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Investigation of the Effect of Coupling Agent on the Properties of Kenaf Fiber/Polypropylene Composites

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Abstract. In a competitive product market, manufacturers ought to utilize eco-friendly materials to reduce the environmental impact of their products. Kenaf (*Hibiscus cannabinus*) is an annual plant that produces bast fiber and develops rapidly. These fibers have excellent properties and could serve as ideal reinforcing fillers for wood-based products. This study aims to investigate Maleated Polypropylene (MAPP) as a coupling agent for Kenaf fiber (KF)/Polypropylene(PP) composite. Three distinct composites were created using the following ratios: Sample 1 (Kenaf 20g: PP 180g), Sample 2 (Kenaf 10g: PP 180g: MAPP 10g), and Sample 3 (Kenaf 15g: PP 180g: MAPP 5g). All materials are mixed and extruded in a single-screw extruder at 185 to 200 ^{o}C at 50 rpm. A granulator is then utilized to palletize the samples. For testing, tensile and impact testing is conducted for mechanical properties, water absorption for physical properties, and Scanning Electron Microscopy (SEM) for morphological characterization. The thermal characteristics of the composites are analyzed using thermogravimetric analysis (TGA). The results indicate that the KF/PP composite with a coupling agent has a higher tensile strength with a value of 29.3 MPa compared to the KF/PP composite without a coupling agent with a value of 22.4 MPa. Water absorption of composite with coupling agent was also less than without coupling agent with a value of 1.05% and 1.31% respectively.

1. Introduction

Recently, numerous researchers have developed a serious interest in using natural fibers. The type of fiber, the use, and the cost of manufacture all influence how environmentally friendly a fiber is. Compared to conventional reinforcing or synthetic fibers, research shows that natural fibers offered a significant advantage. Both economic and environmental advantages can be obtained from natural fibers. The mechanical and physical qualities of these natural fibers were also impressive. Due to their abundance and low cost, which reduced maintenance costs, researchers claimed that natural fibers are more cost-effective in terms of raw materials than synthetic fibers [1][2]. Despite this, the widespread use of kenaf fibres in composites faces numerous obstacles. Standardizing the quality is essential because the quality of natural fibre varies depending on the type, soil, cultivation, and fibre separation techniques. According to the most recent price list published by the National Kenaf and Tobacco Board in 2022, the price of kenaf ranges between RM 3000 and RM 4,200 per tan, depending on quality and cleanliness. However, the price may decrease as seed quality improves due to higher yields, improved separation techniques, and rising demand [3-4].

In the Malvacea family, kenaf, commonly known as (*Hibiscus cannabinus L.*), is a non-wood plant fiber that can be used as reinforcing or filler in matrix materials. Kenaf fibre is a typical crop of the developing world, similar to bamboo and wood, which have origins in Asia and Africa, respectively. After the seeds have been dispersed, kenaf grows rapidly, typically within four to five months. The kenaf plant can grow in a variety of climates. The plant can reach heights of more than 3 m and has a base diameter of 3 to 5 cm [5]. Kenaf has advantages compared with other lignocellulosic fibre because kenaf has a fast growth cycle and flexibility to environmental conditions. Kenaf fibres is preferably to be choose by the industrial as its potential to be a polymer reinforcements in the natural fibre composite industry. Numerous previous studied had claimed that mechanical strength and thermal properties of kenaf composite are superior to other type of natural fibre polymer composites, thus regarded as a suitable applicant for high-performance kenaf fibre polymer composites [6-8].

The use of kenaf fiber-reinforced composites has increased in the building, maritime, packaging, sports, and automotive industries in recent years, with an emphasis on environmentally friendly and sustainable methods. In addition, the surface morphology of reinforcements and hybridization of reinforcements present opportunities for the development of materials with a cost advantage for specific applications [8-9]. A select number of automakers use kenaf fibre reinforced composites to manufacture automobile interior components including door panels, armrests, seat backs, dashboards, package trays, trunk liners, headliners, and consoles. More research is being conducted to determine whether kenaf fiber-reinforced composites can be used for more others automobile components [10-13].

Polypropylene is a thermoplastic polymer produced by polymerizing molecules of propylene. It is versatile due to its high heat distortion temperature, transparency, flame resistance, and dimensional stability. Creating natural-synthetic polymer composites using PP composites made from natural fibre is one of the best methods [14-15]. PP polymeric composites reinforced with natural fibres are widespread due to the development of novel production methods and processing tools [15]. To increase the compatibility of fibres with plastic matrices, researchers have focused heavily on coupling agents. Other studies have also demonstrated the efficacy of MAPP, with acknowledged sources dating back to the late 1970s. According to recent studies, fiber-matrix bonding can be significantly improved by using Maleated Polypropylene (MAPP) with a sufficiently high molecular weight and anhydride concentration. According to IR and electron spectroscopy for chemical analysis, the hydroxyl groups of cellulose and the MAPP have developed covalent bonds [16-17].

Composites composed of natural fibres such as kenaf and a polymer matrix are a promising alternative to synthetic composites. However, fiber-matrix adhesion and compatibility can have an effect on the performance and properties of these composites. A coupling agent can improve the interfacial adhesion between kenaf fibres and polypropylene matrix, thereby improving the composite's mechanical properties and overall performance [4]. Therefore, the objective of this study was to investigate the effect of MAPP as a coupling agent on the fiber-matrix interface bonding of kenaf fiber/polypropylene composites, as well as the composites' mechanical, thermal, and morphological properties. Environmental friendliness and potential applications of natural fibre composites have been the subject of previous research [4-6]. However, the performance of these composites can be inconsistent, particularly regarding issues involving fiber-matrix interface bonding. By examining the effect of a coupling agent on kenaf fiber/polypropylene composites, this study aims to address and overcome the limitations of prior research. Moreover, the findings from this research could lead to the development of high-performance, eco-friendly composites with enhanced mechanical properties, creating new opportunities for sustainable materials in a variety of industries, such as the automotive, construction, and packaging sectors. In addition, the study may contribute to the expanding body of knowledge in the field of composite materials, paving the way for future developments in eco-friendly material science.

2. Methodology

The dried kenaf used as a reinforcement in this work was purchased from Urich Technology. The matrix, Polypropylene (PP) in granules forms, and Maleated Polypropylene (MAPP) in this work were used as the coupling agent to improve the properties of the composite. NaOH was used for alkaline treatment.

In order to get rid of any debris or foreign objects and improve the bonding strength, the kenaf was cut to the desired length and cleaned in 10% of NaOH for alkaline treatment. The kenaf fibers were soaked in NaOH solutions, and then the moisture was drawn out of the fibers by drying them at 50°C in the oven for 24 hours. Kenaf was then ground using a grinder machine, the kenaf was then sieved to get size fibers of 0.5 mm. Lastly, the kenaf was then kept in a zip lock bag before being used in the next process.

In the extrusion process of the sample between kenaf fibers, PP, and coupling agent (MAPP), the mixture was mixed according to the respective weight ratio as shown in Table 1. The mixture of materials melts inside the extruder machine (Figure 1) to produce wired-shaped products. After undergoing the melting process in an extruder machine, the samples were then palletized using a palletizer or granulator machine as shown in Figure 2. The temperature for the extrusion process is 180°C with an average screw speed of 50 rpm.

A pelletizer machine was used to cut the composite fiber into pellet form after the KF/PP and KF/PP/MAPP mixture melted within the extruder machine. Then samples were proceeded to the next process which is injection molding. This process for producing sample sizes for testing tensile and impact tests. The temperatures used for injection molding are between 185◦C - 200◦C, For the tensile test, dumbbell-shaped specimens with dimensions 162mm long, 19mm wide, and 13mm are required. For impact test specimens with dimensions, 60mm x 10mm x 3mm and a V-notch with a 45° angle and 0.25 mm depth in the middle are required.

Table 1. The ratio of Kenaf fibers, Polypropylene (PP), and Coupling agent (MAPP)

Composite	Kenaf (g)	PP (g)	Coupling agent (g)
Sample 1	20	180	0
Sample 2	15	180	5
Sample 3	10	180	10

Figure 1. A wired-shaped product is produced by mixing sieved kenaf, PP granules, and MAPP granules.

Figure 2. A pelletizer machine is used to cut wired-shaped composite into pellets form.

2.1 Tensile Test

A universal testing machine was used to conduct tensile tests. Before testing, the specimens were stored in desiccators to prevent any moisture absorption. Following ASTM D628 and D570, tensile tests were conducted using a load cell with a 10 kN capacity and a crosshead speed of 5 mm/min at room temperature. The specimens were deformed to 1% strain at a crosshead speed of 5 mm/min during the tensile test to determine their elastic modulus and tensile strength. The strain was computed using a 50 mm gauge length. For each batch, three samples were examined, and average values were used for comparison.

2.2 Impact Test

The impact strength test determines a material's impact resistance. The impact test was conducted at room temperature using impact testing equipment in accordance with ASTM D256 (Izod V-notch) (hammer 4H). The impact strength of a specimen with dimensions of 60mm x 10mm x 3mm and a V-notch with a 45^{o} angle and 0.25 mm depth in the middle were also determined. For every batch, three samples were examined and as for the comparison, average values were considered.

2.3 Water Absorption Test

According to ASTM D-570 (Standard Test Method for Water Absorption of Plastics), the water absorption test was executed by weighing the sample both the mass and the time prior to immersion in water were recorded. A 60mm x 10mm x 3mm composite sample was submerged at room temperature 25 to 28℃ for 24 hours. The first sample's weight was determined. After being submerged for 24 hours, the sample was taken out of the water and dried using absorbent paper. The sample was quickly reweighed and submerged in water once more. Periodically, this technique was carried out. The water absorption was calculated using the formula shown below:

$$\text{Water Absorption Percentage (\%)} = \frac{w2 - w1}{w1} x\ 100\%$$

Where;
w1 = Weight of sample before soaking
w2 = Weight of sample after soaking

2.4 Thermal Analysis

To evaluate the thermal stability of the composites, thermogravimetric analysis (TGA) was carried out. The weight of kenaf/pp without coupling agent was 16.597mg and the weight of kenaf/pp with 10g of coupling agent was 17.375mg. The temperature that was used was 700◦C under nitrogen atmosphere and air at a constant heating rate of 20◦C/min to determine the temperature at which kenaf/PP and kenaf/PP/MAPP degraded.

2.5 Morphology Analysis

A focused electron beam was employed by a scanning electron microscope (SEM). The structural features, particles fiber and matrix, pull-out fibers, and void at the surface of the kenaf fibers were observed using the HITACHI scanning electron microscope. The surface of the samples was sputtered and coated with an ultrathin layer of gold. Coating the samples is necessary to enable or improve the imaging of materials using electron microscopy. By coating the sample with a conductive layer of metal such as gold, it is possible to prevent charging, reduce heat damage, and boost the secondary electron signal required for topographic analysis in the SEM (Au). The composite samples adhered to the support using double-sided carbon tape that was coated in a gold (Au) layer for metallization. The analysis used two samples only, one containing 10g of MAPP and the other 0g of MAPP were selected from each group of specimens for morphological analysis.

3. Results and Discussion

3.1 Tensile Properties

The tensile strength of KF/PP composites with and without various MAPP is shown in Table 2. The results indicate that sample 2 and sample 3 which consist of 5g and 10g of MAPP have higher tensile strength than sample 1 (without coupling agent) with a value of 28.393 MPa and 29.359 MPa respectively. It is demonstrated that MAPP improves fiber-polymer bonding interaction. The increase in tensile strength is attributable to the enhanced adhesion between the fibre and matrix, which permits greater stress transmission by bonding to the fibres. Eszer and Ishak (2018) claim that in the presence of MAPP, which interacted with the hydroxyl group on the surface of the kenaf fibre, a good stress transmission interface is generated due to probable covalent contact between the anhydride and hydroxyl groups of the kenaf, as well as chain tangling between the MAPP and PP chains [7]. Due to that reason, additional MAPP is proof that the tensile strength improved by around 28% compared to the sample without additional MAPP.

Table 2. Tensile strength of KF/PP with and without MAPP

Sample	**Tensile Strength (Mpa)**
Sample 1 [KF/PP (20g/180g)]	22.404 (0.02)*
Sample 2 [KF/PP/MAPP (15g/180g/5g)]	28.393 (0.15)
Sample 3 [KF/PP/MAPP (10g/180g/10g)]	29.359 (0.18)

*Value in parentheses is standard deviation

3.2 Impact properties

An impact test is being tested to determine how much energy is absorbed during fracture. Table 3 shows the impact strength of KF/PP composites with and without a coupling agent (MAPP). The greatest value is achieved by the combination ratio of KF/PP/MAPP (15g/180g/5g) and (10g/180g/5g), which was measured with the same value of 1971.994 J/m^2. These results show MAPP has a significant effect on the impact strength of PP composites. Compared to KF/PP composites without MAPP, the strengthening effect of KF/PP containing 5g MAPP and KF/PP containing 10g MAPP both showed an increase in impact strength. However, composite samples that do not contain MAPP additives slightly reduce the impact strength. MAPP are purposely use because of the function of strengthened the composite. MAPP not only use for fiber surface but also to achieve better interfacial bonding between fiber and matrix. The PP allows MAPP to be cohesive and produce maleic anhydride grafted polypropylene. By adding MAPP the surface energy of kenaf fiber is increase approximately to the surface energy of the matrix. Due to that, resulting in better wettability and higher interfacial adhesion of the fiber. Therefore it prove by the increasing mechanical properties such as tensil and impact strength of the composite.

Table 3. Impact strength of KF/PP with and without MAPP

Sample	Average of Impact Strength (JM^2)
Sample 1 [KF/PP (20g/180g)]	1321.392 (0.03)*
Sample 2 [KF/PP/MAPP (15g/180g/5g)]	1971.994 (0.01)
Sample 3 [KF/PP/MAPP (10g/180g/10g)]	1971.994 (0.05)

*Value in parentheses is standard deviation

3.3 Water Absorption Properties

Table 4 shows the water absorption of the samples. It was found that samples of KF/PP without a coupling agent (MAPP) absorbed more water (approximately 1.31%) than other samples. KF/PP had the highest observed water absorption value when no coupling agent was present. This might be because of the coupling agent (MAPP), which limits water absorption into the composite. The presence of MAPP, which interacts with the hydroxyl group on the surface of the kenaf fiber reduces the tendency of bonding from water molecules. The hydroxyl groups of cellulose and the MAPP developed covalent bonds increasing the strength of the composite and reducing the water absorption of the composite.

Table 4. Water absorption test data of KF/PP composites with and without MAPP.

Sample	Weight Before Soaking, g (w1)	Weight After Soaking, g (w2)	Total Average of Water Absorption Percentage (%)
Sample 1	1.9698	2.0078	1.31
Sample 2	1.9827	2.0007	1.07
Sample 3	2.2346	2.0049	1.05

3.4 Thermal Properties

The influence of MAPP on the thermal stability of composites with and without coupling agents was evaluated by TGA, and the results were shown in Figure 3. Even with variable amounts of filler loading, all composites demonstrated the corresponding pattern in the thermal degradation profile. There was a considerable difference between the KF/PP composites (Figure 3 (a) and (b)). Two distinct results were recorded, KF/PP Figure 3 (a) by weight 16.597 mg shows Onset T: 422.4 Cel; while the % weight loss: 91.49% (15.19 mg). Meanwhile, the Residue @ T: 698.5 Cel; 8.14 % (1.35 mg). On the other hand, KF/PP/MAPP (10g) shown in Figure 4.3.1 (b) by weight 17.375 mg shows Onset T: 420.8 Cel; while the % weight loss: 99.67 % (17.32 mg). Therefore, the Residue at T: 699.0 Cel; 0.25 % (0.04 mg). Hence this study discovered that when the ratio of coupling agent (MAPP) increased, the composite without MAPP shows more thermal stability compared to the composite with 10g MAPP.

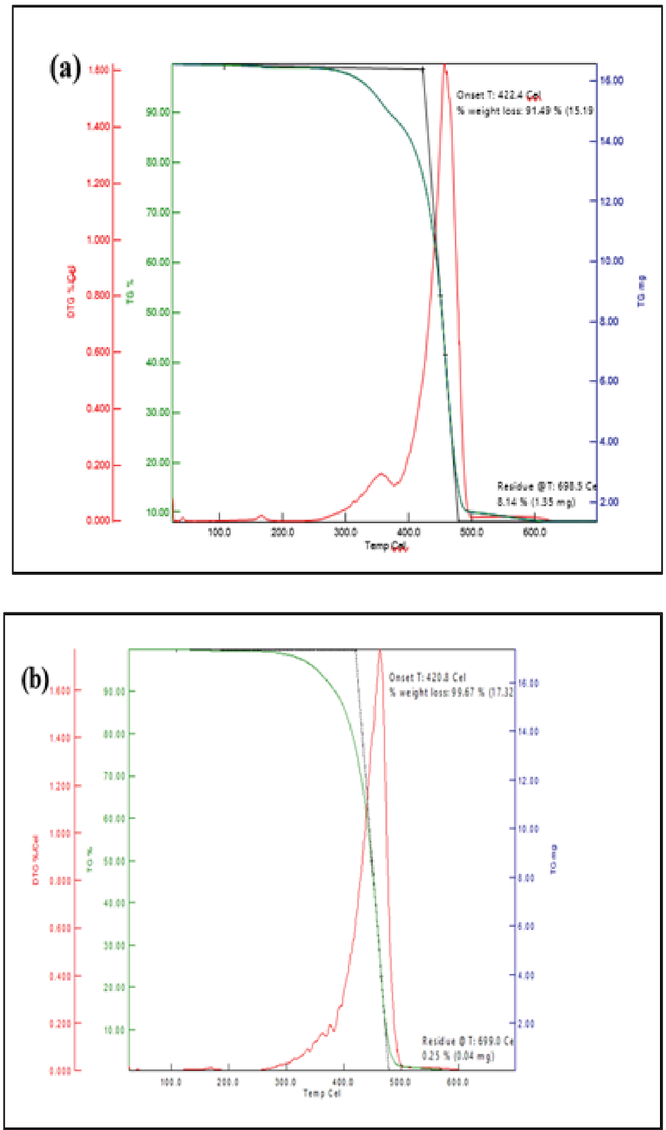


Figure 3 (a) TGA analysis of composite without MAPP, **(b)** TGA analysis of composite with 10g of MAPP

Figure 4 displays SEM images of the surfaces of the KF/PP and KF/PP/MAPP (10g). These SEM micrographs are used to examine how well KF/PP and the coupling agent (MAPP) get along. The existence of voids, pull-out of fibers at the surface, and particle fiber and matrix are all observable. Insufficient wettability or matrix/fiber adhesion is indicated by fiber pull-out. Based on Figure 4 (a) showed a smooth surface morphology of the KF/PP composite. Meanwhile, Figure 4 (b) showed several pull-out fibers. For Figure 4 (c) shows that the cross-sectional area of the KF/PP (20g/180g/0g) composite specimen has several holes. On the other hand, Figure 5 (a) showed an unsmooth surface morphology of the KF/PP/MAPP composite which the composite is a bit clumped. Additionally, although it included fewer kenaf fibers than KF/PP composite, the addition of a coupling agent (MAPP) to the KF/PP/MAPP (15g/180g/10g) composite revealed superior compatibility to that of the KF/PP composite (Figure 5 (c)). Additionally, it was established from SEM observations that the matrix and fiber had a solid interface binding. When using a modified polypropylene matrix, the covering ratio of fiber surfaces was somewhat increased. Plastic deformation of the matrix occurred before a full fracture, as seen in Figure. 5 (b).

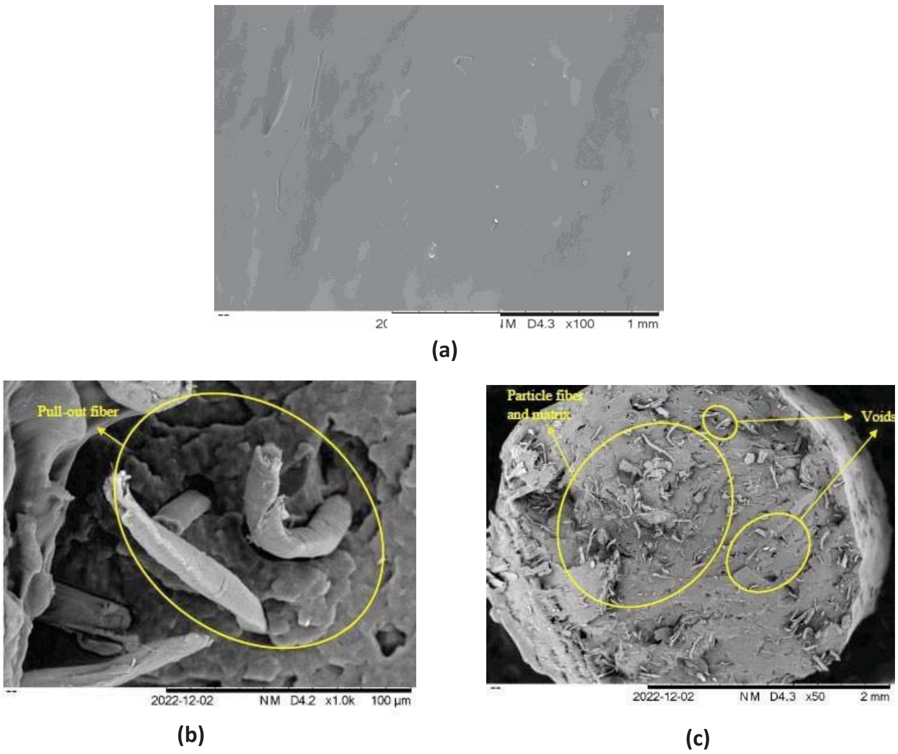
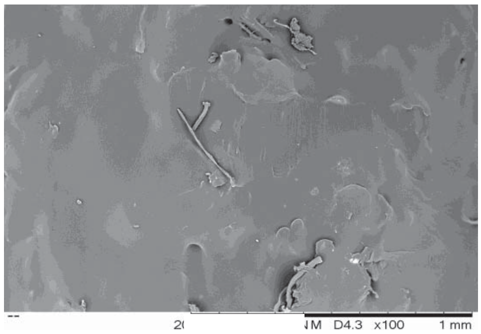
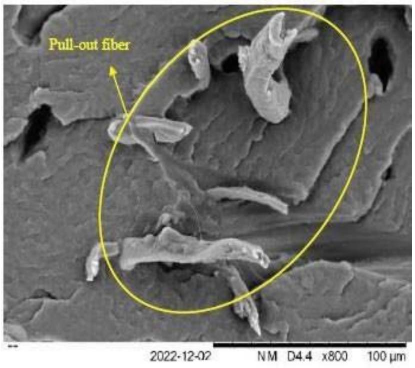


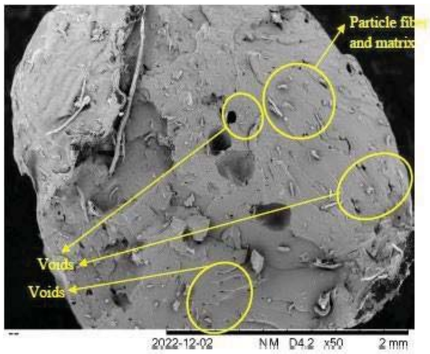
Figure 4 SEM micrographs of the composite sample without coupling agent using SEM of **(a)** KF/PP surface morphology **(b)** a clear view of pull-out fiber at (x1000) and **(c)** shows the particle fiber and matrix and voids from cross-sectional view at (x50)



(a)



(b)



(c)

Figure 5. SEM micrographs of the composite sample with coupling agent using SEM of **(a)** KF/PP/MAPP surface morphology **(b)** KF/PP/MAPP in cross-sectional view (pull-out fiber) (800) and **(c)** shows the particle fiber and matrix from cross-sectional view (x50).

4. Conclusion

This paper studies the investigation effect of a coupling agent (MAPP) on the mechanical, physical, thermal, and morphological properties of kenaf fiber/polypropylene composites. It was found that using MAPP has a significant effect on composite properties. This is proved by adding the MAPP, the tensile strength for 5g of MAPP and 10g of MAPP slightly increased. Besides, for the impact test, the comparison of KF/PP composites' impact resistance with and without a coupling agent (MAPP) shows MAPP improves the impact strength of the composite. Besides, for the water absorption test, it was discovered that the composite without adding a coupling agent (MAPP) absorbs more water. Meanwhile, the composite that was added with the coupling agent has low water absorption. Moreover, the increased interfacial adhesion of the kenaf fibers resulted in improved thermal stability of KF/PP composites when MAPP was added. Furthermore, it is also observed that the SEM micrograph illustrates a good adhesion between the polymer matrix and the reinforcement with coupling agent (MAPP), revealing greater compatibility in the composite material. However, this study has limitations as it primarily focuses on the effect of the coupling agent on composites. It is suggested that future research be conducted to optimize the composition ratios of kenaf fibres, polypropylene matrix, and coupling agent. This can aid in determining the optimal combination for superior mechanical properties and overall performance.

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