

MECHANICAL PERFORMANCE AND WATER ABSORPTION OF COCONUT FIBER/POLYETHYLENE WASTE BIOCOMPOSITE PREPARED VIA A CHEMICAL-FREE APPROACH

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Plastic wastes have caused serious environmental issues worldwide, and thus viable solutions for their replacement are now urgently needed. This work aimed to develop biocomposite materials based on polyethylene (PE) wastes as matrix reinforced with coconut fiber, without any additional chemical treatments, using extrusion and compression molding. The effects of polymer matrix type (high-density and low-density PE (HDPE and LDPE)) and fiber loading (5-15 wt%) on the mechanical properties and long-term water absorption behaviour of the materials were evaluated. Tensile strength results showed the optimum performance at 5 wt% fiber – of 16.6 MPa for the HDPE matrix and 7.3 MPa for the LDPE matrix, but flexural and impact strengths reduced with the fiber loading. An increasing trend of water absorption capacity was noted as a function of filler loading and of the water temperature during immersion, with a weight gain of up to 5%, following the trend: cold water > room temperature tap water > hot water. From the results, HDPE based biocomposites had better mechanical performance and lower water absorption capacity, compared with LDPE based biocomposites.

Keywords: recycled HDPE, recycled LDPE, natural fiber composite, mechanical property, physical property

INTRODUCTION

Natural fibers, such as kenaf, oil palm empty fruit bunch fiber, rice husk, pineapple leaves, coconut, *etc.*, have demonstrated their practical usage to replace synthetic fibers in engineering applications and construction industry as a step towards a greener environment.¹⁻³ In the last decades, natural fiber reinforced polymer composites have been widely studied due to their light weight, longer shelf-life, greater mechanical properties, environmental friendliness, as well as cheap and widely available raw materials.⁴⁻⁷ In order to reduce plastic wastes, researchers investigated biocomposites from wood sawdust and post-consumer plastics, prepared via melt-blending technology, and reported an increase of about 30% (up to 34.30 MPa) in mechanical strength of the biocomposite.⁴ The great thermal and wettability performances of the biocomposite supported it as a promising green and sustainable material.

Coconut fiber, the so-called coir fiber, is a common natural fiber that has been often considered as a reinforcing material, owing to its low cost, ease of availability and separation, biodegradability and recyclability.⁸ Coconut is abundant in tropical regions, being produced in more than 90 countries, in quantities of over 59 million tonnes per year.⁹ Approximately 12.75 tonnes of coconut wastes are produced annually from husks, spathes, peduncles, petioles and leaves.¹ The main constituents of coconut fiber are cellulose (42 wt%), hemicelluloses (0.25 wt%), lignin (47 wt%), moisture (5 wt%), pectin (3 wt%) and ashes (2 wt%).¹⁰ Bazan *et al.*¹¹ claimed that the high contents of lignin and cellulose make the coconut fiber elastic, strong and more durable. When compared with other vegetable fibers, coconut fibers have a low cellulose content and a high microfibril angle, leading to great elongations, but lower tensile strengths.¹² Coconut fiber generally has a density of 1.1-1.5 g/cm³, tensile strength of 105-593 MPa and Young's modulus of 2-8 GPa.¹³ Compared to other natural fibers,

the coconut fiber is highly resistant to moisture and heat, besides having non-toxic properties, good acoustic resistance, and being resistant to fungal and microbial degradation. Owing to its high hardness and hard-wearing quality, coconut fiber has attracted a great interest in the production of composites for the automotive industry.¹⁴

Polyethylene (PE) is a thermoplastic polymer, a long-chain hydrocarbon compound formed from ethylene monomer through polymerization.¹⁴ It is worth noting that the light PE is an odor-free material, with good resistance to water absorption, as well as great chemical, electrical and mechanical properties. PE has been claimed mostly as an exceedingly demanded plastic, and yet, it is urgently needed to search for new alternative materials because of its slow degradation issue, requiring about a thousand years.¹ The slow rate of degradation is related to its durability and chemical bonds that resist the natural degradation process.¹¹ As a result of their longevity and low recycling rate, most plastics end up in landfills or into the ocean, consequently affecting the ecosystems.¹⁵ Therefore, while the non-biodegradability of PE would be the main obstacle in its industrial production, utilizing recycled PE can be a good option for decreasing its carbon footprint.¹

Dharmaratne and coresearchers¹⁴ have reported that their fabricated PE waste reinforced coconut fiber biocomposite, with tensile strength and bending strength up to 6.75 N/mm² and 29.85 N/mm², respectively, could be utilized as a sustainable construction material. In order to produce a new engineering material, the heat and corrosion resistance, as well as the mechanical and electrical properties of the material would be the main consideration; the mechanical performance being usually the focus of preliminary studies in many research developments of new materials. Akter *et al.*¹ analysed for the potential of HDPE and coconut fiber biocomposites as new engineering materials, and found that the great distribution of fiber and interfacial bonding between fiber and matrix improved the bending and load bearing capacity. This can be supported by a study on HDPE/coconut spathe fiber, where improved mechanical performance was reported for 5 wt% and 15 wt% fiber loadings, but decreased when 10 wt% and 20 wt% of fiber was incorporated.¹ Bukar *et al.*⁸ developed coconut fiber reinforced LDPE composites, after extracting the fiber from coconut husks via the water retting process, followed by cleaning and dewaxing processes. The investigation showed the optimum combined tensile and impact strengths were obtained with 20-30 wt% fiber loadings. Ihueze's research group¹⁶ reported that a smaller particle size of coconut fiber improved the hardness of HDPE based composites, but had no effect on their elastic modulus.

Numerous research works reported in the literature have developed natural fiber-reinforced composites or wood-plastic composites using coupling agents or surface modification to enhance interfacial interaction.^{4,8,17,18} However, the question arises: in the absence of chemical treatment, how much can the mechanical properties be improved by incorporating coconut fiber into a PE matrix? Water absorption is another important aspect investigated in these biocomposites. However, limited studies have been conducted on immersion in various water media beyond distilled water or normal tap water.^{8,19,20} Additionally, in most cases, composite materials typically exhibit excellent properties, but involve high production costs.⁸ Nevertheless, it might not be feasible to solve the waste disposal issue through high-cost production, involving expensive chemicals, complex cleaning and refining processes, even though the agro-industrial waste itself is inexpensive. Taking into account both industrial and environmental perspectives, as well as the various advantages of natural fiber-reinforced composites, this study investigated the impact of different coconut fiber loadings (ranging from 0 to 15 wt%) on two types of polymer matrices (HDPE and LDPE). Importantly, no chemicals, such as cleaning reagents, compatibilizer or coupling agents, were used in this investigation. In this study, the mechanical performance, including tensile, flexural and impact properties, as well as long-term water absorption in different temperature media (room temperature tap water, cold and hot water), were analysed in detail.

EXPERIMENTAL

Materials

In this research work, all raw materials were obtained from recycled sources. Two types of polymer matrices were used, namely, high-density polyethylene (HDPE) and low-density polyethylene (LDPE); they were obtained from a commercial chemical container and a film roll purchased from the market (as shown in Fig. 1), respectively. Meanwhile, the reinforcing filler used in the study was coconut fiber.

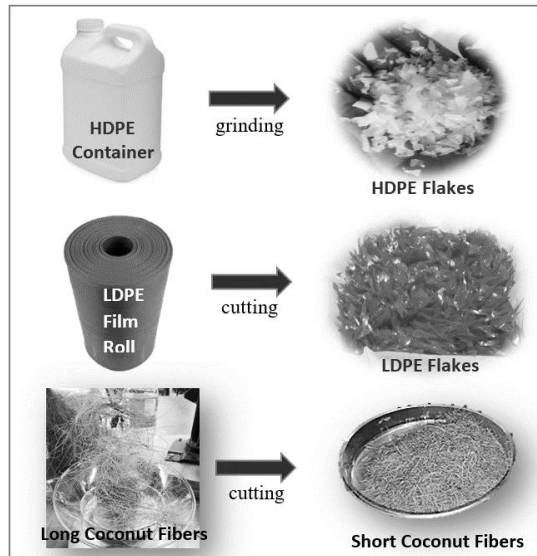


Figure 1: Preparation of HDPE and LDPE flakes, and short coconut fibers

Preparation of PE flakes and short coconut fibers

Before mixing, the plastic and coconut fibers need to be prepared. As shown in Figure 1, the HDPE chemical container was ground using a plastic shredder, whereas the flexible LDPE film roll was cut into PE flakes. The HDPE chemical container was cleaned with soap before grinding. Meanwhile, the long coconut fibers were cut into a 1-2 cm length of short fibers. The PE flakes and short coconut fibers were oven-dried at 90 °C overnight prior to the compounding process.

Preparation of PE/coconut fiber biocomposites

The biocomposites of HDPE/coconut fiber and LDPE/coconut fiber were separately prepared via extrusion and compression molding. The melt-blending of PE flakes and coconut fiber was performed using a co-rotating twin-screw extruder (model: Thermo Prisma TSE 16PC, screw diameter of 16 mm and length/diameter ratio of 24), at the following temperature regime: 110/115/120/115 °C and the screw rotating speed of 25 rpm. The coconut fiber loading was varied as follows: 5, 10 and 15 wt%. The extruded compounds were then subjected to hot and cold pressing (hot pressing at 120 °C), and 1000 psi to mold the composite panels for characterization. During compression molding, the hot-pressing cycle included 3 min pre-heating, 3 min venting and 4 min full pressing; whereas the cool pressing was fixed at 4 min.

Characterization

The tensile, flexural and impact properties of compression molded specimens were evaluated according to ASTM D638-03 (type I, with 3 mm thickness dumbbell specimen), ASTM D790-03 (sample dimension: 127.0 × 12.7 × 3.0 mm³) and ASTM D 256 (sample dimension of 63.5 × 12.7 × 3.0 mm³), respectively. The first two mechanical tests were conducted utilizing a Testometric M350-10CT universal machine, operating at a crosshead speed of 5 mm/min. The impact test was conducted using a Ray-Ran Impact tester, with a velocity of 3.46 m/s and a load of 0.452 kg. Five samples were run for each mechanical testing to obtain the average value.

Based on ASTM D570-98, the water absorption test was performed on the specimen with the dimension of 76.2 × 25.4 × 3.2 mm³. Before testing, the specimens were heated in an oven and weighed (W_o) using a digital weighing balance at 0.01 g precision. The specimens were immersed in water of various temperatures, namely, room temperature tap water (maintained at ambient conditions, 20-24 °C), cold water (stored in the refrigerator at temperatures in the range of 4-8 °C) and hot water (maintained in an oven at a controlled temperature of 105 °C) for a month. The immersed specimens were removed periodically, wiped with tissue and their weights were measured (W_t). The percentages of water absorption (WA%) were determined using the following equation: $WA\% = (W_t - W_o)/W_o \times 100$, where W_o is the oven-dried (initial) weight and W_t is the specimen weight after a certain water immersion time.

Statistical comparisons of the measured data for mechanical and water absorption properties were performed using two-way analysis of variance (ANOVA), with the aid of Data Analysis ToolPak in MS Excel in order to determine the effects of polymer type (HDPE and LDPE) and coconut fiber loading, at the 5% significance level.

RESULTS AND DISCUSSION

Tensile properties

The tensile results of strength, Young's modulus and strain at break of HDPE and LDPE biocomposites reinforced with various loadings of coconut fibers are shown in Figure 2. In general, the HDPE system had higher tensile properties, as compared to the LDPE system. The neat HDPE and LDPE exhibited the tensile strength of 16.5 MPa and 7.1 MPa, Young's modulus of 330.3 MPa and 109.5 MPa, and strain at break of 16.9% and 13.6%, respectively. This result was expected; as reported by Salim *et al.*,²¹ HDPE and LDPE have several different properties and uses, although both are thermoplastic ethylene polymers. In terms of molecular structure, as compared to HDPE, LDPE has a higher degree of chain branching (where the polymeric chains are bound to secondary chains by substituting an atom in the primary chain with a monomer group), thereby weakening the polymer intermolecular forces (as shown in Scheme 1). In summary, close packing (high density) in HDPE results in stronger intermolecular forces. This is why, HDPE exhibited a higher tensile strength due to the higher strength-to-density ratio than that of LDPE.

In the presence of coconut fiber, the tensile strength and modulus of biocomposites were improved at a certain fiber loading. As depicted in Figure 2 (a), the addition of 5 wt% coconut fiber resulted in a slight improvement in tensile strength, where 16.6 MPa and 7.3 MPa were recorded for HDPE and LDPE based biocomposites, respectively. However, when further increasing the coconut fiber loading up to 15 wt%, the tensile strength decreased. These findings are reasonable, because there is no fiber pretreatment or coupling used herein, in comparison with a previous study on coconut shell particle composites, where the filler content used was at 10 wt% only, showing improvements in tensile and flexural strengths.²² This result can be correlated to the volume fractions, degree of matrix–fiber adhesion, level of filler dispersion and presence of surface related defects.⁸ The early increase in tensile strength may be explained by acceptable physical interaction between the coconut fiber and the PE matrix, while the latter decrease in tensile strength could be caused by poor wettability, which caused a weak interface as a result of the formation of voids. The declined strength is expected as the presence of fibers could generate points of stress concentration, thereby leading to premature fraction of the polymer matrix, as supported by de Almeida *et al.*²³ At the same loading, an improvement in Young's modulus of both composites was noticed: of 387.1 MPa and 151.3 MPa, with increments of 17% and 38%, respectively. These values decreased for the 10 wt% loading (the lowest values) and again increased for 15 wt% fiber composites. According to Akter *et al.*,¹ Young's modulus is a measurement indicator for the stiffness of a material. Therefore, based on this result, the composite with 10 wt% fiber is less stiff than other compositions.

The strain at break results (Fig. 2 (c)) of the composites showed a significantly decreasing trend when the coconut fiber loading increased from 0 to 15 wt%, irrespective of the matrix type. This is a common trend for most natural fiber composites, as similar findings have been reported for sugarcane-derived green HDPE/coconut fiber composites with the same fiber content.²³

Flexural properties

Figure 3 shows the effect of fiber loading on the flexural strength and modulus of composites based on HDPE and LDPE. When coconut fiber was added into polymer matrix composites, the flexural strength was found to decrease and the flexural modulus increased with increasing fiber content up to 15 wt%. The result of the decreased flexural strength differs from the increment of tensile strength at 5 wt%, which could be explained by the different fracture mode. The failure of flexural motion is due to the results of two concurrent stresses, where the extension occurred at the convex side and the compression at the concave side of the specimen; on the contrary, the tensile fracture is due to the one-direction extension.²⁴ In this respect, the interaction between the melted PE matrix and the coconut fiber is insufficient to hold the flexural stress since no coupling agent was used in these systems. The increase in flexural modulus can be explained by the stiffening effect, which is caused by the decreased polymeric chain mobility.¹⁰ Comparing HDPE and LDPE matrices, similar trends are observed as in the case of the tensile properties.

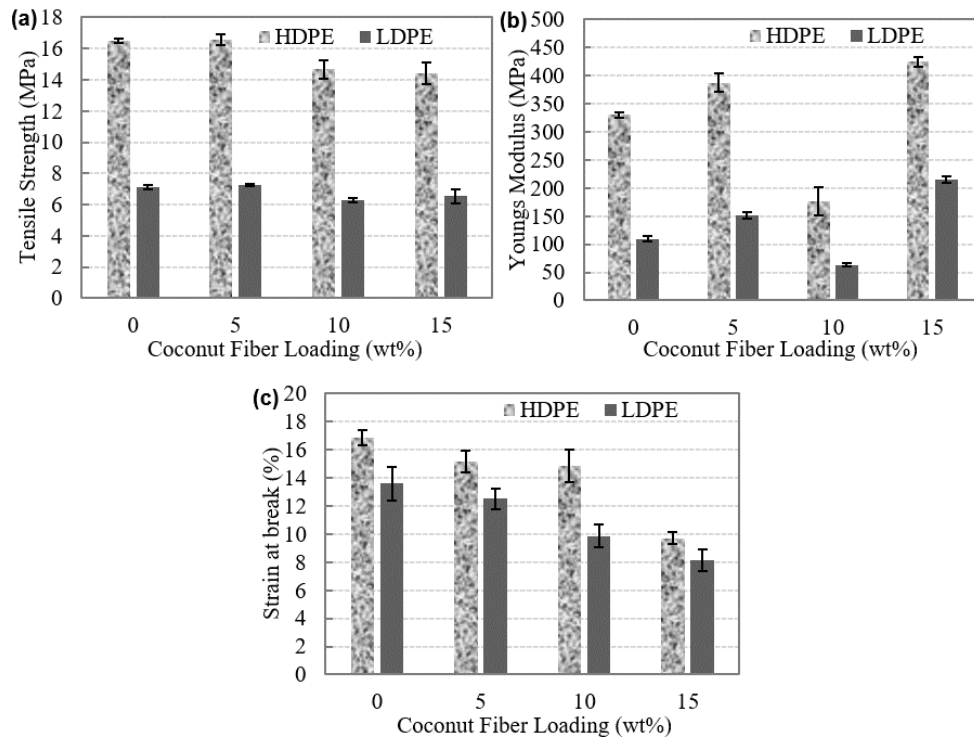
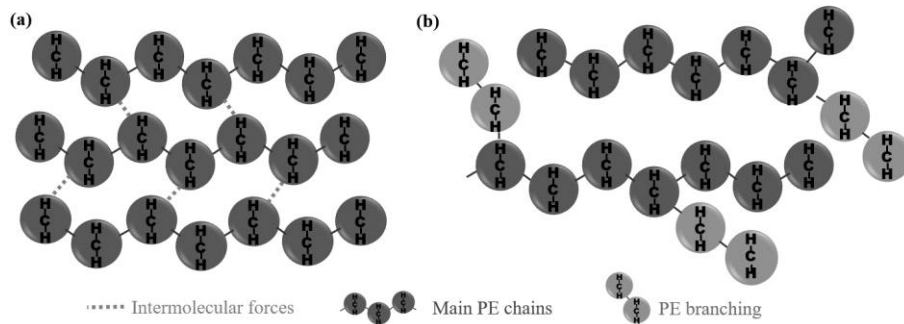


Figure 2: Tensile properties of coconut fiber biocomposites based on HDPE and LDPE



Scheme 1: Molecular structures of (a) HDPE (with stronger intermolecular forces) and (b) LDPE

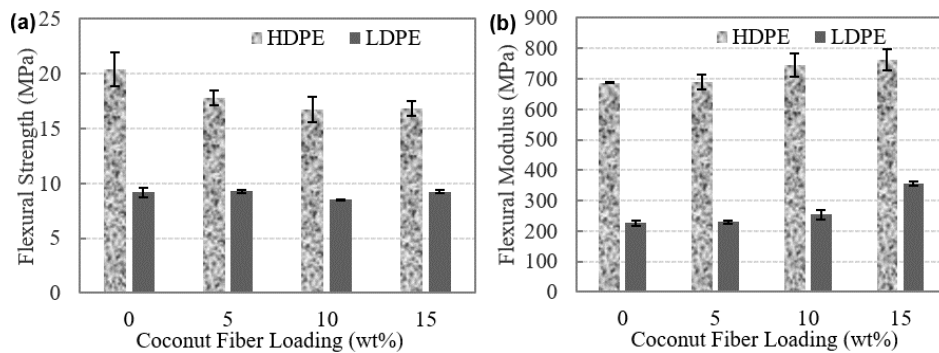


Figure 3: Flexural properties of coconut fiber biocomposites based on HDPE and LDPE

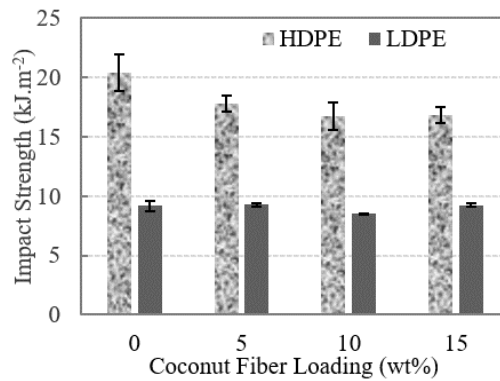


Figure 4: Impact strength of coconut fiber biocomposites based on HDPE and LDPE

Impact properties

Impact toughness measures the ability of a polymer material to withstand the load imposed upon being struck by an object at high velocity via measurement of energy required to propagate the induced crack across the material.²⁵ As can be seen from Figure 4, the impact strength exhibited a similar trend to that of the strain at break. It can be observed that HDPE and the corresponding composites had greater impact strengths than LDPE and its composites. This can be correlated with higher modulus (as shown in Figs. 2 (b) and 3 (b)) and higher density of HDPE than that of LDPE, in which the high weight and narrow molecular distribution could generally improve the impact resistance.¹⁹ The obviously reduced impact strength with respect to fiber loading in both composite systems indicates the increment of the materials' brittleness by the addition of coconut fiber.¹

Water absorption

Figure 5 displays the percentage of water absorption measured via weight gain for the HDPE and LDPE biocomposites immersed in room temperature tap water, cold water and hot water. In general, it can be observed that the water absorption rate increased speedily in the first few days (~100 hours) and it slowed down after 8 days (192 hours),^{1,4,18,26} irrespective of the temperature of the immersion medium. The weight gain caused by the water absorption process continued with the prolonged immersion period until the specimen attained an equilibrium state (saturation) after 4 weeks (672 hours). Both neat HDPE and LDPE specimens exhibited the least water absorption percentages, which were approximately below 2%. The PE polymer is hydrophobic with non-polar functionality and thus absorbed much less water (a negligible amount).⁴ When the coconut fiber was added into the polymer matrix, it generally led to an increment in water absorption with the fiber loadings. This was expected as the coconut fiber is a lignocellulosic material, which can absorb more water contents due to the presence of polar hydroxyl groups (hydrophilic nature).^{1,8} Comparing different fiber loadings, it can be seen that a significantly high water absorption rate was recorded for the 5 wt% loading, and a lower increment – for the 10-15 wt% loadings for all the systems. At this loading, the highest water absorption was achieved at approximately 3-5%, which is in agreement with a previously reported study.⁸ This could be attributed to the fiber swelling and the consequent formation of microcracks in the polymeric matrix, which resulted in the largest transport of water molecules through the matrix-fiber interface.⁸

When observing the effects of the polymer matrix type, in Figure 5, it is obvious that the LDPE based biocomposites (solid lines) exhibited slightly greater water absorption percentages, as compared to those of the HDPE matrix based ones (dashed lines). A similar finding was reported by Gulitah and Liew,²⁷ and Murat *et al.*¹⁹ As explained by Murat *et al.*,¹⁹ this could be related to the microstructure of the LDPE, which has more pores and voids as the polymer chains are branched in LDPE; whereas HDPE is much more linear, with a crystalline structure, as shown in Scheme 1.

By comparing the water absorption results obtained for different temperatures of the immersion media, the three graphs in Figure 5 reveal an increasing trend, as follows: hot water > room temperature tap water > cold water. For instance, at 15 wt% coconut fiber, the weight gains recorded were (i) 2.5% and 3.9% for cold water immersion, (ii) 3% and 4.2% for tap water immersion, and (iii) 3.9% and 4.5% for hot water immersion for HDPE and LDPE composites, respectively. These results are similar to those reported for other natural fiber composites, where room temperature immersion

caused almost doubled the weight gain in cold water immersion (0 °C), and the hot water (50 and 75 °C) immersion caused an about 20-30% higher weight gain compared to that at room temperature.²⁸ The highest water absorption of biocomposites in hot water can be explained by the fact that increasing temperatures will reduce the viscosity of water and thus possibly induce the delamination process.²⁹ According to Kamau-Devers *et al.*,³⁰ the high water temperature immersion condition will also cause enhanced crazing and formation of cracking, thereby increasing the equilibrium moisture content (maximum water content at saturation level).

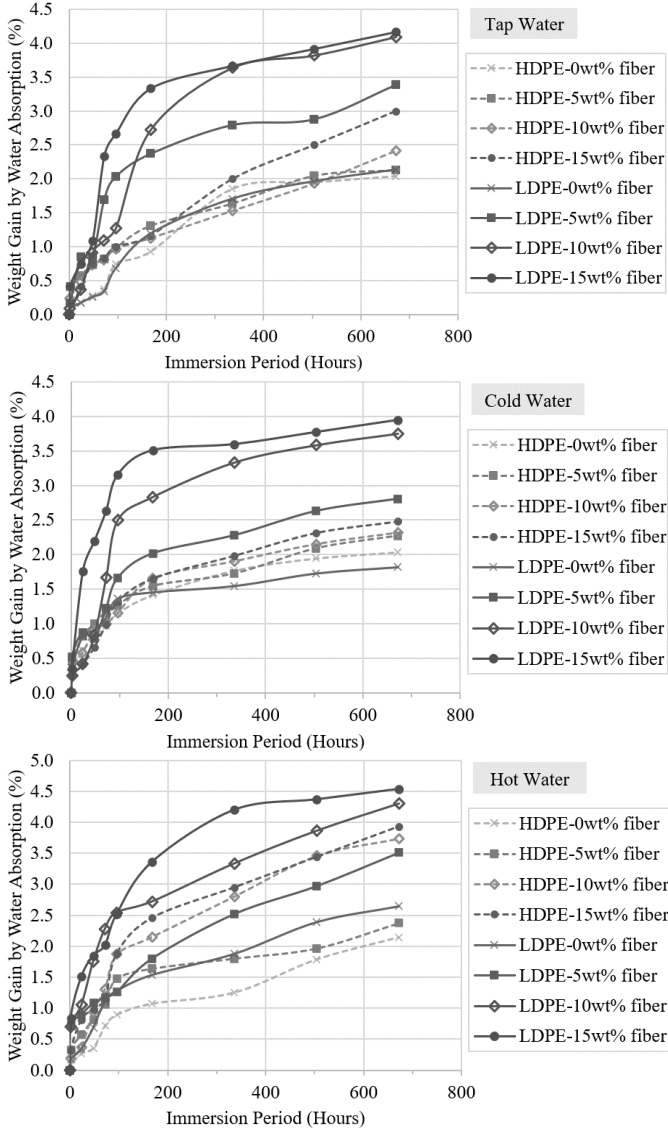


Figure 5: Weight gain by water absorption of HDPE and LDPE biocomposites reinforced with coconut fiber in immersion media of different temperatures

Statistical significance determination for experimental results based on ANOVA

Two-way ANOVA of the tensile, flexural, impact and water absorption properties data as a function of polymer matrix type and fiber loading was performed and the results are provided in Table 1. When *p*-value < 0.05 (which is usually aligned with F value > F critical), it means there is a significant statistical difference between the data, with a confidence level of 95%. Based on the results in Table 1, *p*-values below 0.05 (the numbers in bold) were obtained for the dependence of tensile and flexural properties, and water absorption in hot water on polymer matrix type; and the dependence of Young’s modulus, strain at break, impact strength and water absorption in hot water on the fiber loading. These results confirmed the statistically significant difference between the investigated properties of the composites with the two polymer matrices and various fiber loadings. From this

statistical analysis, it can be simply evidenced that the polymer matrix type (HDPE and LDPE) was a greater influence factor as a significant source of variation in terms of mechanical results, except for impact strength, compared with fiber loading. The loading of coconut fiber used in this study is considered low, therefore, it shows no statistically significant effect for the water absorption in room temperature tap water.

Table 1
Two-way ANOVA test results on the mechanical and water absorption properties

Properties	Polymer type			Fiber loading		
	F value	F critical	p-value	F value	F critical	p-value
Tensile strength	597.045	10.128	0.0002	4.944	9.277	0.111
Young modulus	48.954	10.128	0.006	9.399	9.277	0.049
Strain at break	19.391	10.128	0.022	14.520	9.277	0.028
Flexural strength	121.914	10.128	0.002	1.404	9.277	0.393
Flexural modulus	638.195	10.128	0.0001	6.947	9.277	0.073
Impact strength	7.236	10.128	0.074	24.940	9.277	0.013
Water absorption (tap water)	9.809	10.128	0.052	3.778	9.277	0.152
Water absorption (cold water)	3.978	10.128	0.140	2.036	9.277	0.287
Water absorption (hot water)	23.003	10.128	0.0172	35.758	9.277	0.008

Note: F value – mean between/within groups variance, F critical – critical F value based on F distribution, and p-value – probability from 0 to 1

Table 2
Production, recycling percentage, energy and prices of virgin and recycled PE (HDPE, LDPE)

Plastics	Virgin		Recycled	
	HDPE	LDPE	HDPE	LDPE
Production (%)	17 ³¹	23 ³¹	N/A	N/A
Post-consumer material recycled (%)	10.3 ³¹	5.3 ³¹	N/A	N/A
Embodied energy (MJ/kg)	80 ³¹	68 ³¹	40 ³¹	50 ³¹
Price (\$/kg)	1.9-2.0 ³²	N/A	0.84-0.97 ³²	N/A
Price of scrap (prior to processing) (\$/kg)	N/A	N/A	0.45-0.65 ²⁶	0.10-0.45 ²⁶

Overview of HDPE and LDPE market

The production and recycling percentages of HDPE and LDPE shown in Table 2 generally indicate the still high demand of these plastic products and yet their recycling is low. The recycling of post-consumer HDPE (~10.3%, based on 17% production) is found to be much higher than that of LDPE (~5.3%, based on ~23% production). This may be due to the higher recyclability of HDPE, which is easier to transport and run through the recycling equipment. In contrast, LDPE is softer and can be easily stacked in the recycling machinery. There is a possible reason that HDPE and HDPE based biocomposites would have better mechanical performance, as shown in Figures 2-4, which makes them suitable for more applications. However, the price of HDPE is found to be higher than that of LDPE. Certainly, only about half of the virgin plastics are recycled, while recycling can help save up to 50% energy. Therefore, the more research efforts on plastics recycling methods, including the performance and cost analysis, as well as formulation design, are necessary.

CONCLUSION

HDPE and LDPE biocomposites reinforced with 5-15 wt% coconut fiber were prepared via the melt-blending method. Mechanical and water absorption properties were investigated. Both HDPE and LDPE based biocomposites, containing 5 wt% fiber, exhibited the highest tensile strength in comparison with their own neat polymer matrix and other fiber loadings. Except for modulus, other mechanical properties and water absorption resistance of biocomposites were reduced upon incorporation of the coconut fiber. However, the maximum weight gain by water absorption of 4.5% is

still considered acceptable in many industries. These results indicate that further research on treatment or modification of polyethylene waste reinforced with coconut fiber is necessary to attain the performance levels required for potential applications in the automotive and construction fields.

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