

Sustainable Polymer Composites: Recycled LDPE/LLDPE Reinforced with Pineapple Leaf Fibre

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ABSTRACT

The substantial generation of plastic waste has drawn researchers' attention toward developing environmentally friendly bio-composite materials through polymer recycling. Among synthetic polymers, recycled low-density polyethylene (rLDPE) and recycled linear low-density polyethylene (rLLDPE) are notable for their cost-effectiveness, reduced density, superior surface hardness, and excellent electrical insulation properties. However, they suffer from reduced mechanical strength and quality due to chain degradation and contaminants. This research aims to produce a composite by incorporating sustainable pineapple leaf fiber (PALF) as reinforcement in an rLDPE/rLLDPE blend. The methodology involved processing PALF to ensure uniformity and purity, then blending it at 5%, 10%, 15%, and 20% ratios. The composites were compounded using a laboratory melt mixer at 150 °C and 40 rpm. Tensile and impact strengths, density, water absorption, and chemical interactions (via Fourier Transform Infrared (FTIR) spectroscopy) were examined. Results showed that tensile strength reached 27.7 MPa at 10% PALF, while Young's modulus increased to 1072 MPa at 20% PALF. Impact strength decreased to 17 kJ/m², whereas density increased to 1.25 g/cm³ at 20% PALF. Water absorption also increased with increasing PALF loading. Overall, the incorporation of PALF increased the composite stiffness, with Young's modulus reaching 1072 MPa, while density increased to 1.25 g/cm³ and impact strength decreased to 17 kJ/m² at 20% PALF loading.

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1. INTRODUCTION

Nowadays, plastic pollution and its environmental impact have gained worldwide attention in recent years. The excessive use and improper disposal of non-biodegradable plastics, particularly polyethylene (PE), pose significant ecological threats [1]. To address these challenges, there is a growing emphasis on finding sustainable alternatives to conventional plastics and encouraging the adoption of circular economy principles. Recycling polyethylene (PE) plastics hold significant environmental benefits. This practice conserves valuable resources, curbing the need for raw materials derived from fossil fuels and reducing energy consumption and greenhouse gas emissions. By diverting plastic waste from landfills, recycling contributes to a cleaner environment and a decrease in overall waste [2]. Additionally, recycling mitigates pollution risks associated with improper disposal, particularly in oceans and waterways, thereby protecting ecosystems and wildlife.

Recycling plastic, specifically in terms of strength properties, impact resistance, and structural integrity, is a nuanced process that involves both challenges and advantages. When plastic is recycled, especially in the case of thermoplastics like LDPE and polyethylene terephthalate (PET), the mechanical properties can undergo changes [1]. The strength of recycled plastic may be somewhat diminished compared to its virgin counterpart due to potential degradation during the initial manufacturing and recycling cycles. However, advancements in recycling technologies, such as improved sorting and processing techniques, have contributed to maintaining and enhancing the strength properties of recycled plastics [3]. Continuous advancements in recycling technologies and quality control measures are essential for optimizing the mechanical properties of recycled plastics and ensuring their effective use in diverse structural applications. Balancing the environmental benefits of recycling with the engineering requirements for strength and structural integrity remains a key focus in the ongoing efforts to create sustainable and resilient materials.

Although rLDPE and rLLDPE offers substantial environmental benefits, there are some disadvantages associated with the process. One notable drawback is the potential reduction in mechanical strength during the recycling of the materials. Each cycle of recycling can lead to a degradation of polymer chains, resulting in a decrease in the material's tensile strength and other mechanical properties [4]. This reduction in strength may limit the potential applications of rLDPE in certain high-performance or load-bearing scenarios. Therefore, fibres such as PALF, can help compensate for the potential loss in strength during recycling cycles. By reinforcing the polymer matrix, fibres contribute to improved tensile strength, flexural strength, and overall structural integrity [5]. Additionally, fibers can help overcome challenges related to the homogeneity and consistency of recycled plastics, ensuring a more uniform and predictable material performance [6].

The use of natural fibres as a substitute for synthetic fibres in reinforced composites has gained momentum, creating new possibilities for various industries. Natural fibres are attractive due to their impressive mechanical properties, cost-effectiveness, environmental friendliness, biodegradability, low weight, and renewable nature [7]. In today's technological landscape, bio-based composites present the chance to benefit the environment. This natural fibre offers numerous advantages for both the environment and industries seeking sustainable alternatives. Approximately 30 million tonnes of natural fibres are annually produced and utilized in various manufacturing processes such as clothing, packaging, paper-making, automobiles, building materials, and sports equipment [8]. The attractiveness of natural fibre composites to industries stems from their lower density and eco-friendly characteristics compared to

traditional composites [9]. These fibers, composed of cellulose and lignin, exhibit strength and flexibility due to hydrogen bonding among fibrils along their length.

Apart from Mexico, South Africa, Nigeria, Brazil and China, pineapple is a major tropical and subtropical plant cultivated in Philippines, Thailand, Malaysia [10]. Certainly, this results in pineapple leaves waste, and farmers seek avenues to dispose of it responsibly. PALF has been widely used in polymer composites due to its superior mechanical strength because of their highest cellulose content (ranges up to 80%) [11] and relatively low microfibrillar angle which imparts significant mechanical strength [12]. PALF-based composites also offered a notable benefit in terms of weight reduction when compared to conventional synthetic fiber composites [13].

This research contributes to the existing scientific knowledge by exploring the feasibility of using PALF as a sustainable reinforcement material in rLDPE/rLLDPE composites. The objective of the research is to incorporate PALF into rLDPE blended with rLLDPE and to evaluate the resulting mechanical and physical properties. Utilizing rLDPE/rLLDPE blends exploits the synergistic effects of both materials, combining the enhanced melt strength and processability of rLDPE with the superior toughness and puncture resistance of rLLDPE. The methodology involves systematically varying the PALF to rLDPE ratio to determine the optimal mix that strikes a balance between sustainability and mechanical performance, considering factors such as tensile strength, impact strength and physical properties. The study recognizes the need for ongoing advancements in recycling technologies and quality control measures to optimize the mechanical properties of recycled plastics, aligning with the global efforts to create sustainable and resilient materials for various applications.

2. MATERIALS AND METHODOLOGY

2.1 Materials

The organic raw material used in this research study was pineapples leaves sourced from Ladang Nanas Mos, Kuala Kangsar in Perak. The rLDPE/rLLDPE (blend ratio 2:8) was supplied by Lean Lee Trading Co Sdn Bhd. Sodium hydroxide (NaOH) used for the chemical treatments was purchased from Merck Millipore.

2.2 PALF Treatment and Preparation

Firstly, the pineapple leaves were cut and scraped until the fibers appeared. The fibers were then allowed to dry under the sun for two days. The dried PALF were immersed in 6% concentration of NaOH for 5 hours [14], and subsequently dried in the oven for 24 hours at 80 °C. FTIR analysis were conducted to obtain the functional group of both treated and untreated PALF. The functional groups of both untreated and treated PALF were subsequently determined using FTIR analysis in order to verify the effectiveness of the treatment process. The second step involved pulverizing the treated PALF in a ball mill. To reach the targeted particle size reduction of 200 µm, the milling process was carried out continuously for two days. After milling, the PALF powder was sieved through a 200 µm sieve. The transformation of PALF into powder form is shown in Fig.1.

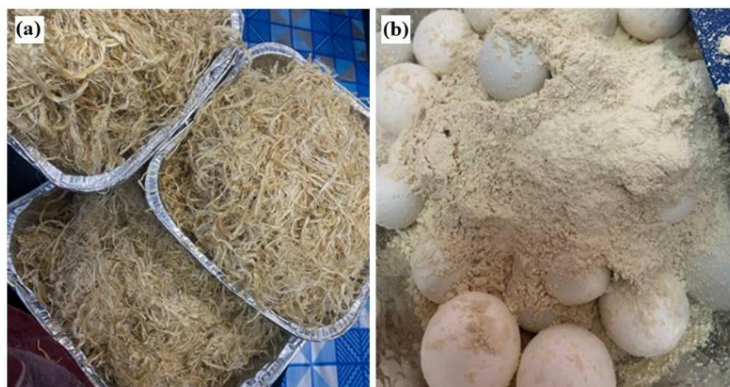


Fig. 1. (a) Treated PALF and (b) Pulverized PALF

2.3 Compounding of PALF with rLDPE/rLLDPE

The rLDPE/rLLDPE and PALF were compounded at a melting temperature of 150 °C according to the formulation shown in Table 1, using the Banbury Mixer (Model DW5310A). The rotation speed was set at 40 rpm. The compound was subsequently placed into square mould having a dimension of 150 mm x 150 and a thickness of 2 mm using a compression molding machine at 150 °C for 5 minutes. Next, the compounds were cut to the desired dimensions in preparation for subsequent mechanical and physical testing.

Table 1. The composition of rLDPE/rLLDPE and PALF composite

Compound	rLDPE/rLLDPE (%)	PALF (%)
0	100	0
1	95	5
2	90	10
3	85	15
4	80	20

2.4 FTIR Spectroscopy

Perkin Elmer FTIR spectrometer was used to observe the chemical structures of treated and untreated PALF, as well as the composites sample. The samples underwent scanning within the range of 4000 - 600 cm^{-1} . The absorption accuracy stood at 0.01%T, while the wavenumber accuracy was 0.01 cm^{-1} . The scanning resolution was set at 4 cm^{-1} , and each specimen underwent 16 scans for analysis.

2.5 Tensile Strength Test

The tensile strength test was conducted in adherence to ASTM D3039 standard using Instron Tensile Tester. For each compound, five samples with dimensions of 150 mm x 20 mm x 2 mm were tested, applying a 50 kN load and a crosshead speed of 100 mm/s. Tensile strength, percentage of elongation at break, and Young's modulus values was determined based on the averages derived from a minimum of three samples.

2.6 Impact Strength Test

The Notched Izod Impact test, in accordance with ASTM D256 standards, is a singular assessment gauging a material's resilience against the impact initiated by a swinging pendulum [15]. In this method, the Izod test was applied to a 10 mm x 64 mm x 2 mm of notched sample. The Gotech Izod Impact Testing Machine pendulum exerted a force of 2.7609 J for this evaluation. To determine the impact strength of PALF/rLDPE/LLDPE composites, the impact energy was divided by the cross-sectional area of the sample, and the average of three readings were documented as the impact strength.

2.7 Density Test

The density of PALF/rLDPE composites was assessed following ASTM D792 guidelines employing an Electronic Densitometer MD-3005. Square specimens measuring approximately 2 mm x 2 mm were utilized for this examination, with three replicates conducted for each compound.

2.8 Water Absorption Test

The ASTM D570 test method was employed to quantify the absorbed water content under specified conditions. Water absorption assessments was carried out by immersing the specimens in distilled water at room temperature. Following a 24-hours immersion, each sample was individually removed, surface water was wiped off using a dry cloth, and the immediate weight was measured with a digital scale. This process was reiterated on days 3, 5, 8, and 24. Each compound underwent testing with five samples, and the average values were subsequently computed. The percentage of water absorption was determined by the weight difference as shown in Eq. (1), where W_t represents the sample's weight at time (t), and W_o is the initial weight of the sample:

$$\text{Weight absorption (\%)} = \frac{(W_t - W_o)}{W_o} \times 100 \quad (1)$$

3. RESULTS AND DISCUSSION

3.1 FTIR Analysis

By referring to Fig. 2, FTIR spectra for untreated PALF display several characteristic peaks. A broad peak between 3400-3200 cm^{-1} indicates the presence of hydroxyl (O-H) groups, which are abundant in cellulose and hemicellulose. Peaks observed in the range of 2900-2800 cm^{-1} are associated with aliphatic C-H stretching vibrations. The region around 1731 cm^{-1} corresponds to the C=O stretching of acetyl groups in hemicellulose, while peaks near 1600 cm^{-1} and 1500 cm^{-1} are attributed to the aromatic skeletal vibrations of lignin. Lastly, the peaks around 1050-1030 cm^{-1} are indicative of C-O-C stretching vibrations in cellulose and hemicellulose. After alkaline treatment with NaOH, significant changes were observed in the FTIR spectrum of PALF. The O-H stretching peak, originally between 3400-3200 cm^{-1} , had shifted to a slightly lower frequency due to the removal of some hydroxyl groups and the reduction of hemicellulose and lignin content. The peak around 1731 cm^{-1} , corresponding to the C=O stretching had also shifted to a lower frequency, indicating the deacetylation of hemicellulose. Moreover, the peaks associated with lignin aromatic skeletal vibrations (1600 cm^{-1} and 1500 cm^{-1}) are reduced, reflecting the partial removal of lignin.

These changes confirm the effectiveness of NaOH treatment in modifying the surface chemistry of PALF by removing non-cellulosic components and increasing the relative cellulose content [14].

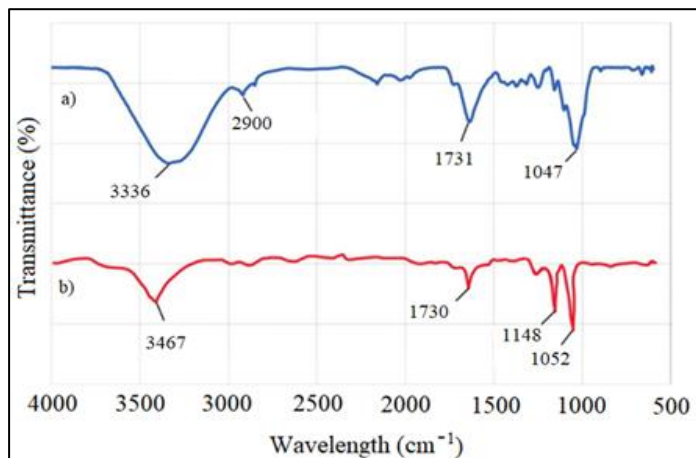


Fig. 2. FTIR Spectra of a) untreated PALF and b) treated PALF

The FTIR spectra for rLDPE/rLLDPE presented in Fig. 3 typically shows characteristic peaks corresponding to the molecular structure of PE. These include the symmetric and asymmetric C-H stretching vibration appearing at 2916 and 2848 cm^{-1} , C-H bending vibrations detected at 1468 cm^{-1} and C-H rocking vibrations appears at 719 cm^{-1} were associated to the methylene ($-\text{CH}_2-$) groups in the PE chain. When 10% PALF was incorporated into rLDPE/rLLDPE, the FTIR spectra of the composite exhibits additional peaks or changes in peak intensity, indicating the interaction between the fibers and the polymer matrix. The spectra show a slight reduction in the C-H stretching peak intensity to 2914 cm^{-1} due to the dilution effect of adding PALF, which has different chemical constituents compared to pure rLDPE/rLLDPE. New peaks corresponding to PALF, such as O-H stretching around 3379 cm^{-1} and C-O-C stretching around 1145 cm^{-1} , reflecting the presence of fiber within the composite. Additionally, shifts or changes in peak intensity associated with C-H bending and rocking vibrations might indicate interactions between PALF and the rLDPE/rLLDPE matrix.

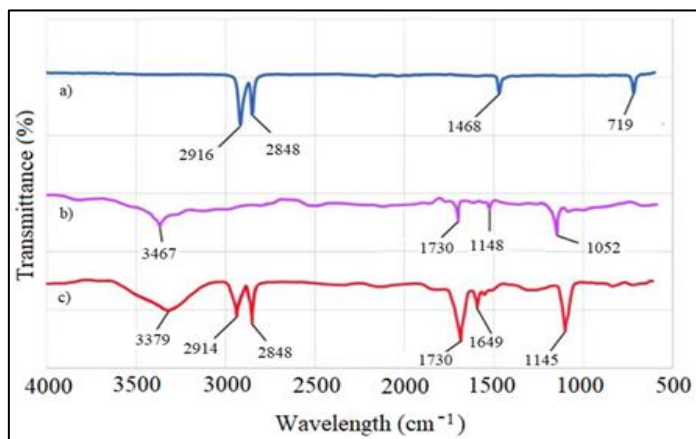


Fig. 3. FTIR spectra of a) rLDPE/rLLDPE, b) Treated PALF and c) 10% PALF/rLDPE/rLLDPE composites

3.2 Tensile Properties

Fig. 4 shows the tensile strength and elongation at break test results of rLDPE/rLLDPE composites filled with various PALF loading. The incorporation of PALF into rLDPE/rLLDPE had initially increased the tensile strength. The results indicated that the composite containing 10% PALF exhibits the highest tensile strength of 27.7 MPa. The increased in tensile strength at 10% PALF can be attributed to the effective reinforcement provided by the treated PALF fibers. NaOH treatment of PALF removes impurities and amorphous components such as hemicellulose and lignin, which enhances the fiber-matrix adhesion, leading to better stress transfer between the fibers and the polymer matrix [14]. This improved interfacial bonding results in a more efficient load-bearing capacity of the composite, thus enhancing its tensile strength [16]. However, further addition of PALF up to 20% had consequently decreased the tensile strength to the lowest value of 21.1 MPa. The tensile strength reduction can be due to the agglomeration and poor dispersion of fibers within the polymer matrix, hence creating stress concentration points and weakens the composite structure. Additionally, higher fiber content may lead to a reduction in the polymer content, which can compromise the overall flexibility and ductility of the composite, further contributing to the decrease in tensile strength [17].

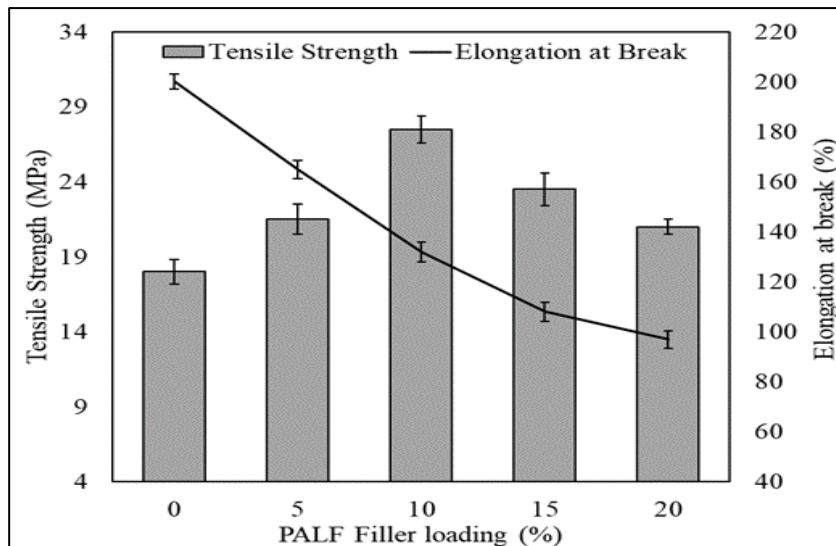


Fig.4. Tensile strength and elongation at break of PALF/rLDPE/rLLDPE composites

Fig. 4 also shows the increased in PALF loading had gradually decreased the elongation at break of the composites. Unfilled rLDPE/rLLDPE exhibits the highest elongation at break which could be attributed to its inherent ductility and flexibility. LDPE is a polymer known for its high elongation and ability to deform plastically under tensile stress. This characteristic allows the material to stretch significantly before breaking, making it suitable for applications requiring high flexibility. The reduction in elongation at break of the composites was due to the rigid nature of the PALF fibers, which restricts the polymer chains' mobility and decreases the composite's overall flexibility [18]. While the addition of fibers can improve the tensile strength of the composite, it often compromises its ductility. The fibers introduce points of stress concentration and limit the matrix's ability to undergo large deformations, resulting in a lower elongation at break [17].

Fig. 5 illustrates the addition of PALF into rLDPE/rLLDPE had significantly increased the Young's modulus of the composites. The increased in Young's modulus with rising PALF content can be attributed to the stiffening effect of the fibers. PALF fibers are significantly stiffer than the rLDPE/rLLDPE matrix, and their incorporation into the polymer enhances the composite's overall rigidity. As the fiber content increases, the composite becomes less flexible and more resistant to deformation under applied stress, resulting in a higher Young's modulus, which resulted from effective load-bearing and stress distribution capabilities provided by the PALF fibers, especially when well-dispersed within the matrix [19]. At 20% PALF content, the Young's modulus reached its maximum value of 1072 MPa. The substantial increased in modulus signifies that the composite has become much stiffer and more capable of resisting elastic deformation. However, it is essential to balance stiffness with other mechanical properties such as tensile strength and elongation at break, as excessive stiffness can lead to brittleness [20]. The composites with higher fiber loading contribute to greater stiffness. This improvement underscores the reinforcing effect of PALF fibers, making the composites suitable for applications requiring high rigidity and mechanical stability.

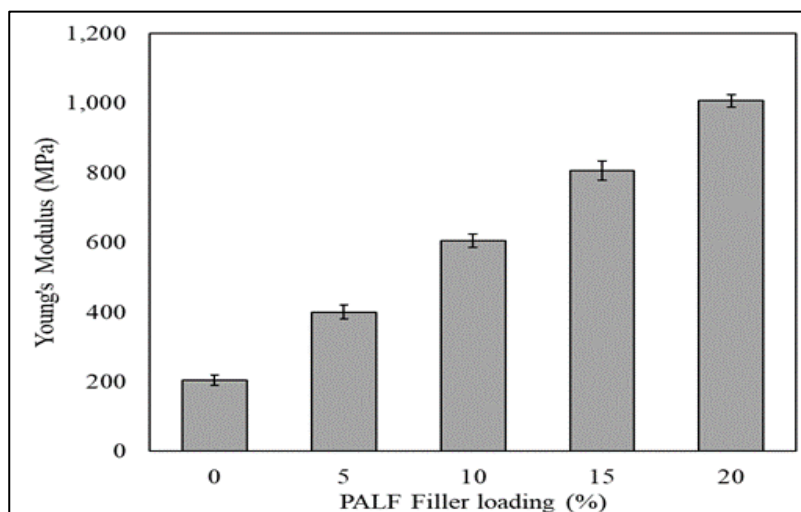


Fig. 5. Young's modulus of PALF/rLDPE/rLLDPE composites

3.3 Impact Strength

Fig. 6 shows the notched Izod impact strength results for PALF/rLDPE/rLLDPE composites. The data reveals that the impact strength of the composites decreased steadily with the increased in PALF loading. Specifically, the composite with 20% PALF has the lowest impact strength of 17 kJ/m², while unfilled rLDPE/rLLDPE exhibits the highest impact strength at 25 kJ/m². The decreased in impact strength can be attributed to the inherent brittleness introduced by the rigid fibers. The rLDPE/rLLDPE, being a polymer, possesses high impact strength due to its ability to absorb and dissipate energy through plastic deformation [21]. This ductility allows unfilled rLDPE/rLLDPE to withstand impacts and resist crack propagation effectively, resulting in higher impact strength values [18]. However, when PALF was incorporated into the rLDPE/rLLDPE matrix, the composite's ability to absorb impact energy diminished. The rigid PALF fibers restricted the polymer matrix's mobility and introduce points of stress concentration, which can initiate and propagate cracks more easily upon impact [13]. As a result, the energy absorption

capacity of the composite decreases, leading to lower impact strength values as the PALF content increases. Furthermore, the significant reduction in impact strength also indicated that the composite has become more brittle and less capable of withstanding sudden impacts. While the addition of PALF fibers enhanced other mechanical properties such as tensile strength and Young's modulus, it compromises the composite's toughness and impact resistance.

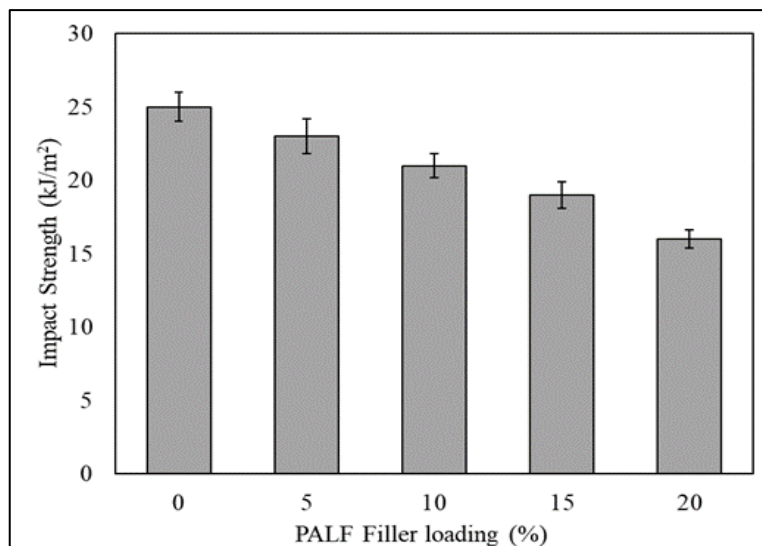


Fig. 6. Izod impact strength of PALF/rLDPE/rLLDPE composites

3.4 Density

Fig. 7 presents the density test results of PALF/rLDPE/rLLDPE composites. The data shows a significant increase in density upon the addition of PALF. Specifically, unfilled rLDPE/rLLDPE typically has a density of 0.91-0.94 g/cm³ [21], while the composite containing 20% PALF exhibits the highest density at 1.52 g/cm³. The increase in density with higher PALF content can be attributed to the denser packing of the composite materials. PALF fibers, being denser than the rLDPE/rLLDPE matrix, contribute to the overall density of the composite as their volume fraction increases. The addition of PALF increases the mass per unit volume of the composite, resulting in higher density measurements [22]. Furthermore, as the PALF content rises, the fibers tend to agglomerate more due to inadequate dispersion within the matrix [23]. This agglomeration further increases the composite's apparent density, as regions with higher fiber concentration occupy less volume than the surrounding matrix. The denser packing of fibers and matrix components also reduces the void space within the composite, contributing to the observed increase in density [24]. The highest density of 1.51 g/cm³ in the composite with 20% PALF indicates a significant increase in material compactness. This densification is beneficial for applications where material weight and dimensional stability are critical factors but may also affect other mechanical properties such as flexibility and impact resistance. This densification results in higher composite densities, demonstrating the effective incorporation of PALF fillers to modify the material's physical characteristics, as depicted in Fig. 7.

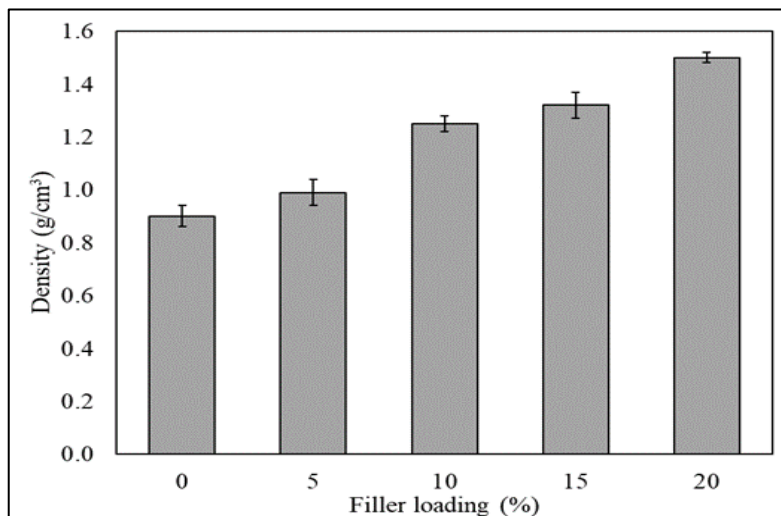


Fig. 7. Density of PALF/rLDPE/rLLDPE composites

3.5 Water Absorption

Water absorption property of PALF/rLDPE/rLLDPE composites is shown in Fig. 8.

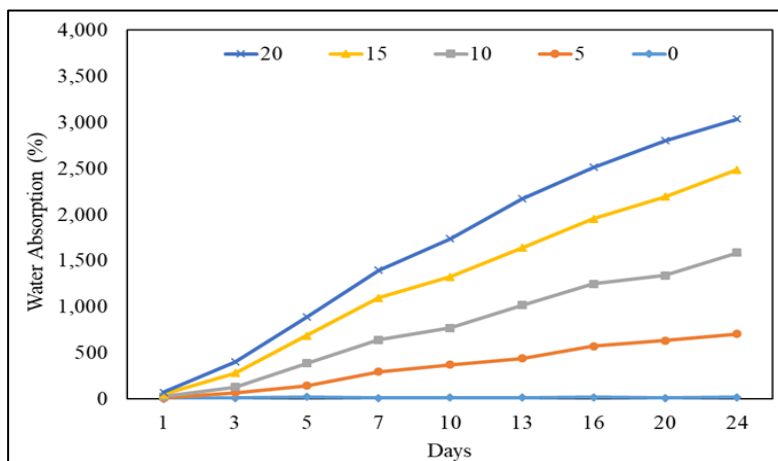


Fig. 8. Water absorption of rLDPE/rLLDPE composites filled with various PALF loading

The data reveals a trend where water absorption increased with higher PALF content in the composites. Water absorption in polymer composites typically increased with the addition of natural fibers such as PALF. This phenomenon occurs because natural fibers, even when treated with NaOH to improve compatibility with the polymer matrix, still retain some hydrophilic characteristics [25]. As the PALF content increases, more hydrophilic fibers are incorporated into the composite, providing additional pathways for water to penetrate and absorb within the material. Additionally, the agglomeration of fibers at higher PALF contents can create voids and gaps within the composite structure [25]. These voids act as reservoirs for water, further increasing the overall water absorption capacity of the material. The presence

of water within the composite can also weaken the fiber-matrix interface over time, potentially compromising mechanical properties such as tensile strength and modulus. As observed in Fig. 8, the composites with higher PALF content, such as 15% and 20%, exhibited the highest water absorption values. This indicates that as the fiber content increases, so does the composite's susceptibility to moisture ingress and retention.

4. CONCLUSION

In conclusion, FTIR analysis indicated effective removal of non-cellulosic content, thus enhancing fiber-matrix adhesion by removing hemicellulose and lignin. The rLDPE/rLLDPE composite containing 10% PALF exhibited the highest tensile strength of 27.7 MPa due to effective reinforcement, but higher PALF content had decreased the tensile strength due to fiber agglomeration. Unfilled rLDPE/rLLDPE had the highest elongation at break and impact strength due to its ductility, while the increased in PALF content had reduced the elongation at break and impact strength, making the composite more brittle due to stress concentration points introduced by the rigid fibers. In contrast the Young's modulus increased with rising PALF content, reaching a maximum of 1072 MPa at 20% PALF, indicating increased stiffness but also potential brittleness. Likewise, the density of the composites had also increased linearly with higher PALF content, reflecting the higher density of PALF compared to unfilled rLDPE/rLLDPE. Finally, water absorption capacity had also increased with higher PALF content due to the hydrophilic nature of natural fibers and the creation of voids within the composite. With 10% PALF being the ideal amount, these results imply that PALF/rLDPE/rLLDPE composites have potential for applications needing enhanced mechanical performance with the highest tensile strength property and intermediate values of impact strength, elongation at break, Young's modulus, impact strength, density, and water absorption.

5. ACKNOWLEDGEMENTS/FUNDING

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6. CONFLICT OF INTEREST STATEMENT

The authors affirm that there were no financial, economic, or self-benefitting conflicts of interest, and had no conflicts of interest with the funders.

7. AUTHORS' CONTRIBUTIONS

Siti Nor Din: Project administration, conceptualisation, data analysis and validation, supervision and writing-review and editing; Lily Shazleen Ghazali: Methodology, investigation, data analysis and writing-original draft; Faiezah Hashim: Data analysis and validation.

8. DECLARATION OF GENERATIVE AI IN THE WRITING PROCESS

During the preparation of this work, the authors used ChatGPT (OpenAI) to improve the readability, grammar, language quality, and clarity of the manuscript. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

9. DATA AVAILABILITY/SUPPLEMENTARY MATERIALS

The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

10. ETHICS STATEMENT

The authors declare that this research did not involve human or animal subjects. All experimental procedures were performed following the institutional Safety, Health, and Environmental (HSE) protocols of Universiti Teknologi MARA Cawangan Perlis.

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