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Experimental Investigation on the Influence of Processing Pressure on Mechanical Properties of Pineapple Leaf Fiber Reinforced Tapioca Biopolymer Composites

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ABSTRACT

The parameters employed in composite manufacturing are the primary factors influencing composite performance. This study assessed the effect of varying molding pressures on the mechanical characteristics of tapioca biopolymer (TBP) reinforced with pineapple leaf fiber (PALF) in the creation of entirely biodegradable materials. The present study selected four different molding pressures to identify the optimal processing pressure for achieving the highest mechanical properties. Samples of 30% PALF and 70% TBP were created using four different molding pressures (2 MPa, 4 MPa, 6 MPa, and 8 MPa). The findings demonstrate that PALF-TBP composites with 6 MPa molding pressure application yield the highest mechanical properties with 14.94 MPa of tensile strength, 17.46 MPa of flexural strength, and 15.31 KJ/m² of impact strength. Additionally, scanning electron microscopy (SEM) images of the fracture samples demonstrate a notable level of interfacial adhesion between the fibers and the matrix, as well as efficient stress transfer from the matrix to the fibers. The outcomes of the current study indicate that employing optimal molding pressure is essential for producing composites with superior mechanical properties. The capability to produce PALF-TBP composites is anticipated as a viable substitute for petroleum-based polymers across various applications, thereby advancing sustainable development efforts parallel to the

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Sustainable Development Goals (SDGs), specifically SDG 12 concerning responsible production and consumption.

INTRODUCTION

Composites made of natural fibers have gained popularity as a substitute for polymers derived from petroleum. It is because bio-based composites have many benefits, including being recyclable, renewable, biodegradable, non-irritating to the skin, lowering energy consumption, and posing fewer health hazards (Hadi et al., 2022a; Hassan et al., 2023; Wan-Mohtar et al., 2023). Furthermore, numerous automakers in the automotive sector have utilized bio-composites as materials for various parts and components. Kenaf fiber-reinforced plastic utilized in BMW series door panels and flax and sisal composites employed in the Mercedes-Benz E-Class trunk liner exemplify bio-composites in the automotive sector (Ahmad et al., 2014; Hassan et al., 2017; Moshood et al., 2022). Nevertheless, most bio-composites still depend on a petroleum-based polymer as a composite matrix. Therefore, most scientists and engineers are looking for a potential combination of natural fiber and biodegradable matrix as a future solution.

Previous studies indicate that the amalgamation of tapioca biopolymer (TBP) and pineapple leaf fiber (PALF) possesses significant potential as a fully biodegradable composite material (Hadi et al., 2022b). The parameter configuration in composite processing, particularly in compression molding, is a crucial determinant of composite performance. The appropriate parameters in composite manufacturing are essential for creating a comprehensive melt matrix that effectively infiltrates the fiber bundles, enhancing the quality and mechanical characteristics of bio-composites (Jaafar et al., 2019a).

The three primary sub-factors in the parameter setup of bio-composite manufacture are molding temperature, pressure, and holding duration (Jaafar et al., 2019b). Nonetheless, the majority of prior studies examining the correlation between material qualities and manufacturing parameters of bio-composites focus solely on the influence of a singular molding parameter. Furthermore, the majority of previous research has been on identifying the optimal temperature and length of exposure (Jaafar et al., 2019b). Furthermore, previous studies have determined that 165 °C with a 10-minute retention period is the ideal configuration for PALF-TBP composites (Jamiluddin et al., 2016). Conversely, the impact of utilizing varying pressures has garnered insufficient scrutiny. Processing variables exert nonlinear influences on composite mechanical characteristics. Three multi-parameter dimensions, which are compression pressure, holding time, and mold temperature, interact to affect the composite mechanical performance, as illustrated in Fig 1 (Lee et al., 2019).

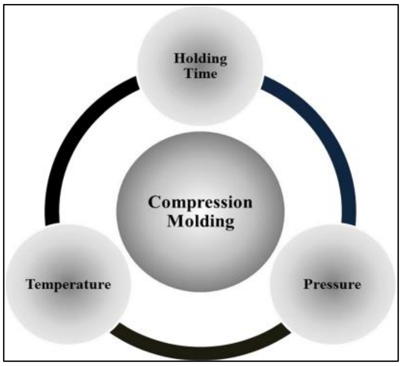

Fig. 1. The key parameters of the compression molding process (Peng-Gang et al., 2016).

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Fig 2 presents the spectrum of compressive pressures applied during the molding process for the prior production of bio-composites, such as jute-polypropylene (PP) composite, bagasse-based composite, low-density polyethylene (LDPE) reinforced with banana fiber, PALF reinforced polylactic acid (PLA), among others. Fig 2 illustrates that the compression molding pressure applied in earlier investigations for the production of bio-composites varied between 2 MPa and 20 MPa.

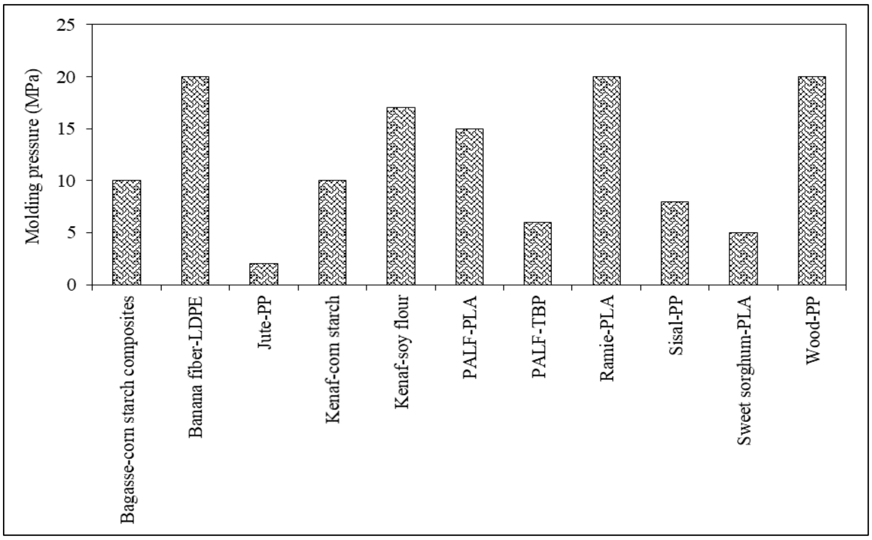

Fig. 2. Compression pressure employed in prior research for the purpose of compression moulding in the production of bio-composites (Khondker et al., 2005; Prasad et al., 2016; Zulkefli et al., 2025).

The application of elevated pressure during the moulding process improves the interface bonding between the matrix and reinforcing agent (Kim et al., 2019; Rubio-López et al., 2015; Saha & Kumari, 2023). It is imperative to remember that augmenting the compression molding capacity may compromise the natural fiber structure, adversely affecting the mechanical performance (Medina et al., 2009). This study sought to assess the impact of various stresses and identify the optimal molding pressure for the production of PALF reinforced TBP composites to achieve superior mechanical qualities. Consequently, develop a competitive, entirely eco-friendly composite that will ultimately supplant petroleum-derived polymers. Moreover, it is anticipated to endorse sustainable development measures in alignment with SDG 12, which pertains to responsible production and consumption. The decreased dependence on non-renewable materials will advocate for the sustainable management and efficient utilization of natural resources, which with Target 12.2 in SDG 12. Moreover, the results are expected to advance Target 12.4, which advocates environmentally sustainable chemical and waste management.

METHODOLOGY

Materials

The PALF bundles were obtained from Pemalang, Indonesia. The fiber originated from the process of scraping pineapple leaves. The RL-L10 MPL crushing equipment was subsequently employed to reduce the fiber length. The subsequent phase involves the screening of fibers that are shorter than 2.00 mm in length, utilizing a semi-automatic sieving machine, specifically the India-made For Bro SS304 GMP model.

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The TBP utilized in the present study was produced by Indochine Bio Plastiques Sdn. Bhd., located in Johor Bahru City, Johor, Malaysia. The biopolymer examined in this study was derived from Manihot esculenta, a species acknowledged for its industrial significance. The unpleasant flavor profile of these tapioca varieties makes them unsuitable for human consumption. Table 1 presents the attributes of TBP used in this research.

Table 1. The thermal and mechanical characteristics of TBP (Jamiluddin et al., 2016, 2018)

Properties	Value
Tensile strength	14 MPa
Maximum strain	37%
Molding temperature	165 °C to 185 °C
Density	1.1 g/cm³

Composites manufacturing

Short PALF and TBP were dried for 24 hours in a vacuum oven at 80 °C of drying temperature. Previous studies have determined that the optimal fiber content is 30% by weight (Jaafar et al., 2018). In the compounding procedure, the Germany-made Brabender Plastograph EC has a speed range from 0.2 min^{-1} to 150 min^{-1} and a 200 Nm of torque range. The internal mixing apparatus was preheated to 165 °C and adjusted to 40 revolutions per minute of rotation speed. Before the insertion of the fiber, a TBP was heated at the specified temperature and allowed to reach a stable state. The composites were mixed for a total of 20 min. Then, the internal mixer's mixed compounds were cut into pallets. Fig 3 shows the PALF-TBP pallet after the compounding process.

Fig. 3. The physical state of PALF-TBP compound.

The subsequent step was fabricating a composite plate through compression molding using Wabash G30H-CL, a 30-ton hydraulic press machine manufactured in the United States of America, creating a 180 mm × 180 mm × 6 mm plate. The mold temperature was set to 165 °C, the preheating phase lasting 5 minutes, followed by a subsequent press phase lasting 10 minutes. Following this, a cooling period of 10 minutes was seen under identical pressure conditions. The different compression molding pressure used in sample fabrication is presented in Table 2. According to the literature, most previous studies used 2 MPa to 20 MPa of compression molding pressure to fabricate natural fiber composites. However, prior research by Medina et al. (2009) have found that the characteristics of natural fiber samples have decreased when the molding pressure was beyond 6 MPa. Therefore, in this research, the 2 MPa to 8 MPa compression molding pressure was selected for producing PALF-TBP composite samples. Meanwhile, Fig 4 shows the PALF-TBP plate after the compression process.

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Table 2. Information regarding the compounding and molding pressure utilized in the preparation of PALF-TBP samples

Sample	Components composition (%)		Compression molding pressure (MPa)
	PALF	TBP	
CM2	30	70	2
CM4	30	70	4
CM6	30	70	6
CM8	30	70	8

Fig. 4. The composite plate of PALF-TBP following the compression process.

Subsequently, the composite plates were processed with a Thye Hong TH-13 laser cutting machine sourced from China to produce samples for tensile, flexural, and impact tests, adhering to ASTM D638, ASTM D790, and ASTM D6110 standards, respectively. Fig 5 illustrates the tensile test sample after the laser cutting process.

Fig. 5. A tensile test specimen of CM2, CM4, CM6 and CM8 following laser cutting.

Furthermore, Fig 6 illustrates the overview of the sample preparation and fabrication process of PALF-TBP composites in this study, commencing with the drying process and concluding with the laser cutting process.

Physical testing

Thickness measurement was conducted by using Mitutoyo 293-340-32 Digimatic Micrometer on all samples to investigate the effect of different pressures on sample thickness distribution. After that, the

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density test was performed using United States of America brand density meter Mettler Toledo Alfa Mirage MD-300S Desimeter according to ASTM D792 standard.

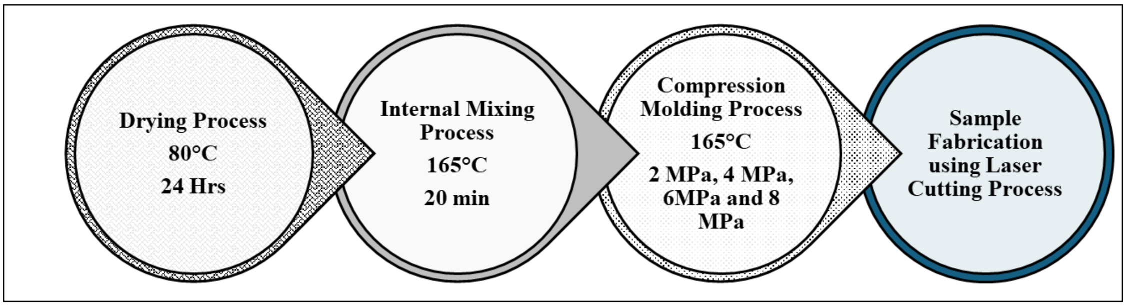

Fig. 6. The summary of the sample preparation and fabrication process of PALF-TBP composites in this study.

Mechanical testing

The 5 kN Shimadzu AGS-J universal testing machine was utilized in the tensile tests following ASTM D638 and flexural tests according to ASTM D790. A 1 mm/min and 1.28 mm/min crosshead speed was set for flexural and tensile tests, respectively. Five samples of each sample type were tested for failure. In addition, the elastic modulus for tensile and flexural was determined using the Chord modulus, E, where the modulus value was computed between 0.1% and 0.5% strain value. In the interim, the Charpy impact test was performed utilizing the Wolpert impact tester. The measurement was carried out following the guidelines set forth in ASTM D6110.

Fracture morphology

The tensile samples were visually inspected for their fracture surfaces using a JEOL JSM-6380, a high-performance SEM manufactured in Japan, featuring a resolution of 3.0 nm. The study sought to investigate the effect of various molding pressures on the fiber behaviour and interface bonding between the reinforcement agent and matrix interface by comparing and examining the surface morphology of composites.

RESULTS AND DISCUSSION

Influence of different molding pressures on physical properties

The physical examination of all samples revealed that the thickness diminished as molding pressure increased. Sample CM2 has the most thickness, averaging 7.13 mm, while sample CM8 displays the least thickness, averaging 6.73 mm. Concurrently, the density of the samples escalates with the augmentation of molding pressure. Sample CM2 exhibited the lowest density at 1.09 g/cm³, however, sample CM8 showed a notable rise in density, measuring 1.15 g/cm³. Fig 7 illustrates the thickness and density values of the samples influenced by various compression molding techniques.

The findings from this study showed that increasing pressure produced samples with lower thickness but higher density. This is because higher pressure forces the material into the mold more completely, aligning the polymer chains or compressing the material. Previous studies have also shown that high pressure also ensures better material distribution, reduces porosity, and increases crystallinity (Medina et al., 2009).

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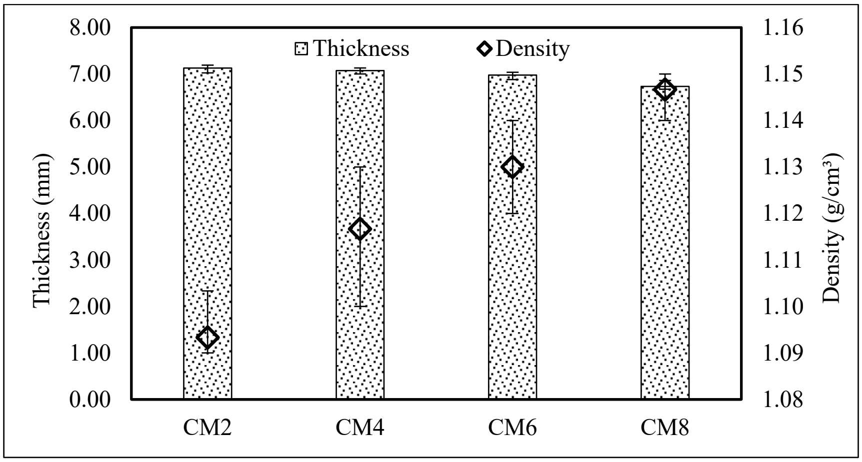

Fig. 7. The samples' thickness and density values are affected by different compression moulding values.

Influence of different molding pressures on mechanical properties

Fig 8 depicts the relationship between tensile properties and compression molding pressure. Using higher compression molding up to 6 MPa is observed to be capable of increasing tensile strength and modulus, respectively. It's worth noting that the utilization of higher molding pressure is expected to be capable of improving fiber binding in PALF-TBP composites. Tensile strength and tensile modulus increased about 33% and 74%, respectively, as the processing pressure rose from 2 MPa to 6 MPa.

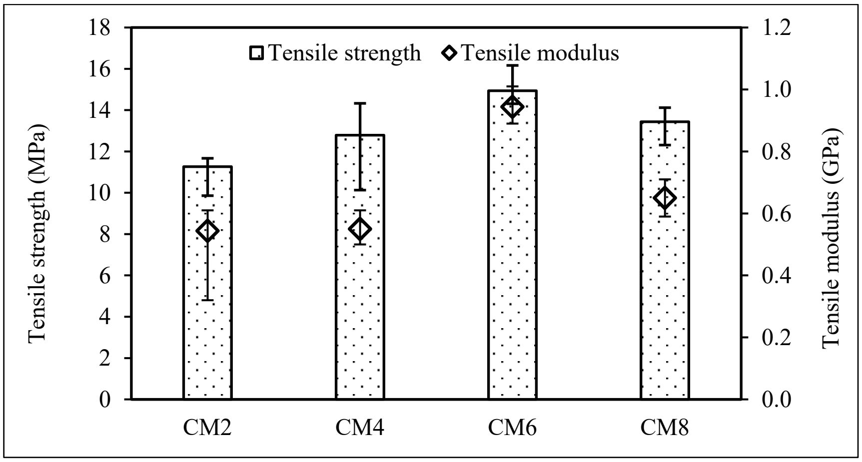

Fig. 8. The influence of molding pressure on the tensile characteristics of PALF-TBP composites.

Each incremental increase in compression moulding beyond 6 MPa resulted in a reduction of tensile strength and modulus by 10% and 31%, respectively. Nevertheless, the results obtained from CM8 continue to exceed the tensile properties of the CM2 and CM4 specimens. A comparable result was noted in the data regarding the flexural properties. The enhancement of compression moulding resulted in improved flexural properties, as illustrated in Fig 9. The CM6 sample exhibits the highest recorded flexural strength and modulus, measuring 17.46 MPa and 1.63 GPa, respectively. Data shows that the flexural strength and

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modulus decrease by 21% and 18%, respectively, as the compression moulding pressure increases from 6 MPa to 8 MPa.

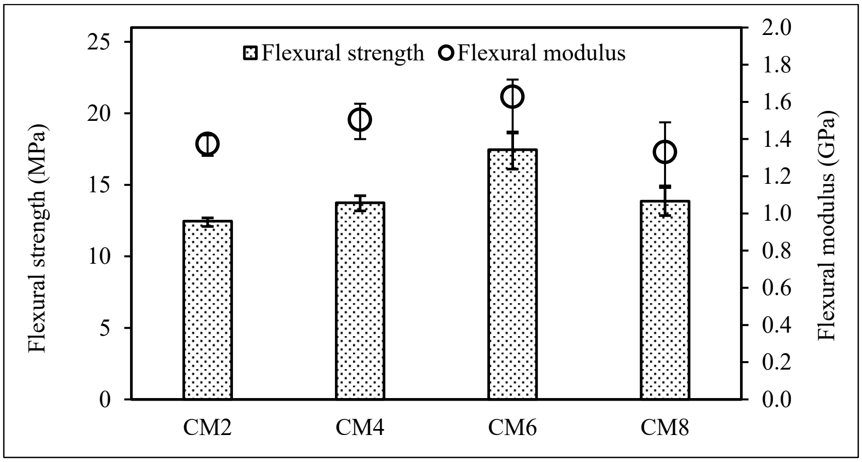

Fig. 9. The effect of varying processing pressures on the flexural characteristics of PALF-TBP composites.

Recent research demonstrates that the mechanical characteristrics of natural bio-composites, specifically PALF-TBP composites, are significantly affected by the compression molding pressure. The increase in the molding pressure enhanced the mechanical properties in the dynamic testing evaluation, which is the impact test result. In addition, the impact test result also presents the same pattern of the strength value as the tensile and flexural test results, with the highest impact strength produced by sample CM6, which is 15.31 KJ/m² as shown in Fig 10. The subsequent enhancement of compression moulding diminished the impact strength value similarly to the behaviour of tensile and flexural properties.

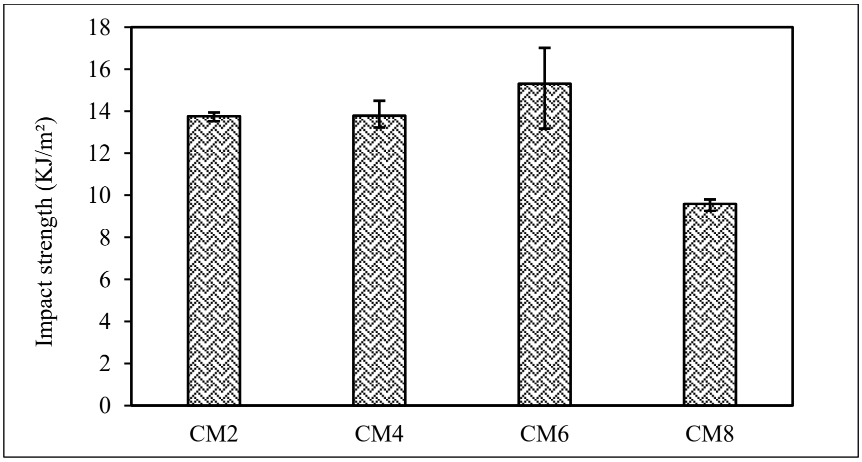

Fig. 10. The impact strength result of PALF-TBP composites affected by different molding pressures.

The current study concluded that increased molding pressure results in enhanced mechanical characteristics of composites. Prior research suggests that this phenomenon results from the widespread distribution of dispersed matrix particles, efficient fibers-matrix interfacial adhesion, and robust

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consolidation of the composite components (Jamal et al., 2024; Medina et al., 2009). The results regarding the influence of compression molding on physical properties corroborated this observation, as the reduction in thickness and increased density from elevated molding pressure indicated improved matrix dispersion, resulting in enhanced interfacial adhesion of the composites. Nonetheless, exceeding the optimal molding pressure will diminish the mechanical qualities of the sample. A key factor contributing to this occurrence is fiber degradation resulting from elevated pressure. The study revealed that a compression pressure of 6 MPa produced optimal mechanical performance in tensile, flexural, and impact strength, with values of 14.94 MPa, 17.46 MPa, and 15.31 KJ/m², respectively. This study demonstrates that the optimum application of pressure in the composite manufacturing process is a critical aspect in achieving superior mechanical characteristics of composites. A comparable result was noted in an earlier work by Medina et al. (2009), which indicated that composites composed of kenaf and hemp reinforced acrylic exhibit a decline in mechanical characteristics when the processing pressure exceeds 6 MPa during the impregnation process.

Fracture morphology on failure samples

Fig 11 presents SEM images comparing the TBP matrix and PALF fiber contact at 40X magnification for the CM2, CM4, CM6, and CM8 samples. Fig 11(a) illustrates that the interface adhesion between the matrix and the fiber in the CM2 sample is inadequate, as indicated by the observable fiber pull-out from the matrix. Furthermore, the uneven surface indicates inadequate compaction, resulting in a sparse dispersion of the dispersed matrix particles in CM2 samples. Sample CM2 exhibited significant and distinct porosity. The condition arose due to insufficient pressure during the composite manufacturing process, which resulted in air being entrapped and volatilized within the composite material. Nonetheless, the SEM picture of the CM4 sample in Fig 11(b) exhibits a more polished fracture surface in comparison to CM2. The combination of fiber breakage and fiber pull-out indicates an improvement in the fiber-matrix interfacial adhesion. Consequently, in comparison to the CM2 sample, the mechanical properties of the previously mentioned CM4 exhibit a greater value.

The SEM picture of the CM6 sample, depicted in Fig 11(c), demonstrates effective fiber-matrix adhesion characterized by small cracks, predominantly influenced by fiber breakage, fibrillation, and cracking rather than fiber pullout. The matrix exhibited excellent wetting of the fiber surface, characterized by minimal voids and a dense appearance at the boundary surface (Alias et al., 2024; Cionita et al., 2024) This occurrence demonstrates that elevating the molding pressure from 4 MPa to 6 MPa has effectively compressed the composite mixture and significantly improved the interfacial adhesion, where the stress is suspected to be well transferred from the matrix to the fiber. The anticipated outcome of this study is predicted to be a significant contributing factor in yielding improved mechanical properties for CM6 samples.

In contrast, CM8 exhibits a complete 100% fiber pull-out, and numerous holes detected on the fractured samples are presumed to result from the fiber pull-out phenomena. Additionally, the compromised fiber is also present in CM8. The observations of the CM8 sample yielded a noteworthy discovery. Two primary concerns arise when elevated pressures are employed in the composite manufacturing process. The primary concern is that excessive pressure during compression molding has resulted in the excessive expulsion of resin from the mold. This results in several fiber-rich regions or matrix deficiency, which causes inadequate bonding and a compromised composite. This condition was determined to be the cause of the fiber pullout phenomenon.

Furthermore, the application of high pressure was determined to have inflicted damage on the fiber structure. The compromised fiber was believed to have contributed to the stress resulting from incomplete transfer from the matrix to the fiber. The current findings align with and are corroborated by prior research on kenaf and hemp fibers subjected to varying pressures during the vacuum impregnation procedure. Exceeding the optimal pressure of 6 MPa renders additional compaction of the material impractical, leading to harm to the fundamental fiber structure. The predominant fiber structure experienced considerable

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damage when subjected to increased pressures. The two conditions led to insufficient interfacial adhesion in the CM8 sample, which impeded effective stress transfer from the matrix to the fibers. This situation was expected to explain the lower mechanical performance of the CM8 sample in comparison to the CM6 sample.

Thus, the finding deduces that the mechanical performance and interfacial adhesion of PALF and TBP with 6 MPa compression molding pressure are markedly better than those of other samples. In summary, the production parameters for composites must be meticulously tuned to guarantee that the resultant composite adheres to the requisite mechanical standards while preventing damage to the natural fibers.

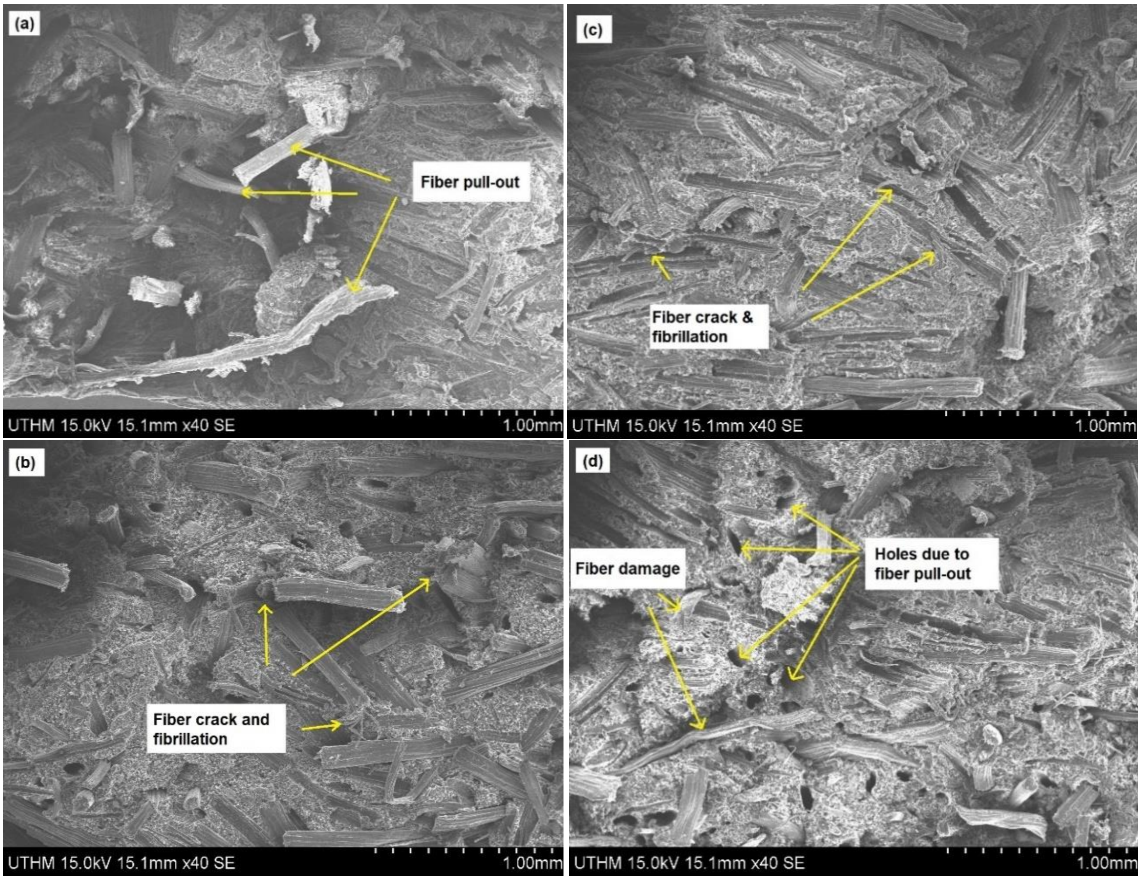

Fig. 11. The SEM images of PALF-reinforced TBP composites exposed to varying compression molding pressures: (a) CM2, (b) CM4, (c) CM6, and (d) CM8.

CONCLUSIONS

The utilization of optimal processing parameters, such as ideal molding pressure, in the fabrication of PALF-TBP composites seeks to enhance the mechanical properties of these composites while simultaneously advancing the creation of a sustainable and environmentally friendly polymer material poised to compete in the marketplace. This study aimed to explore the effects of varying moulding pressure applications on the mechanical characteristics of PALF-TBP composites. The results of the physical

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attributes indicate that CM8, processed at 8 MPa, exhibits the thinnest thickness, averaging 6.73 mm, and the highest density of 1.15 g/cm³, signifying effective material compaction and distribution, which minimizes porosity and enhances crystallinity. The application of 6 MPa moulding pressure produced the most favourable mechanical properties, achieving a tensile strength of 14.94 MPa, a flexural strength of 17.46 MPa, and an impact strength of 15.31 KJ/m². The SEM images of the fractured samples indicate a notable degree of interfacial adhesion between the matrix and fibers, along with effective stress transfer from the matrix to the fibers. As a result, the most recent research concluded that a pressure of 6 MPa is ideal for the synthesis of PALF-TBP composites. The findings suggest that the optimal application of pressure during the moulding process facilitates a superior distribution of the dispersed matrix particles within PALF-TBP composites, thereby enhancing the adhesion of PALF to the TBP matrix. The observed phenomena clarify the significant enhancement in the mechanical properties of the CM6 sample. The results of the present study demonstrate that the utilization of optimal molding pressure markedly improves the mechanical characteristics of PALF-TBP composites. Moreover, the application of optimal molding pressure is anticipated to enhance the feasibility of manufacturing fully biodegradable composites, such as PALF-TBP composites, as substitutes for petroleum-derived polymers in various future applications, particularly within engineering sectors like packaging and automotive industries.

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CONFLICT OF INTEREST STATEMENT

The authors agree that this research was conducted in the absence of any self-benefits, commercial or financial conflicts and declare the absence of conflicting interests with the funders.

AUTHORS' CONTRIBUTIONS

The authors confirm their contribution to the paper as follows: study conception and design: Mohamed Abdirahman Mohamed, Jamiluddin Jaafar, Deni Fajar Fitriyana; data collection: Muhammad Akmal Afiq Ibrahim, Ahmed Nurye Oumer; analysis and interpretation of results: Mohamed Abdirahman Mohamed, Eliza M. Yusup, Januar Parlaungan Siregar; draft manuscript preparation: Mohamed Abdirahman Mohamed, Shahrul Azmir Osman, Tezara Cionita. All authors reviewed the results and approved the final version of the manuscript.

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