].

3.5 FT-IR

Figure 6 shows the functional group of pure PVA and its biocomposite observed by FT-IR. The spectrum peak obtained for all samples tested at 3,290 cm^{-1} is for the OH and H groups, which are the bonds between the hydroxyl groups [57]. The C–H bond stretching in methyl and methylene for hemicellulose and cellulose groups is shown at 2,855–2,950 cm^{-1} peaks [69]. Furthermore, the peak at 1,640 cm^{-1} is due to the strain for the C=O bond [68].

Due to the hydroxyl group being stretched, the FT-IR spectrum exhibits a significant absorption peak at 1,640 and 3,290 cm^{-1} (PVA) [57]. On the other hand, the FT-IR spectrum of the fiber shows a distinctive amide-I band at 1,640 cm^{-1} and a wide peak at 3,290 cm^{-1} according to the O–H hydroxyl stretch [69].

For all PVA/MSW filler biocomposite with ultrasonication, the C=O and O–H stretching appeared broader and more intense than pure PVA. This widening is mainly due to the strong hydrogen bonds that occur between the hydroxyl and –N–H or C=O groups of the PVA and the MSW fillers [14]. No prominent shifts in the FT-IR peaks were observed, confirming that there was no functional group change in the biocomposite film.

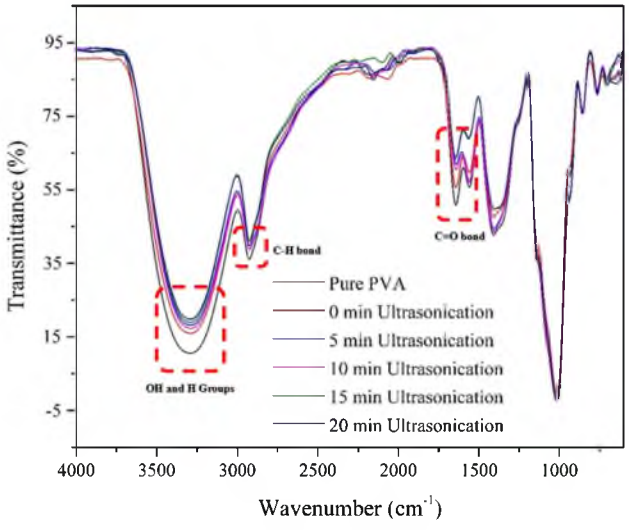

Figure 6: FT-IR spectra of all biocomposite samples.

4 Conclusion

The development of a PVA-based biocomposite with MSW filler has been successfully made. The tensile properties of the biocomposites varied with the variation in the duration of the ultrasonication. The tensile strength of biocomposite without ultrasonication was found to be 2.65 MPa. However, the biocomposite with 5-min ultrasonication exhibited a strength of 2.61 MPa, which is lower than the composites without ultrasonication. This could be attributed to the porosity and voids present at the interface of matrix and mango seed filler reinforcement. Poor performance results from porosity and gaps because the reinforcement's matrix and reinforcement cannot transmit loads effectively. On the other hand, a biocomposite sample with a 20-min ultrasonication had the greatest tensile strength result (3.95 MPa). This value was 170% higher than the pure PVA sample. SEM observations also showed a good structure, followed by an even distribution of fibers in the matrix. Thermal resistance increased significantly by 53°C compared to pure PVA film. The addition of ultrasonication duration has also succeeded in increasing the resistance to moisture. The findings in the research are results that are quite superior around 25–30% in terms of tensile strength compared to packaging products made from polypropylene (film grade). PVA-based biocomposite and MSW powder can be recommended as potential economic and environmentally friendly food packaging materials that can serve as a substitute for synthetic plastic.

Acknowledgments: The authors would like to thank Dr. Patrick Ehi Imoisili for his contribution to reviewing this manuscript.

Funding information: This research was funded and supported by the Institute of Research and Community Service, University of Jember, for reworking the thesis scheme with project number: 3018/UN25.3.1/LT/2021. We sincerely thank the Centre for Research Management and Innovation, Universiti Pertahanan Nasional Malaysia, for the financial support. The authors would like to thank the Universiti Teknologi Malaysia and the Ministry of Education, Malaysia, for their financial support. The authors would like to express gratitude for the financial support received from the Universiti Teknologi Malaysia for the project "The impact of Malaysian bamboos' chemical and fibre characteristics on their pulp and paper properties, grant number PY/2022/02318 – Q. J130000.3851.21H99". The research has been carried out under the program Research Excellence Consortium (JPT (BPKI) 1000/016/018/25 (57)) provided by the Ministry of Higher Education, Malaysia.

Author contributions: Conceptualization: Mochamad Asrofi; data curation: Mochamad Asrofi and L. Rajeshkumar; formal analysis: Mochamad Asrofi, Rizki Setyobudi, Senthil Muthu Thiagamani, and C. D. Midhun Dominic; funding acquisition: M. N. F. Norrrahim and V. F. Knight; investigation: Mochamad Asrofi, Rizki Setyobudi, R. A. Ilyas, M. L. Sanyang, A. O. Adegbenjo, Iylia Idris, and Senthil Muthu Thiagamani; methodology: Mochamad Asrofi and Rizki Setyobudi; project administration: Mochamad Asrofi and R. A. Ilyas; validation: Mochamad Asrofi, R. A. Ilyas, M. L. Sanyang, A. O. Adegbenjo, Iylia Idris, Senthil Muthu Thiagamani, C. D. Midhun Dominic, Victor Feizal Knight, and M. R. M. Asyraf; writing – original draft: Mochamad Asrofi, Rizki Setyobudi, and L. Rajeshkumar; writing – review & editing: R. A. Ilyas, M. L. Sanyang, A. O. Adegbenjo, Iylia Idris, Senthil Muthu Thiagamani, C. D. Midhun Dominic, M. R. M. Asyraf, M. N. F. Norrrahim, V. F. Knight, and L. Rajeshkumar. All authors have accepted responsibility for the entire content of this manuscript and approved its submission.

Conflict of interest: The authors state no conflict of interest.

References

[1] Agboola, O., R. Sadiku, T. Mokrani, I. Amer, and O. Imoru. 4 – Polyolefins and the environment. In *Polyolefin fibres*, 2nd edn, Ugbolue, S. C. O., ed, Woodhead Publishing, USA, 2017, pp. 89–133. https://www.sciencedirect.com/science/article/pii/B978008101132400047.

[2] Rajeshkumar, L., P. S. Kumar, M. Ramesh, M. R. Sanjay, and S. Siengchin. Assessment of biodegradation of lignocellulosic fiber-based composites – A systematic review. *International Journal of Biological Macromolecules*, Vol. 253, 2023, id. 127237. https://linkinghub.elsevier.com/retrieve/pii/S014181302304134X.

[3] Ilyas, R. A., S. M. Sapuan, R. Ibrahim, M. S. N. Atikah, M. R. M. Asyraf, M. N. F. Norrrahim, et al. Environmental advantages and challenges of bio-based packaging materials. In *Bio-based packaging: Material, environmental and economic aspects*, Wiley, Germany, 2021, pp. 371–380.

[4] Norrrahim, M. N. F., H. Ariffin, M. A. Hassan, N. A. Ibrahim, and H. Nishida. Performance evaluation and chemical recyclability of a polyethylene/poly(3-hydroxybutyrate-co-3-hydroxyvalerate) blend for sustainable packaging. *RSC Advances*, Vol. 3, No. 46, 2013, pp. 24378–24388.

[5] Ariffin, H., M. N. F. Norrrahim, T. A. T. Yasim-Anuar, H. Nishida, M. A. Hassan, N. A. Ibrahim, et al. Oil palm biomass cellulose-fabricated polylactic acid composites for packaging applications. In *Bionanocomposites for packaging applications*, Springer, Cham, 2018, pp. 95–105.

[6] Thiagamani, S. M. K., S. Krishnasamy, and S. Siengchin. Challenges of biodegradable polymers: An environmental perspective. *Applied Science and Engineering Progress*, Vol. 12, No. 3, 2019, id. 149.

[7] Dhandapani, A., S. Krishnasamy, T. Ungtrakul, S. M. K. Thiagamani, R. Nagarajan, C. Muthukumar, et al. Desirability of tribo-performance of natural based thermoset and thermoplastic composites: A concise review. *Applied Science and Engineering Progress*, Vol. 14, No. 4, 2021, pp. 606–613.

[8] Gulihonenahali Rajkumar, A., M. Hemath, B. Kurki Nagaraja, S. Neerakallu, S. M. K. Thiagamani, and M. Asrofi. An artificial neural network prediction on physical, mechanical, and thermal characteristics of giant reed fiber reinforced polyethylene terephthalate composite. *Journal of Industrial Textiles*, Vol. 51(1_suppl), 2022, pp. 769S–803S.

[9] Senthil Muthu Kumar, T., K. Senthilkumar, M. Chandrasekar, S. Karthikeyan, N. Ayrilmis, N. Rajini, et al. Mechanical, thermal, tribological, and dielectric properties of biobased composites. In *Biobased composites: Processing, characterization, properties, and applications*, Wiley, New Jersey, 2021, pp. 53–73.

[10] Dhandapani, A. Desirability of tribo-performance of natural based thermoset and thermoplastic composites: A concise review, *Applied Science and Engineering Progress*, Vol 14, No. 4, 2021, pp. 606–613.

[11] Senthilkumar, K., T. Ungtrakul, M. Chandrasekar, T. Senthil Muthu Kumar, N. Rajini, S. Siengchin, et al. Performance of sisal/hemp bio-based epoxy composites under accelerated weathering. *Journal of Polymers and the Environment*, Vol. 29, No. 2, 2021, pp. 624–636.

[12] Akhil, U. V., N. Radhika, B. Saleh, S. Aravind Krishna, N. Noble, and L. Rajeshkumar. A comprehensive review on plant-based natural fiber reinforced polymer composites: Fabrication, properties, and applications. *Polymer Composites*, Vol. 44, No. 5, 2023, pp. 2598–2633.

[13] Ilyas, R. A., S. M. Sapuan, M. M. Harussani, M. Y. A. Y. Hakimi, M. Z. M. Haziq, M. S. N. Atikah, et al. Polylactic acid (PLA) biocomposite: Processing, additive manufacturing and advanced applications. *Polymers (Basel)*, Vol. 13, No. 8, 2021 Apr, id. 1326.

[14] Abral, H., A. Hartono, F. Hafizulhaq, D. Handayani, E. Sugiarti, and O. Pradipta. Characterization of PVA/cassava starch biocomposites fabricated with and without sonication using bacterial cellulose fiber loadings. *Carbohydrate Polymers*, Vol. 206, 2019, pp. 593–601. https://www.sciencedirect.com/science/article/pii/S0144861718313882.

[15] Syafri, E., Sudirman, Mashadi, E. Yulianti, Deswita, M. Asrofi, et al. Effect of sonication time on the thermal stability, moisture absorption, and biodegradation of water hyacinth (*Eichhornia crassipes*) nanocellulose-filled bengkuang (*Pachyrhizus erosus*) starch biocomposites. *Journal of Materials Research and Technology*, Vol. 8, No. 6, 2019 Nov, pp. 6223–6231.

[16] Asrofi, M., D. Dwilaksana, H. Abral, and R. Fajrul. Tensile, thermal and moisture absorption properties of polyvinyl alcohol (PVA)/bengkuang (*Pachyrhizus erosus*) starch blend films. *Material Science Research India*, Vol. 16, No. 1, 2019 Apr, pp. 70–75.

[17] Rajeshkumar, L. Biodegradable polymer blends and composites from renewable resources. In *Biodegradable polymers, blends and composites*, Mavinkere Rangappa, S., J. Parameswaranpillai, S. Siengchin, and M. Ramesh, eds, Elsevier, UK, 2022, pp. 527–549. https://www.sciencedirect.com/science/article/pii/B9780128237915000156.

[18] Sukudom, N., P. Jariyasakoolroj, L. Jarupan, and K. Tansin. Mechanical, thermal, and biodegradation behaviors of poly (vinyl alcohol) biocomposite with reinforcement of oil palm frond fiber. *Journal of Material Cycles and Waste Management*, Vol. 21, No. 1, 2019, pp. 125–133.

[19] Senthil Muthu Kumar, T., N. Rajini, A. Alavudeen, S. Siengchin, V. Rajulu, and N. Ayrilmis. Development and analysis of completely biodegradable cellulose/banana peel powder composite films. *Journal of Natural Fibers*, Vol. 18, No. 1, 2019, pp. 151–160. https://www.tandfonline.com/doi/full/10.1080/15440478.2019.1612811.

[20] Ramalingam, K., S. M. K. Thiagamani, H. Pulikkalparambil, C. Muthukumar, S. Krishnasamy, S. Siengchin, et al. Novel cellulosic natural fibers from *Abelmoschus ficulneus* weed: Extraction and characterization for potential application in polymer composites. *Journal of Polymers and the Environment*, Vol. 31, 2023, pp. 1323–1334.

[21] Shahroze, R. M., M. Chandrasekar, K. Senthilkumar, T. S. M. Kumar, M. R. Ishak, N. Rajini, et al. Mechanical, interfacial and thermal properties of silica aerogel-infused flax/epoxy composites. *International Polymer Processing*, Vol. 36, No. 1, 2021, pp. 53–59.

[22] Kumar, T. S. M., K. Senthilkumar, M. Chandrasekar, S. Subramaniam, S. M. Rangappa, S. Siengchin, et al. Influence of fillers on the thermal and mechanical properties of biocomposites: An overview. In *Biofibers and biopolymers for biocomposites*, Springer, Cham, 2020, pp. 111–133.

[23] Senthil Muthu Kumar, T., N. Rajini, K. Obi Reddy, A. Varada Rajulu, S. Siengchin, and N. Ayrilmis. All-cellulose composite films with cellulose matrix and Napier grass cellulose fibril fillers. *International Journal of Biological Macromolecules*, Vol. 112, 2018, pp. 1310–1315.

[24] Kumar, T. S. M., N. Rajini, T. Huafeng, A. V. Rajulu, N. Ayrilmis, and S. Siengchin. Improved mechanical and thermal properties of spent coffee bean particulate reinforced poly(propylene carbonate) composites. *Particulate Science and Technology*, Vol. 37, No. 5, 2019, pp. 639–646.

[25] Muthu Kumar, S. T., K. Yorseng, S. Siengchin, N. Ayrilmis, and V. A. Rajulu. Mechanical and thermal properties of spent coffee bean filler/poly(3-hydroxybutyrate-co-3-hydroxyvalerate) biocomposites: Effect of recycling. *Process Safety and Environmental Protection*, Vol. 124, 2019, pp. 187–195.

[26] Thiagamani, S. M. K., R. Nagarajan, M. Jawaid, V. Anumakonda, and S. Siengchin. Utilization of chemically treated municipal solid waste (spent coffee bean powder) as reinforcement in cellulose matrix for packaging applications. *Waste Management*, Vol. 69, 2017, pp. 445–454.

[27] Senthil Muthu Kumar, T., K. Senthilkumar, M. Chandrasekar, N. Rajini, S. Siengchin, and A. Varada Rajulu. Characterization, thermal and dynamic mechanical properties of poly(propylene carbonate) lignocellulosic *Cocos nucifera* shell particulate biocomposites. *Materials Research Express*, Vol. 6, No. 9, 2019, p. 096426.

[28] Indira Devi, M. P., N. Nallamuthu, N. Rajini, T. Senthil Muthu Kumar, S. Siengchin, A. Varada Rajulu, et al. Antimicrobial properties of poly(propylene) carbonate/Ag nanoparticle-modified tamarind seed polysaccharide with composite films. *Ionics (Kiel)*, Vol. 25, 2019, pp. 3461–3471.

[29] Indira Devi, M. P., N. Nallamuthu, N. Rajini, T. Senthil Muthu Kumar, S. Siengchin, A. Varada Rajulu, et al. Biodegradable poly(propylene) carbonate using in-situ generated CuNPs coated *Tamarindus indica* filler for biomedical applications. *Materials Today Communications*, Vol. 19, 2019, pp. 106–113.

[30] Kumar, T. S. M., M. Chandrasekar, K. Senthilkumar, R. A. Ilyas, S. M. Sapuan, N. Hariram, et al. Characterization, thermal and antimicrobial properties of hybrid cellulose nanocomposite films with in-situ generated copper nanoparticles in *Tamarindus indica* nut powder. *Journal of Polymers and the Environment*, Vol. 29, No. 4, 2021, pp. 1134–1142.

[31] Senthil Muthu Kumar, T., N. Rajini, S. Siengchin, A. Varada Rajulu, and N. Ayrilmis. Influence of Musa acuminate bio-filler on the thermal, mechanical and visco-elastic behavior of poly (propylene) carbonate biocomposites. *International Journal of Polymer Analysis and Characterization*, Vol. 24, No. 5, 2019, pp. 439–446.

[32] Thiruganasambanthan, T., S. M. K. Thiagamani, C. Santulli, S. Krishnasamy, and C. Muthukumar. Preparation of sodium alginate/rice starch blend polymer film for soil moisture sensing. *Materials Today: Proceedings*, Vol. 64, 2022 Jan, pp. 352–356.

[33] Sivaram, R. N., S. M. Kumar Thiagamani, S. P, S. M, B. Y. Surya Narayana, H. Ebrahimnezhad-Khaljiri, et al. Isolation and characterization of agro-waste biomass sapodilla seeds as reinforcement in potential polymer composite applications. *Heliyon*, Vol. 9, 2023 Jul, id. e17760.

[34] Joseph Arockiam, A., S. Rajesh, S. Karthikeyan, S. M. K. Thiagamani, R. G. Padmanabhan, M. Hashem, et al. Mechanical and thermal characterization of additive manufactured fish scale powder reinforced PLA biocomposites. *Materials Research Express*, Vol. 10, No. 7, 2023 Jul, id. 075504, https://iopscience.iop.org/article/10.1088/2053-1591/ace41d.

[35] Sari, N. H., P. D. Setyawan, S. M. K. Thiagamani, Suteja, R. Tamimi, S. M. Rangappa, et al. Evaluation of mechanical, thermal and morphological properties of corn husk modified pumice powder reinforced polyester composites. *Polymer Composites*, Vol. 43, No. 3, 2022, pp. 1763–1771.

[36] Legesse, M. B. and S. A. Emire. Functional and physicochemical properties of mango seed kernels and wheat flour and their blends for biscuit production. *African Journal of Food Science and Technology*, Vol. 3, No. 9, 2012, pp. 193–203.

[37] Henrique, M. A., H. A. Silvério, W. P. Flauzino Neto, and D. Pasquini. Valorization of an agro-industrial waste, mango seed, by the extraction and characterization of its cellulose nanocrystals. *Journal of Environmental Management*, Vol. 121, 2013 May, pp. 202–209.

[38] Wang, Z., X. Qiao, and K. Sun. Rice straw cellulose nanofibrils reinforced poly(vinyl alcohol) composite films. *Carbohydrate Polymers*, Vol. 197, 2018, pp. 442–450, https://www.sciencedirect.com/science/article/pii/S0144861718306805.

[39] Thiagamani, S. M. K., H. Pulikkalparambil, S. Siengchin, R. A. Ilyas, S. Krishnasamy, C. Muthukumar, et al. Mechanical, absorption, and swelling properties of jute/kenaf/banana reinforced epoxy hybrid composites: Influence of various stacking sequences. *Polymer Composites*, Vol. 43, No. 11, 2022, pp. 8297–8307.

[40] Jeyaguru, S., S. M. K. Thiagamani, H. Pulikkalparambil, S. Siengchin, J. Subramaniam, S. M. Rangappa, et al. Mechanical, acoustic and vibration performance of intra-ply Kevlar/PALF epoxy hybrid composites: Effects of different weaving patterns. *Polymer Composites*, Vol. 43, No. 6, 2022, pp. 3902–3914.

[41] Nasimudeen, N. A., S. Karounamourthy, J. Selvarathinam, S. M. K. Thiagamani, H. Pulikkalparambil, S. Krishnasamy, et al. Mechanical, absorption and swelling properties of vinyl ester based natural fibre hybrid composites. *Applied Science and Engineering Progress*, Vol. 14, No. 4, 2021, pp. 680–688.

[42] Senthilkumar, K., S. Subramaniam, T. Ungtrakul, T. S. M. Kumar, M. Chandrasekar, N. Rajini, et al. Dual cantilever creep and recovery behavior of sisal/hemp fibre reinforced hybrid biocomposites: Effects of layering sequence, accelerated weathering and temperature. *Journal of Industrial Textiles*, Vol. 51, No. 2, 2022, pp. 2372S–2390S.

[43] Atiqah, A., M. Chandrasekar, T. Senthil Muthu Kumar, K. Senthilkumar, M. N. M. Ansari. Characterization and interface of natural and synthetic hybrid composites. In *Reference module in materials science and materials engineering*, Elsevier, US, 2019.

[44] Krishnasamy, S., C. Muthukumar, R. Nagarajan, S. M. K. Thiagamani, N. Saba, M. Jawaid, et al. Effect of fibre loading and Ca $(OH)_2$ treatment on thermal, mechanical, and physical properties of pineapple leaf fibre/polyester reinforced composites. *Materials Research Express*, Vol. 6, No. 8, 2019, id. 085545.

[45] Asrofi, M., S. M. Sapuan, R. A. Ilyas, and M. Ramesh. Characteristic of composite bioplastics from tapioca starch and sugarcane bagasse fiber: Effect of time duration of ultrasonication (Bath-Type). *Materials Today: Proceedings*, Vol. 46, No. 4, 2021, pp. 1626–1630.

[46] Jantasrirad, S., J. Mayakun, A. Numnuam, and K. Kaewtatip. Effect of filler and sonication time on the performance of brown alga (*Sargassum plagiophyllum*) filled cassava starch biocomposites. *Algal Research*, Vol. 56, 2021, id. 102321. https://www.sciencedirect.com/science/article/pii/S2211926421001405.

[47] Osman, A. F., L. Siah, A. A. Alrashdi, A. Ul-Hamid, and I. Ibrahim. Improving the tensile and tear properties of thermoplastic starch/dolomite biocomposite film through sonication process. *Polymers (Basel)*, Vol. 13, No. 2, 2021 Jan, pp. 1–21.

[48] Asrofi, M., H. Abral, A. Kasim, A. Pratoto, M. Mahardika, and F. Hafizulhaq. Mechanical properties of a water hyacinth nanofiber cellulose reinforced thermoplastic starch bionanocomposite: Effect of ultrasonic vibration during processing. *Fibers*, Vol. 6, No. 2, 2018, p. 40.

[49] Abral, H., G. J. Putra, M. Asrofi, J. W. Park, and H. J. Kim. Effect of vibration duration of high ultrasound applied to bio-composite while gelatinized on its properties. *Ultrasonics Sonochemistry*, Vol. 40, 2018, pp. 697–702. https://www.sciencedirect.com/science/article/pii/S1350417717303735.

[50] Atiqah, A., M. Jawaid, S. M. Sapuan, M. R. Ishak, M. N. M. Ansari, and R. A. Ilyas. Physical and thermal properties of treated sugar palm/glass fibre reinforced thermoplastic polyurethane hybrid composites. *Journal of Materials Research and Technology*, Vol. 8, No. 5, 2019, pp. 3726–3732.

[51] Jeyaguru, S., S. M. K. Thiagamani, A. G. Rajkumar, S. M. Rangappa, and S. Siengchin. Solid particle erosion, water absorption and thickness swelling behavior of intra ply Kevlar/PALF fiber epoxy hybrid composites. *Polymer Composites*, Vol. 43, No. 6, 2022, pp. 3929–3943.

[52] Thiagamani, S. M. K., S. Krishnasamy, C. Muthukumar, J. Tengsuthiwat, R. Nagarajan, S. Siengchin, et al. Investigation into mechanical, absorption and swelling behaviour of hemp/sisal fibre reinforced bioepoxy hybrid composites: Effects of stacking sequences. *International Journal of Biological Macromolecules*, Vol. 140, 2019, pp. 637–646.

[53] Nasimudeen, N. A., S. Karounamourthy, J. Selvarathinam, S. M. Kumar Thiagamani, H. Pulikkalparambil, S. Krishnasamy, et al. Mechanical, absorption and swelling properties of vinyl ester based natural fibre hybrid composites. *Applied Science and Engineering Progress*, Vol. 14, No. 4, 2021, pp. 680–688.

[54] Ilyas, R. A., S. M. Sapuan, R. Ibrahim, H. Abral, M. R. Ishak, E. S. Zainudin, et al. Effect of sugar palm nanofibrillated cellulose concentrations on morphological, mechanical and physical properties of biodegradable films based on agro-waste sugar palm (*Arenga pinnata* (*Wurmb.*) *Merr*) starch. *Journal of Materials Research and Technology*, Vol. 8, No. 5, 2019 Sep, pp. 4819–4830.

[55] Santos, A. C. M. Q. S., F. M. Monticeli, H. Ornaghi Jr, L. F. D. P. Santos, and M. O. H. Cioffi. Porosity characterization and respective influence on short-beam strength of advanced composite processed by resin transfer molding and compression molding.

Polymers and Polymer Composites, Vol. 29, No. 8, 2021, pp. 1353–1362.

[56] Puttaswamy, M., G. Srinikethan, and K. V. Shetty. Biocomposite composed of PVA reinforced with cellulose microfibers isolated from biofuel industrial dissipate: *Jatropha curcus* L. seed shell. *Journal of Environmental Chemical Engineering*, Vol. 5, No. 2, 2017, pp. 1990–1997. https://www.sciencedirect.com/science/article/pii/S2213343717301409.

[57] Abral, H., A. Atmajaya, M. Mahardika, F. Hafizulhaq, Kadriadi, D. Handayani, et al. Effect of ultrasonication duration of polyvinyl alcohol (PVA) gel on characterizations of PVA film. *Journal of Materials Research and Technology*, Vol. 9, No. 2, 2020, pp. 2477–2486, https://www.sciencedirect.com/science/article/pii/S2238785419307203.

[58] Asrofi, M., H. Abral, Y. K. Putra, S. M. Sapuan, and H. J. Kim. Effect of duration of sonication during gelatinization on properties of tapioca starch water hyacinth fiber biocomposite. *International Journal of Biological Macromolecules*, Vol. 108, 2018, pp. 167–176, https://www.sciencedirect.com/science/article/pii/S0141813017338916.

[59] Rangappa, S. M., J. Parameswaranpillai, S. M. K. Thiagamani, S. Krishnasamy, and S. Siengchin. *Food packaging: Advanced materials, technologies, and innovations*, CRC Press, New York, 2020.

[60] Sriariyanun, M., A. Tawai, and Y. S. Cheng. Influence of nanoparticles on the shelf life of food in packaging materials. In *Food packaging*, CRC Press, London, 2020, pp. 255–276.

[61] Senthil Muthu Kumar, T., K. Senthil Kumar, N. Rajini, S. Siengchin, N. Ayrilmis, and A. Varada Rajulu. A comprehensive review of electrospun nanofibers: Food and packaging perspective. *Composites Part B: Engineering*, Vol. 175, No. July, 2019, id. 107074.

[62] Abral, H., J. Ariksa, M. Mahardika, D. Handayani, I. Aminah, N. Sandrawati, et al. Highly transparent and antimicrobial PVA based bionanocomposites reinforced by ginger nanofiber. *Polymer Testing*, Vol. 81, 2020 Jan, id. 106186.

[63] Romainor, A. N., S. F. Chin, and S. Lihan. Antimicrobial starch-based film for food packaging application. *Starch - Stärke*, Vol. 74, No. 3–4, 2022 Mar, id. 2100207.

[64] Su, C., X. Zhang, X. Ge, H. Shen, Q. Zhang, Y. Lu, et al. Structural, physical and degradation characteristics of polyvinyl alcohol/esterified mung bean starch/gliadin ternary composite plastic. *Industrial Crops and Products*, Vol. 176, 2022, id. 114365.

[65] Kasim, A. Improvement on the bioplastic properties of polyvinyl alcohol (PVA) with the sago starch nanoparticle addition, Vol. 166, No. 1, 2022, id. 130.

[66] Nayak, S. and J. R. Mohanty. Study of mechanical, thermal, and rheological properties of areca fiber-reinforced polyvinyl alcohol composite. *Journal of Natural Fibers*, Vol. 16, No. 5, 2019 Jul, pp. 688–701.

[67] Niazi, M. B. K., Z. Jahan, A. Ahmed, B. Uzair, A. Mukhtar, and Ø. W. Gregersen. Mechanical and thermal properties of carboxymethyl fibers (CMF)/PVA based nanocomposite membranes. *Journal of Industrial and Engineering Chemistry*, Vol. 90, 2020, pp. 122–131, https://www.sciencedirect.com/science/article/pii/S1226086X20302902.

[68] Mahardika, M., H. Abral, A. Kasim, S. Arief, F. Hafizulhaq, and M. Asrofi. Properties of cellulose nanofiber/bengkoang starch bionanocomposites: Effect of fiber loading. *LWT*, Vol. 116, 2019, id. 108554. https://www.sciencedirect.com/science/article/pii/S0023643819308965.

[69] Asrofi, M., H. Abral, A. Kasim, A. Pratoto, M. Mahardika, and F. Hafizulhaq. Characterization of the sonicated yam bean starch bionanocomposites reinforced by nanocellulose water hyacinth fiber (WHF): The effect of various fiber loading. *Journal of Engineering Science and Technology*, Vol. 13, 2018, pp. 2700–2715.