Research Article

Rigid Polyurethane Foam Reinforced with Microfibrillated Cellulose from Oil Palm Empty Fruit Bunches as Efficient Thermal Insulation

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Abstract
There is increasing interest in enhancing the thermal insulation of rigid polyurethane foam (RPUF) through a sustainable approach. This work presents the utilization of micro-sized biobased fillers, microfibrillated cellulose (MFC) derived from oil palm empty fruit bunches (EFB) waste, to improve the properties of RPUF composite as a potential insulation material. The inclusion of MFC_{EFB} fillers at varied compositions (0.25–1 wt.%) in the RPUF composites demonstrated significant improvement of thermal insulation, as evidenced by their correlation with lower thermal conductivity, without compromising the mechanical properties compared to the control RPUF without MFC_{EFB}. The results showed a decrease in the thermal conductivity of RPUF composite by 7.92% with the addition of 0.5 wt.% MFC_{EFB}. The compressive strength also increased by 11.76% compared to the control RPUF. These enhancements were correlated to MFC_{EFB} acting as a nucleating agent in the RPUF foaming process, where morphological analysis confirmed that the addition of MFC_{EFB} resulted in smaller and more uniform cell sizes compared to the control RPUF. Fourier transform infra-red (FTIR) analysis revealed potential interactions between MFC_{EFB} and polyurethane that could improve foam structural integrity. Moreover, the incorporation of MFC_{EFB} at very low concentration had a negligible effect on the biodegradability compared to the RPUF control, demonstrating similar structural integrity to the control RPUF under natural environmental conditions, thereby ensuring the long-term durability of RPUF biocomposites.

Keywords: Biocomposite, Microfibrillated cellulose, Oil palm empty fruit bunches, Rigid polyurethane foam, Thermal insulation

1 Introduction

Rigid polyurethane foam (RPUF) is one of the most prevalent types of thermoset polyurethane [1], [2]. RPUF is typically made using methylene diphenyl diisocyanate (MDI) and polyols, resulting in high levels of cross-linking and a closed-cell structure [2], [3]. RPUF is widely utilized as a thermal insulating material in building and construction, owing to its low density, low thermal conductivity, excellent water resistance, and high strength-to-weight ratio [3]–[5]. In addition to global concerns about climate change and the need to reduce greenhouse gas emissions, RPUF is of particular interest in building insulation applications due to its excellent thermal insulation properties, energy efficiency, and compatibility with

various environmentally friendly strategies for improving building energy performance [6]. Therefore, there is significant interest in improving both the thermal insulation and mechanical performance of RPUF [7]–[9]. Enhancing the thermal insulation properties is achieved by reducing the thermal conductivity [7]. In polymer foams, thermal conductivity is directly related to heat transfer, which is primarily influenced by the diffusion of gas molecules and collisions within the foam's cellular structure. Cell size plays a crucial role in the heat transfer process. The smaller cell sizes limit the movement of gas molecules, thereby reducing heat transfer and lowering thermal conductivity [4], [10]. Previous studies have reported reducing the cell size in polymeric foams by adding fillers such as nano clays, carbon black, and aluminum oxide. These fillers act as nucleating agents, promoting a smaller cell size and enhancing insulation performance [8], [11], [12].

As a biopolymer, cellulose exhibits potential as a filler in polymer composites owing to its renewable sourcing, biodegradability, and non-toxic nature [13], [14]. It is primarily derived from lignocellulosic biomass, where cellulose strands are naturally bundled into semi-crystalline microfibrils enveloped by hemicellulose within a lignin matrix [14]–[16]. Most notably, microfibrillated cellulose (MFC) possesses a low density, extensive surface area, and a chemically reactive surface, promoting excellent mechanical properties of cellulose-reinforced polymers [3], [17]. Due to its excellent performances and abundant availability, reports of cellulose production have been explored extensively from various renewable resources such as agricultural residue [15], [18]–[20], and domestic wastes [21]. The utilization of cellulose, both microcrystalline cellulose [22] and nanocrystalline cellulose [3], [23], [24], as a reinforcing filler in RPUF has demonstrated significant improvements in both the mechanical and thermal insulation properties [3], [23], [24]. The concentration of cellulose reinforcement in RPUF is a critical parameter that must be optimized to achieve optimum improvement of RPUF performances [25]. In line with the broader trend of bio-based and nano-scale filler utilization in polymer composites, recent studies have reported improved mechanical and thermal properties that strongly depend on the filler type and its composition [26]. Unlike previous studies that primarily employed micro and nano crystalline cellulose, which require complex procedures, intensive chemical usage, and high energy demand, the use of microfibrillated cellulose (MFC) in this study presents a more facile, scalable, and cost–effective alternative while still achieving enhanced RPUF performance. To the best of our knowledge, no study has specifically reported the use of MFC derived from oil palm empty fruit bunches, referred to as MFC_{EFB}, to enhance the thermal conductivity of RPUF. These technical and environmental advantages further underline the novelty of this work, where MFC_{EFB} was effectively employed to enhance both the thermal insulation and mechanical properties of RPUF. In this study, MFC_{EFB} was incorporated into RPUF due to its excellent properties, in addition to its abundant availability in Indonesia and Malaysia. Notably, EFB is particularly promising as a cellulose resource, containing approximately 40%–43% of cellulose content [13].

The primary objective of this study was to investigate the structure-property relationship upon the incorporation of MFC_{EFB} in RPUF in terms of thermal insulation, mechanical properties and physical–chemical properties. The optimal composition of MFC_{EFB} in RPUF composites was investigated comprehensively to offer the optimum formulation to achieve high performance of RPUF biocomposite for insulating materials. Further, the biodegradability response of RPUF over a period of time upon the incorporation of MFC was also studied to promote sustainable building practice through energy efficiency and a more eco-friendly practice.

2 Materials and Methods

2.1 *Materials*

The EFB was generously provided by PT Mandiri Palmera Agrindo, Luwu, Sulawesi, Indonesia. Technical-grade sodium hydroxide was obtained for the alkaline pre-treatment. Additionally, the polyisocyanate, known as polyurethane Part A / Millionate MR–200, and the polyether polyol containing water catalyst and silicon surfactant, named Polyurethane Part B / JKR-7631L, were both acquired from PT. Justus Kimia Raya, Indonesia.

2.2 *Methods*

2.2.1 Extraction of MFC from EFB

Extraction of MFC from EFB is referred to our previous study [27]. The oil palm empty fruit bunches (EFB) fibers were initially mechanically cut into small particles (~1–3 cm in length) prior to treatment.

Afterward, an alkali steam explosion was performed using a CHEMEX steam explosion reactor with 10% NaOH at 150 °C and a pressure of 4 bar for 30 minutes to remove lignin and hemicellulose content. The treated fibers were then washed thoroughly with tap water until the filtrate reached a neutral pH (~pH 7) to eliminate residual chemicals. After washing, the fibers were dried at 80 °C for 24 hours to obtain MFC_{EFB}. To evaluate the cellulose purity of the obtained MFC_{EFB}, component analysis was conducted according to the protocol of the Indonesian National Standard (*Standar Nasional Indonesia* (SNI)) 01–1303–1989 at the Integrated Testing Laboratory, Center for Standardization and Instrument for Sustainable Forest Management, Ministry of Environment and Forestry, Bogor, Indonesia. Briefly, the oven-dried sample was first treated with cold water and chlorinated for several cycles. Residual chlorine and hydrochloric acid were removed by washing with 95% ethanol. The sample was then extracted using a 3% hot monoethanolamine solution at a temperature of 75 °C and subsequently washed with ethanol and cold water until the residue appeared white and showed a neutral pH. Then the residue was dried in an oven at 105 °C until a constant weight was achieved, and the cellulose content was calculated based on the weight difference before and after treatment. Based on the component analysis, the MFC_{EFB} contained 78.21±11.6% cellulose with individual fibrillated structures as depicted in Figure 1. The fibers on MFC_{EFB} had an average diameter of 16.23 ±3.07 µm and lengths of about 418.69 ± 70.31 µm. The dimension of MFC_{EFB} was determined using ImageJ software, with a minimum of 20 measurements of individual fibers from the SEM images to ensure accuracy. These microfibrillated cellulose fibers were utilized as reinforcing fillers in the RPUF formulation.

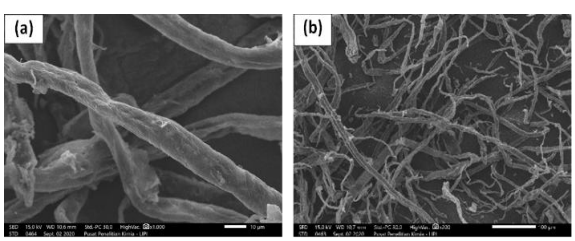

Figure 1: SEM of MFC_{EFB} at (a) magnification of X1000 and (b) magnification of X200.

2.2.2 *RPUF/MFC_{EFB} composite preparation*

The process of producing RPUF composites reinforced with MFC_{EFB} refers to the optimum conditions as reported in our previous research [5]. A varied composition of MFC_{EFB} at 0.25%, 0.5%, 0.75%, and 1% in weight percent was mixed into Polyurethane Part A using a digital overhead stirrer (IKA RW 20, Germany) at 500 rpm for 20 s. The selected MFC_{EFB} concentration range was determined based on previous studies on the incorporation of nanocellulose from EFB in RPUF [24] as well as other reports on microfiller reinforcement in RPUF [28]–[30]. Then, polyurethane Part B was added to Part A in a 3:2 ratio and mixed at 500 rpm for 20 minutes using the same digital stirrer. This ratio was chosen in accordance with the commercial manufacturer's formulation guidelines to ensure optimal foam formation. The mixture was poured into a mold and allowed to cure completely for up to 24 h at room temperature. The obtained RPUF composites were denoted as FMC_x, where x referred to the percentage weight composition of MFC_{EFB}. The control sample was also prepared without the addition of MFC_{EFB}, denoted as F_0. The detailed formulation of foam composites is represented in Table 1.

Table 1: Formulation of RPUF/MFC composite.

Sample Code	Formulation		
	PU Part A (gram)	MFC (gram)	PU Part B (gram)
F_0	30	0	20
$FMC_{0.25}$	30	0.125	20
$FMC_{0.5}$	30	0.25	20
$FMC_{0.75}$	30	0.375	20
FMC_1	30	0.5	20

2.2.3 *RPUF/MFC_{EFB} composite characterization*

Foam formation behaviour test. The foam formation behavior was measured using a cup test by measuring the cream, gel, and rise times during the foaming process. Creaming time was measured by the duration from completion of mixing parts A and B until the foam starts to rise, indicating that the mixture is saturated with the blowing agent. Gel time is the time calculated when the formation of a thin polymer forms, indicating that the cross–linking reaction is taking place. Free–rise time is when the foam reaches its maximum foaming expansion. Tack–free time is the time when the surface of the foam is no longer sticky, meaning when the foaming reaction is completely formed [31], [32].

Fourier transform infrared (FTIR) spectroscopy. The functional group and morphological analyses

were performed on the control and sample with the best thermal and mechanical insulation. The analysis used the Fourier transform infrared (FTIR)–Hyperion 2000 Bruker, Germany. The functional group analysis was performed using the Attenuated total reflectance (ATR) method with wave numbers ranging from 4000 cm^{-1} to 600 cm^{-1}, 32 scans, and a resolution of 4 cm^{-1}.

Mechanical properties. A Universal testing machine (Orientec UCT–5T, Japan) with a 100 Kgf load cell was employed to determine the foam's compressive strength and modulus, following the procedures outlined in ASTM D1621. Square-shaped specimens measuring 20 mm × 20 mm × 20 mm were prepared for the tests. Five specimens from each sample were tested, with the average value and standard deviation subsequently calculated. Tests were carried out in parallel and perpendicular directions of foam rise.

Morphological analysis. The morphology analysis of the RPUF/MFC_{EFB} was observed using a Scanning electron microscope (SEM - (JEOL JSM-6510LA) with a working voltage of 20 kV and magnification of 27X.

Thermal conductivity measurement. The foam's thermal conductivity was measured using the QTM-500 Meter (Kyoto Electronics). This involved placing the PD 11 probe above the sample and recording the thermal conductivity value. Using an electric current of 0.25 A at 20–30 °C for 60 seconds, the experiments were conducted within a thermal conductivity range of 0.00116–6 W/mK. Each sample was measured three times, with dimensions of 120 mm × 60 mm × 20 mm.

Degradability test. RPUF control (F_0) and the selected formulation RPUF/MFC composite were tested for degradability by burying them in the soil for 5 months and evaluating them monthly. Before the foam is buried in the soil, it is weighed. The debris on the specimens was cleaned with water and dried for 24 hours in an oven at 100–105 °C. The percentage of weight changes of the foams throughout the burial period was used to assess biodegradation. Further, the morphology before and after the biodegradability test was observed using a Scanning electron microscope (SEM) with a working voltage of 20 kV and 65X magnification.

3 Results and Discussion

3.1 *The effect of MFC_{EFB} on foam formation behavior*

In developing rigid polyurethane foam (RPUF) composites with cellulose filler, analysis of the foam formation behavior plays an important role in evaluating various aspects related to RPUF processing. To achieve this objective, the study measured specific parameters, including cream time, gel time, free-rise time, and tack-free time (Table 2).

Table 2: Foam formation behavior.

Sample	Cream Time (s)	Gel Time (s)	Free-Rise-Time (s)	Tack-Free-Time (s)
F_0	38.86 ± 0.05	198.75 ± 8.81	429.98 ± 15.0	663.80 ± 24.0
$FMC_{0.25}$	38.04 ± 0.24	210.40 ± 12.6	434.75 ± 18.1	673.16 ± 8.7
$FMC_{0.5}$	39.27 ± 0.16	217.58 ± 1.4	425.87 ± 8.1	672.78 ± 31.7
$FMC_{0.75}$	39.47 ± 0.01	219.51 ± 0.01	430.31 ± 8.3	680.65 ± 28.9
FMC_1	39.84 ± 0.4	225.00 ± 1.91	440.21 ± 10.9	693.31 ± 24.4

Incorporation of MFC_{EFB} with the composition, as in Table 1, showed an insignificant difference in cream formation time, which was in the range of 38–39 s. Further, generally, the addition of MFC_{EFB} in RPUF composites substantially increased the gel, rise, and tack-free time compared to F_0. The increased time during the foam formation was also reported in literature, suggesting that upon MFC addition, it was expected to increase the viscosity of the polyol premix [33], [34], thus slowing the formation of RPUF. Higher viscosity is known to reduce molecular mobility and introduce physical resistance during the foaming reaction, which can hinder the expansion of CO_2 bubbles produced from the isocyanate–water reaction.

Interestingly, $FMC_{0.5}$ is the only formulation with a shorter free rise time than F_0. This is probably because the incorporation at 0.5 wt.% of MFC_{EFB} creates a more homogenous dispersion of filler compared to other formulations. In addition to the aforementioned increase in viscosity, this improved dispersion provides more nucleation sites for foam formation. This higher density of nucleation points accelerates bubble formation during the foaming process, which causes the foam to rise faster because more bubbles are generated simultaneously [35]. This effect contributes to the formation of smaller and more uniform cell structures as cellulose filler acts as a

nucleating agent, which was observed in previous studies [17], [36] and further discussed in the morphological analysis section.

3.2 *The effect of MFC_{EFB} on the foam chemical structure*

Fourier transform infrared (FTIR) analysis was employed to examine the interaction between the matrix of polyurethane and MFC_{EFB}. The spectra were normalized with the absorbance of the phenyl group at a wavelength of 1595 cm^{-1} for consistency in comparison [33], [37]. The FTIR spectra of RPUF containing different amounts of MFC_{EFB} are presented in Figure 2a consistent with previous studies, the characteristics bands were observed at 3300 cm^{-1} corresponding to N–H stretching, at 1513 cm^{-1} (N-H bending), at 1700 cm^{-1} (C=O stretching), and at 1222 cm^{-1} (C-O stretching) confirming the successful synthesis of RPUF [3], [23], [38], [39]. Further, the incorporation of MFC_{EFB} was found to influence the chemical interaction in RPUF as the presence of hydroxyl groups on the MFC_{EFB} can affect the interaction and -NCO from isocyanates and -OH groups from during RPUF formation. This was evidenced by the increased intensity of the unreacted N=C=O peak at the wavenumber of 2270 cm^{-1} after the addition of MFC_{EFB}, as shown in Figure 2(b).

This phenomenon is attributed to the rise in viscosity, which may hinder effective mixing and reduce the reactivity between polyols and isocyanates. This is consistent with prior observations in polyurethane systems containing cellulose-based fillers, where higher viscosity limits reactive mobility and affects network formation [22]. This suggests that while the polyol remains the primary reactive component with –NCO, its reaction efficiency slightly decreases as cellulose content increases. This observation is further supported by the slight reduction in C=O (urethane) band intensity with increasing addition of MFC_{EFB} (Figure 2(c)), suggesting fewer urethane linkages.

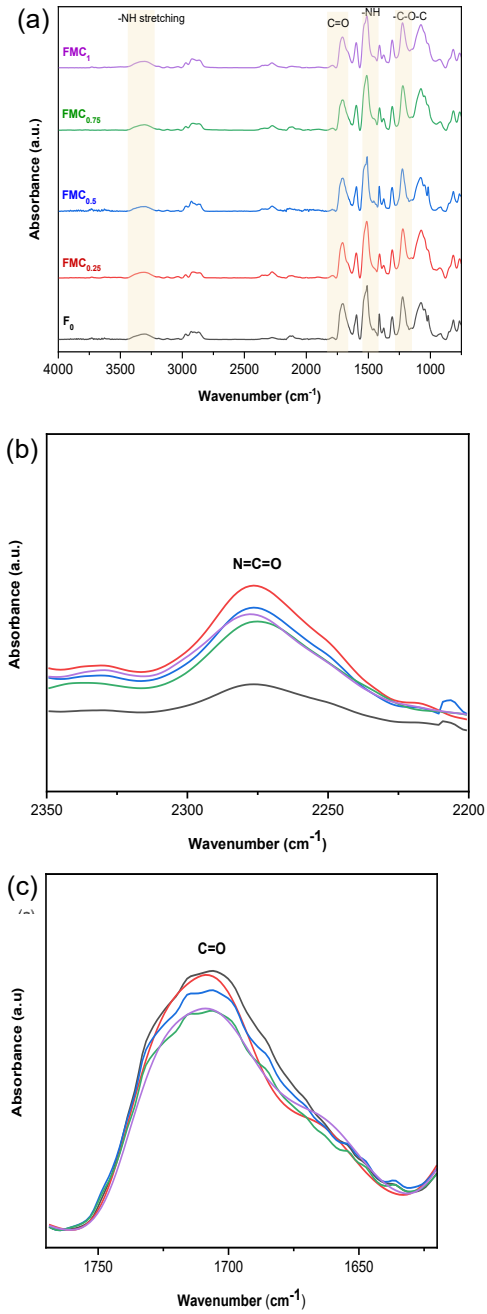

Figure 2: FTIR spectra of (a) RPUF and RPUF/MFC_{EFB}, with (b) magnification of N=C=O functional group, and (c) magnification of C=O urethane group.

Moreover, it was reported that OH groups on the cellulose surface of MFC can interact with polar groups such as C=O and NH bonds on the polyurethane backbone structure to form hydrogen bonds [23], [24]. This was evidenced by the shift in wave numbers at C=O and N-H, from 1711 to 1704 cm^{-1} and 1508 to 1513 cm^{-1}, respectively. These hydrogen bonds are likely to form an intermolecular network between MFC and the polyurethane matrix, thereby contributing to improved mechanical properties [24]. A schematic of the proposed interaction is illustrated in Figure 3. Although the OH groups on the surface of MFC could potentially react with isocyanates, such reactions were less favorable because the polyol used in this study was a polyether polyol, which contains primary hydroxyl alcohols that were significantly more reactive toward isocyanates than the secondary hydroxyl groups predominantly present in cellulose [33].

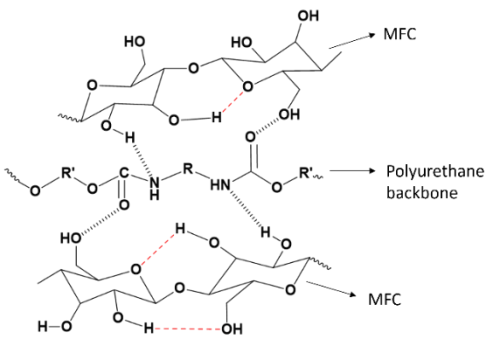

Figure 3: Schematic of the proposed interaction between MFC and RPUF backbone.

The findings from this study support the understanding that MFC_{EFB} interacts with the urethane matrix primarily through non-covalent hydrogen bonding, rather than covalent bonding with isocyanates, a mechanism similar to that reported for nanocrystalline cellulose [24]. MFC_{EFB} can form hydrogen bonds with NH and C=O in the polyurethane backbone, which contribute to structural reinforcement and foam stability. As a result, MFC_{EFB} plays a vital role in modifying foam morphology and mechanical behavior without significantly changing the formation of the polyurethane core network.

3.3 *The effect of MFC_{EFB} on the mechanical properties of RPUF*

To investigate the effect of MFC as a reinforcement, the compressive mechanical properties of RPUF samples were evaluated both parallel and perpendicular to the foam rise, as shown in Figure 4. The addition of MFC_{EFB} at a concentration of 0.25 wt.% initially reduced the compressive strength of the RPUF. An improvement in compressive strength was observed when the concentration of MFC_{EFB} was increased to 0.5 wt.%. Beyond this point, at the concentration higher than 0.5 wt.%, a significant decline was noted in both the compressive strength parallel and perpendicular to the foam rise direction. Meanwhile, the compressive modulus of neat RPUF increased with the addition of MFC_{EFB} in various concentrations. This enhancement was attributed to a reduction in elasticity, resulting in a stiffer material upon the incorporation of MFC_{EFB} [40].

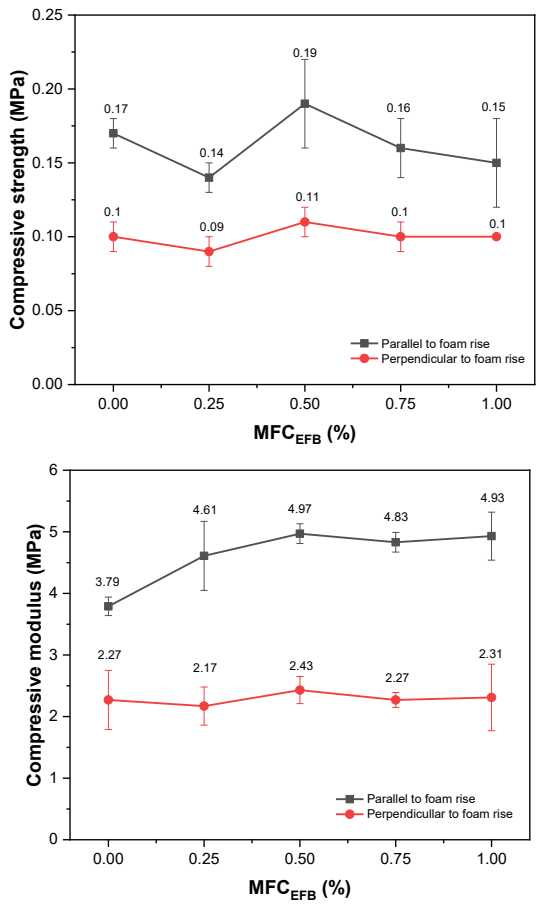

Figure 4: Mechanical properties of RPUF/MFC_{EFB} composite.

Overall, the compressive strength and modulus values in the parallel foam rise direction were higher

than those in the perpendicular direction. This result is closely related to the anisotropic behavior of rigid polyurethane foams, which arises from the oriented cellular structure developed during the foaming process. As the blowing agents drive the mixture upward, the expanding cells elongate and align along with the direction of rise. This orientation creates more uniform and continuous cell walls that can better withstand applied loads. In contrast, in the perpendicular direction, the cells are less oriented and more randomly distributed, resulting in lower mechanical resistance under compression. Similar anisotropic behavior has also been reported in previous studies on foam materials [7], [41].

At low concentrations ($FMC_{0.25}$), the amount of MFC_{EFB} in RPUF might be less than the threshold required to achieve a reinforcing effect. The decrease in compressive properties with the addition of 0.25 wt.% MFC_{EFB} could be attributed to insufficient and uneven dispersion of MFC_{EFB} within the polymer matrix. Under optimal conditions ($FMC_{0.5}$), an increased volume fraction and improved distribution of MFC_{EFB} could facilitate a greater formation of hydrogen bonds, as previously indicated in FTIR results. This enhanced interfacial interaction contributes to the improvement in compression properties.

At this optimal filler composition in RPUF, MFC_{EFB} was reported to promote a nucleation point, due to smaller and more evenly distributed cell sizes within the foam structure [13]. Smaller cell sizes were reported to result in a more even load distribution and packing density throughout the material, thus increasing its strength and resistance to stress and deformation [42]. The combined effects of additional hydrogen bonds (as noted by FTIR analysis, Figure 2) and smaller cell size contribution (supported by SEM analysis in the following discussion) upon the incorporation of MFC_{EFB} at optimal composition contributed to the improved mechanical properties of polyurethane foam. However, exceeding 0.5 wt.% of MFC_{EFB} loading in RPUF (FMC_1) induced filler agglomeration, which could distort the foam's cell structure, compromise structural integrity and consequently decrease compressive strength. Similar morphological distortions in RPUF have been reported in previous studies, resulting in reduced mechanical properties at higher reinforcement filler loadings using silica and glass powder [43]. This morphology induced deterioration in the mechanical performance of FMC_1 was supported by SEM analysis, as discussed in the subsequent section.

3.4 *The effect of MFC_{EFB} on the structure and morphology of RPUF*

The RPUF's primary morphological structure consisted mainly of closed cells in both parallel and perpendicular orientations to foam rise as seen in Figure 5. The ImageJ software was used to calculate the average cell size. There were notable differences in cell sizes between the parallel and perpendicular foam rise directions in both the control and selected RPUF composite ($FMC_{0.5}$ and FMC_1). Cell sizes in the parallel direction were smaller and more uniform than those in the perpendicular direction. This discrepancy may be attributed to uneven heat distribution during the open-mold foaming process. Consequently, the compressive performance of the foams (Figure 4) was notably higher in the parallel direction than in the perpendicular direction, as previously discussed.

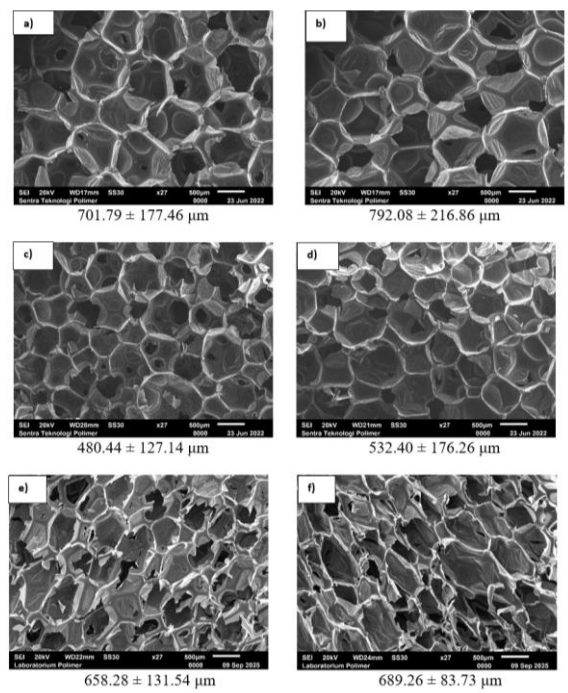

Figure 5: Morphological structure and cell size of (a) F_0 parallel direction to foam rise, (b) F_0 perpendicular direction to foam rise, (c) $FMC_{0.5}$ parallel direction to foam rise, (d) $FMC_{0.5}$ perpendicular direction to foam rise, (e) FMC_1 parallel direction to foam rise, and (f) FMC_1 perpendicular direction to foam rise.

Furthermore, the addition of MFC_{EFB} influenced the cell size in parallel and perpendicular directions [44], [45]. As shown in Figure 5, the addition of

MFC_{EFB} reduced the cell size compared to the control sample. Precisely, the incorporation of 0.5 wt.% MFC_{EFB} resulted in a decrease of 31.54% in cell size in the parallel direction and 32.78% in the perpendicular direction compared to the control sample. In line with previous discussion, MFC_{EFB} could function as a nucleating agent, as noted by the reduction in the time of foam formation (Table 2) and the interconnections in the cellular structure, as previously discussed in FTIR spectra in (Figure 2), resulting in a smaller cell-size foam and thus improving the mechanical properties (Figure 4). This was also reported in the other studies that cellulose fibers can function as a nucleating agent and contribute to the development of an interconnected cellular structure, resulting in a smaller cell-size foam [7], [24].

However, as shown in Figures 5e and 5f, the morphology of RPUF containing 1.0 wt.% MFC_{EFB} (FMC_1) in the parallel and perpendicular, respectively, exhibited more damaged foam cells and larger average cell size compared to the optimal sample containing 0.5 wt.% MFC_{EFB} ($FMC_{0.5}$). These morphological changes indicated that the excessive concentrations of MFC_{EFB} caused fiber agglomeration due to uneven dispersion [46]. These agglomerates act as local stress concentration points, interfering with gas expansion during the foaming process and leading to cell wall rupture. Consequently, the foam developed larger and less uniform cells, which not only reduced the compressive strength (Figure 4) but could also reduce its thermal insulation performance (in the subsequent discussion), as effective insulation depends on a small and uniform cell structure to restrict heat transfer.

3.5 *The effect of MFC_{EFB} on the thermal conductivity of RPUF*

Thermal conductivity is a key parameter in evaluating the insulating performance of polyurethane foams, which must maintain low thermal conductivity values to effectively minimize heat transfer. The typical specification for RPUF as an insulating material ranges from approximately ≤17–26 mW/mK [47]. However, conventional RPUFs often exhibit thermal conductivity values in the range of 17–30 mW/mK [47], indicating the need for strategies to further reduce this value. One effective approach is to reduce the foam cell size, as the dominant heat transfer mechanism in RPUF occurs through gas conduction, which is highly dependent on the mean free path of gas molecules. Smaller cell sizes result in shorter mean free paths, thereby limiting the movement of gas molecules and reducing the overall thermal conductivity of the foam [42].

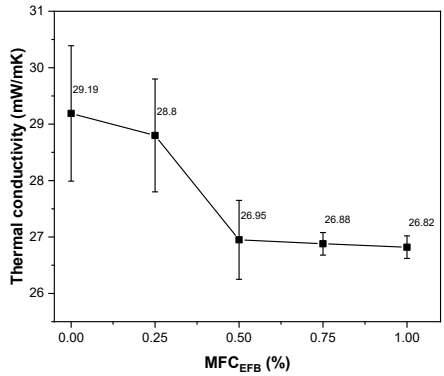

Figure 6: The thermal conductivity of the RPUF/MFC composite with various MFC_{EFB} concentrations.

Figure 6 illustrates the effect of MFC_{EFB} incorporation on the thermal conductivity of RPUF. The addition of MFC_{EFB} decreased the thermal conductivity of RPUF, reaching an optimum at 0.5 wt.% of MFC_{EFB}, with a reduction of the conductivity value of 7.92% compared to the control sample (F_0). In general, the smaller the cell size results the lower the thermal conductivity and, thereby, enhancing insulating properties [7], [30], [48]. As discussed earlier, MFC_{EFB} functions as a nucleating agent, promoting a smaller cell size as noted by SEM analysis (Figure 5), in which the incorporation of 0.5 wt.% MFC_{EFB} resulted in the smallest average cell size of $FMC_{0.5}$ at approximately 480.44 µm. Additionally, the incorporation of MFC_{EFB} at optimal composition provides resistance to excessive cell expansion during the foaming process, as suggested by FTIR analysis (Figure 2). This indicates that the hydroxyl groups on MFC_{EFB} may interact with isocyanate groups, providing improved compatibility and limiting excessive cell expansion during foaming. This phenomenon helps in reducing the possibility of damaged cell formation, as a result, improved mechanical property was observed (Figure 4). However, at higher MFC_{EFB} loadings (≥0.75 wt.%), the benefits begin to diminish. Excess filler content tends to cause fiber agglomeration, resulting in non-uniform dispersion with larger foam cells, as observed in the SEM images (Figure 5(e)–(f)). In addition, the foaming process is also disrupted, leading to weak RPUF matrix–MFC_{EFB} interaction, as evidenced by

FTIR analysis (Figure 2). The morphological disruptions and weakened interfacial MFC_{EFB} – RPUF interaction ultimately reduced the thermal insulation performance as reflected by an increase in thermal conductivity.

Several studies have explored strategies to reduce the thermal conductivity of RPUF by incorporating nanofillers such as NCC and nano–graphene [24], [34], [49]. Our findings suggest that low-cost micro fillers, particularly MFC_{EFB}, compared to nanosized materials, offer promising alternatives to improve the thermal insulation properties of RPUF without compromising the mechanical properties. In addition, compared to previous studies with the addition of nanocrystals cellulose (NCC) as a reinforcement in RPUF at the same loading level (0.5 wt.%), the reduction of thermal conductivity is equally observed at ~9% compared to RPUF without any reinforcement [24].

3.6 *The effect of MFC_{EFB} on the biodegradability of RPUF*

Conventionally produced RPUF derived from fully petrochemical resources decomposes very slowly in the natural environment due to the presence of isocyanurate rings as well as carbon derived from petrochemicals [50], offering long term durability. Hence, biodegradability studies play a crucial role in assessing such low composition of biopolymer fillers (MFC_{EFB}) in RPUF composites could compromise the performance of the lifetime usage of the product in the intended application. It was reported that the incorporation of cellulose into RPUF increases the foam's natural hydrophilic groups, encouraging higher water absorption and thus increasing decomposition ability [50]–[52].

As demonstrated in Figure 7, after 6 months of soil burial, the $FMC_{0.5}$ sample exhibited only a very slight weight loss of approximately 0.003%, while the F_0 sample showed virtually no change. The minimal degradation rate was likely due to the relatively low amount of cellulose incorporated into RPUF. This finding aligns with a study by Behjat, T., who reported that incorporating up to 30% cellulose filler into a synthetic polymer significantly increased biodegradation by around 10% for 4 months because a sufficient amount of cellulose is required for microorganisms to effectively initiate the degradation process [53].

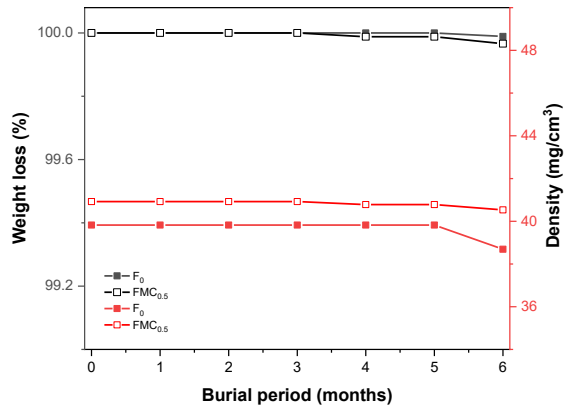

Figure 7: RPUF (F_0) and RPUF/MFC ($FMC_{0.5}$) biodegradability test results.

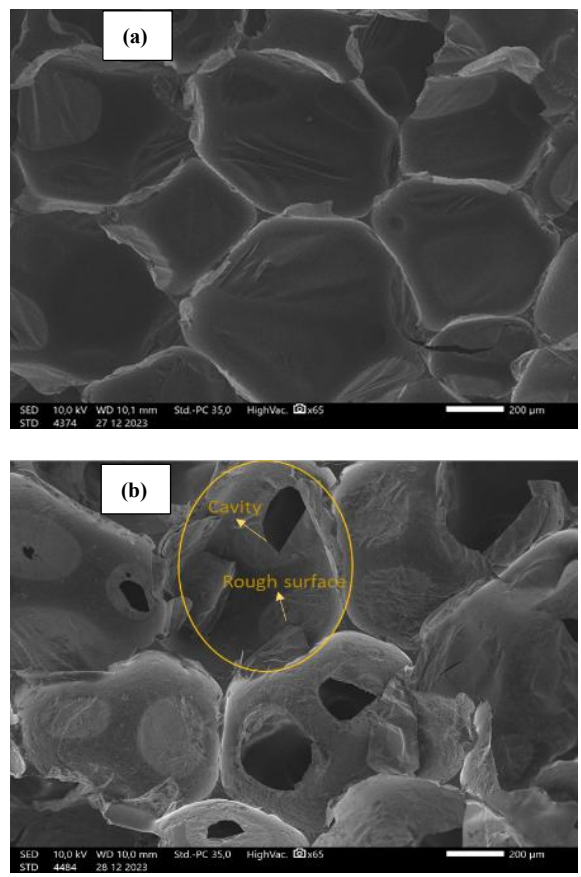

Figure 8: Morphology of $FMC_{0.5}$ (a) before and (b) after (right) biodegradability test.

A comparison of morphological examinations between the fresh control $FMC_{0.5}$ sample and $FMC_{0.5}$ after a 6-month burial period revealed that changes in the cell wall surface, which developed a rougher

texture, as seen in Figure 8, are likely due to surface erosion from the contact with the soil during the degradation test. This observation is supported by a slight decrease in the density of the $FMC_{0.5}$ sample (Figure 6), approximately 0.97%.

4 Conclusions

This study demonstrated that microfibrillated cellulose (MFC) derived from oil palm empty fruit bunches (EFB) is a promising reinforcement material for rigid polyurethane foam (RPUF), capable of enhancing both mechanical and thermal insulation of RPUF. MFC_{EFB} acted as a nucleating agent, promoting the formation of smaller and more uniform cell sizes, thereby improving mechanical and thermal insulation properties. It was important to note that the effectiveness of MFC_{EFB} as a nucleating agent depends on its concentration. In this study, the optimal concentration was achieved at the addition of 0.5 wt.% MFC_{EFB}. At higher concentrations, the tendency for MFC agglomeration becomes more pronounced, which negatively impacts the properties of the RPUF composite.

Table 3: Comparison of thermal and mechanical performance of RPUF composites.

Filler			Improvement (%)		Ref.
Filler Type	**Source**	**Opt. loading (wt. %)**	**Thermal Coduct.**	**Compress Strength**	
Micro fibrillated cellulose (MFC)	EFB	0.5	7.92	11.76	This study
Nano crystalline cellulose (NCC)	EFB	0.5	9.93	23.52	[24]
Microcrystalline cellulose (MCC)	Commercial (analytical standard)	1	0.78	12.5	[54]
Hybrid fillers (silica and alumina particles)	Commercial (analytical standard)	0.01	20	25	[55]

To provide an understanding of the effectiveness of MFC_{EFB} as a reinforcement in RPUF, Table 3 compares the thermal and mechanical performance achieved in this study with those reported in previous work. At an optimal loading of 0.5 wt.% MFC_{EFB}, the RPUF composite exhibited a 7.92% reduction in thermal conductivity and an 11.76% increase in compressive strength. Meanwhile, other studies showed that the incorporation of nanocrystalline cellulose (NCC) extracted from EFB at the same loading (0.5 wt.%) led to a slightly higher improvement, with a 23.52% increase in compressive strength and 9.93% reduction in thermal conductivity [25]. In contrast, microcrystalline cellulose (MCC) required a higher loading of 1 wt.% to improve compressive strength by 12.5%, but only resulted in thermal conductivity reduction of 0.78% [54]. Another study utilizing hybrid fillers consisting of silica and alumina particles at 7.5 wt.% achieved a 14% increase in compressive strength and 9% reduction in thermal conductivity [55]. The present study highlights the feasible and scalable use of MFC_{EFB} as an eco-friendly filler to achieve a balanced improvement in both insulation and mechanical properties of RPUF, using relatively low filler concentration.

Furthermore, the incorporation of MFC_{EFB} showed insignificant changes in the natural degradation compared to the RPUF control. This study supports the advancement of green materials to enhance the performance of RPUF while ensuring its durability to promote global sustainability goals.

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Author Contributions

A.A.S.: conceptualization, supervision, formal analysis, validation, funding acquisition, resources, writing—review and editing; A.R.: formal analysis, investigation, data curation, visualization, writing—original draft preparation; D.N.: methodology, data curation, formal analysis, investigation; Z.Z.: methodology, formal analysis; N.N.: Formal analysis, data curation: Nanda Nagara. All authors reviewed the results and approved the final version of the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

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