Heliyon 8 (2022) e09287

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Research article

CellPress

Fabrication and investigation of the physico-mechanical properties of Jute-PALF reinforced LLDPE hybrid composites: Effect of gamma irradiation



Helivon

Habibur Rahman^{a,*}, Farjana Yeasmin^b, Tarikul Islam^a, Mahmudul Hasan^c, Muhamad Borhan Uddin^d, Ruhul Amin Khan^d

^a Department of Textile Engineering, Jashore University of Science and Technology (JUST), Jashore-7408, Bangladesh

^b Department of Agro Product Processing Technology, Jashore University of Science and Technology (JUST), Jashore-7408, Bangladesh

^c Department of Textile Engineering, Dhaka University of Engineering and Technology (DUET), Gazipur, Bangladesh

^d Atomic Energy Research Establishment (AERE), Savar, Dhaka, Bangladesh

ARTICLE INFO

Keywords: Hybrid composites Jute PALF LLDPE Physico-mechanical properties Gamma irradiation

ABSTRACT

The hybridization effect of agro-waste pineapple leaf fibre (PALF) and jute fibre as reinforcement in linear low-density polyethylene (LLDPE) composites was investigated in this work. The samples were fabricated by using the heat press compression moulding. The effect of gamma irradiation on composite physico-mechanical properties was also investigated in order to determine the best gamma dose among 2.50, 5.00, 7.50, and 10.00 kGy. The composite sample containing 40% PALF and 60% jute (with a total weight of 50% fibres) demonstrated the most feasible tensile strength (33.36 \pm 0.59 MPa), tensile modulus (1494.41 \pm 10.94 MPa), elongation at break (50.92 \pm 0.77%), bending strength (82.58 \pm 0.49 MPa), bending modulus (4932.46 \pm 96.12 MPa), and impact strength (34.38 \pm 0.42 kJ/m²) at 7.50 kGy irradiation. Thermogravimetric analysis (TGA) determined the thermal performance of the samples. Scanning electron microscopy (SEM) images at the tensile fracture surfaces of composites revealed the interfacial interaction between reinforcement fibres and matrix.

1. Introduction

Natural fibre-reinforced polymer composites are gaining popularity in both structural and non-structural applications. All disciplines of materials engineering research are paying close attention to the widespread replacement of expensive synthetic glass and carbon fibres [1, 2]. PALF, jute, sisal, and hemp are examples of lignocellulosic plant fibres that are usually applied as reinforcement in polymer matrix composites. These fibres have outstanding physico-mechanical features such as renewability, non-abrasiveness, ease of handling and processing, better energy recovery, great thermal insulation, increased cellulose percentage and aspect ratio, higher specific tensile and flexural capabilities, and so on [2, 3]. Among them, jute has various distinguishing characteristics, including adequate mechanical strength, lightweight, low cost, environmental friendliness, and widespread availability [4, 5]. This is composed of around 60% cellulose, 20.4% hemicellulose, 0.2% pectin, 13% lignin, 0.5% wax and other minor ingredients [6]. Jute cell walls and fibrils act like an amorphous substance due to the presence of lignin [7]. As a result, jute has good heat, noise, chemical, and UV radiation resistance [4, 5]. PALF is made up of cellulose (67.12-83%), hemicellulose (15-20%), lignin (4.4-15.4%), pectin (1.1-4%), fat and wax (3.2-7%), ash (0.9-6%), and extractives (3.83-0.87%) [8]. Each pineapple plant generates about 2–3 kg of leaves during harvesting. This is frequently referred to as agricultural waste. This huge amount of post-harvest wastage (90-100 tons per hectare) can be utilized by extracting the fibres [3]. PALF shows more advanced tensile properties and fineness than many natural fibres [9]. Because of the high percentage of cellulose content and low microfibrillar angle, researchers have made tremendous efforts to use agro-waste PALF in various value-added technical applications [8]. LLDPE is highly processable, has a high recyclability, and is compatible with natural fibres [10]. It shows remarkable rheological properties, softening temperature (135 °C), specific gravity (0.92), density (0.905 g/cm³), and is more stiffer than branched polymers due to its non-polarity [11]. This is also a cost-effective polyolefin matrix for manufacturing natural fibre-reinforced composites [12]. The hybrid composites are produced by combining two or more reinforcements into a particular matrix by maintaining the complementary equilibrium among the intrinsic attributes of elements with a view to achieve better performances [13]. Natural fibres reinforced hybrid composites are gaining more attention

https://doi.org/10.1016/j.heliyon.2022.e09287

Received 4 February 2021; Received in revised form 31 May 2021; Accepted 12 April 2022

^{*} Corresponding author. E-mail address: habib te@just.edu.bd (H. Rahman).

^{2405-8440/© 2022} The Author(s). Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

with researchers for improved physico-mechanical properties [9]. Lignocellulose-based natural fibre reinforced hybrid composites are used widely in automobiles, aerospace, marine, packaging, and industrial applications [4, 13, 14]. The addition of a small fraction of glass fibres with PALF in polyester composites demonstrated the hybridization effect in the enhancement of mechanical performances [15]. PALF and kenaf fibres reinforced high-density polyethylene (HDPE) composite showed improved tensile, flexural, and impact strength [16]. However, the hybridization effect of jute-glass-polypropylene [17], sisal-glass-polypropylene [18], flax-glass-polypropylene [19], jute-banana fibre-e-poxy [20], kenaf-PALF-HDPE [21], and other fibres has been studied. For improved physico-mechanical properties, hybridization of PALF with jute fibre in LLDPE composites can be investigated.

Gamma irradiation is the application of ionising radiation to a specific body with the highest energy and shortest wavelength gamma rays. It can improve the reinforcing fiber's compatibility with the polymer matrix [22]. As a result, the physico-mechanical properties (tensile strength and modulus, bending or flexural strength and modulus, impact strength, density, dimensional stability, and so on) and thermo-chemical properties (chain scission as well as cross-linking within matrix and reinforcement, interfacial polarisation and orientation, thermal stability, hydrophobicity, glass transition temperature, and so on) of natural fibre reinforced polymer composites increased up to a specific dose [2, 11, 23]. This method is less expensive, easier to use, and takes less time to expose than x-rays or ultraviolet radiation [22, 23, 24, 25]. Various researchers have made remarkable attempts to assess the effects of gamma irradiation on various natural fibre reinforced polymer composites, such as jute-polyester [22], bagasse fibre-waste polypropylene [26], jute-polypropylene [27], PAN-carbon fibre [28], jute-LDPE [29], silk-polypropylene/natural rubber [30], and so on. At 7.5 kGy gamma irradiation, another investigation found that 1:1 (weight fraction) untreated PALF-LDPE composite had the best physico-mechanical properties [23]. There hasn't been any research into the effects of gamma irradiation on PALF and jute-reinforced LLDPE hybrid composites. As a result, the impact of gamma irradiation on various physico-mechanical properties of PALF-jute reinforced LLDPE hybrid composites can be assessed in order to find the best fibre-matrix ratio at a certain gamma dose.

2. Materials and methods

2.1. Raw materials

PALF (*Ananas comosus*), white jute (*Corchorus capsularis*), and LLDPE granules were collected from Madhupur, Tangail, Bangladesh; Bangladesh Jute Research Institute (BJRI); and Polyolefin Company Ltd. Singapore; respectively. The fibres were opened by hand comb and cut into 12.7–15.2 cm (5–6 inches) length.

2.1.1. Tensile properties of fibres

Tensile tests on fibres were carried out in accordance with ASTM D3822 (2020) on a universal testing machine (James Heal Titan SN 1410 series) with a load cell of 200 N, a gauge length of 25 mm, and a cross-head speed of 10 mm/min.

2.1.2. LLDPE sheet preparation and characterization

The LLDPE granules were placed between two shims of stainless-steel platens bounded by a forma (300 mm \times 300 mm). LLDPE sheets were

prepared in a heat press machine (Carver, Inc., USA, Model 3925) at 115 $^{\circ}$ C with a 5.00 bar consolidation pressure. Then, the sheets were released from the heated platens of the machine and allowed to cool naturally for 20 min at a temperature of 25 $^{\circ}$ C. The thickness of the prepared sheets was 0.50–1.50 mm. The density of LLDPE sheet was measured according to ASTM D792 (2020). The tensile characteristics of LLDPE sheets were tested according to ASTM D638 (2002) using a universal testing machine (Model: H50KS-0404, HOUNSFIELD series S, UK).

2.1.3. Density of fibres

A digital microscope was used to determine the single fibre diameter (mm) of a specific length. The final diameter of a single fibre was calculated using the average values of 100 fibres. On the basis of masses and lengths, the linear densities of fibres were measured according to ASTM D2130 (2013). The values of average linear density and diameter were put in the below Eq. (1) to estimate the volume density (ρ_f) of fibres [31].

$$\rho_{\rm f} = \frac{M}{\frac{\pi d^2 l}{4}} \tag{1}$$

where, M represents the mass of fibre, d represents the average diameter and l represents the length of a fibre.

2.1.4. Moisture regain percentage of fibres

The moisture regain percentage of fibres was determined according to ASTM D2495 (2019). The fibre samples were immersed in distilled water at room temperature (25 °C). Those fibres were then kept in a glass beaker containing 150 ml of distilled water for 4 (four) hours. Finally, the fibres were removed from the beaker and wiped properly. The wiped samples were then reweighted after drying for 10 min at 100 °C in a drying oven. The moisture regain percentage was calculated by means of the following Eq. (2).

Moisture regain% =
$$\frac{Wt - W_0}{W_0} \times 100$$
 (2)

where, W_t represents the weight of the sample after immersion in water and W_o represents the weight of the sample before immersion.

The physico-mechanical characteristics of jute fibre, PALF, and LLDPE are presented in Table 1.

2.2. Composite fabrication

Six composite samples were made with various weight percentages of reinforced fibres (jute and PALF) and a 50% LLDPE matrix. In composites, the overall weight percentage of fibres was almost 50%. Table 2 shows the percentages of LLDPE, PALF, and jute fibres utilised to make the composite samples. For fabricating the jute-PALF hybrid composite, the LLDPE sheets were sliced to the preferred dimension for maintaining the weight fraction. To remove any remaining moisture, the jute and PALF were dried in a drier at 100 °C for 60 min. After that, the PALF and jute fibres were combined according to the percentages listed in Table 2. The PALF and jute fibre blended layer had a thickness of 0.58 \pm 0.03 mm. The three LLDPE sheets with a feasible thickness (0.60 \pm 0.04 mm) were initially weighted (50%) before the fibres (50%) were taken according to the amount given in Table 2. Two layers of randomly oriented jute-PALF blended plies were sandwiched between three layers of LLDPE sheets to

Table 1. Physico-mechanical characteristics of jute fibre, PALF and LLDPE.								
Types	Tensile strength (MPa)	Linear density (tex)	Tenacity (g/tex)	Breaking elongation (%)	Young's modulus (MPa)	Density (g/cm ³)	Moisture regain (%)	
Jute	653.21 ± 6.31	2.02 ± 0.03	26.62 ± 0.21	1.04 ± 0.01	14783.12 ± 36.14	1.34 ± 0.01	13.70 ± 0.12	
PALF	1572.32 ± 47.43	2.50 ± 0.06	100.08 ± 0.48	2.69 ± 0.04	6260.64 ± 47.39	1.53 ± 0.02	12.00 ± 0.11	
LLDPE	8.61 ± 0.13	-	-	136.46 ± 0.56	146.72 ± 5.28	$\textbf{0.918} \pm \textbf{0.01}$	-	

Table 2. Jute-PALF reinforcement and LLDPE matrix ratio in composite samp	les
---	-----

Sample ID	LLDPE (wt.% of composite)	Fibres (50 wt.% of composite)		
		PALF (wt.% of fibres)	Jute (wt.% of fibres)	
LP8J2	50	80	20	
LP7J3	50	70	30	
LP6J4	50	60	40	
LP5J5	50	50	50	
LP4J6	50	40	60	
LP3J7	50	30	70	

produce these samples. The sandwich was heated at 160 °C for 5.00 min to soften the LLDPE, then pressed at 6.00 bar pressure for 5.00 min (Carver, Inc., USA, Model 3856). After that, the heat press machine was cooled for 10 min by running water through a water inlet. After releasing the jaw pressure, the sandwiched composite with the shim metal plates was removed from the machine. Finally, the samples were allowed to cool for 25 min at room temperature. The manufactured composite samples had an average thickness of 3.00 ± 0.08 mm.

2.2.1. Gamma irradiation

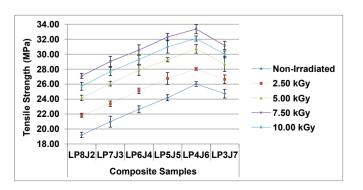
The fabricated hybrid composites were irradiated with a gamma beam model-650, Cobalt-60 (60 Co) source (25 kCi). This was loaded with a source GBS-98 (36 double encapsulated capsules) at the Atomic Energy Research Establishment (AERE), Savar, Dhaka. The irradiation doses were 2.50, 5.00, 7.50 and 10.00 kGy by means of the type C-252 along with 60 Co pellets.

2.2.2. Mechanical performance analysis

The fabricated composite samples were cut to the requisite size according to the standard. The non-irradiated and irradiated samples were tested for tensile strength (TS), tensile modulus (TM), elongation percentage at break (EB%), bending strength (BS), bending modulus (BM), and impact strength (IS). The universal test machine was used to conduct the tensile test in accordance with ASTM D638 (2002) supported by the universal test machine (Model: H50KS-0404, HOUNSFIELD Series S, UK). The ASTM D790 (2017) standard was used to conduct the flexural or bending test. The IS test was performed using the Izod impact testing equipment (HUNG TA INSTRUMENT CO. LTD., Origin-Taiwan; pendulum weight of 2.63 kg, pendulum lift angle of 150°, and a pendulum cycle of 1.13 s) in accordance with ASTM D256 (2018).

2.2.3. Thermogravimetric analysis

TGA was evaluated by means of a Netzsch STA 449 F3 (Germany) with a temperature rate of 10.00 $^{\circ}$ C/min under a constant heating mode in a nitrogen atmosphere with a flow of 10 ml/min. The thermal stability of the optimum rational composite (LP4J6) samples irradiated at 5.00,



7.50, and 10.00 kGy was determined. The following Eq. (3) was used to calculate the consequent percentage of weight loss:

Weight loss% =
$$\frac{W_1 - W_2}{W_2} \times 100$$
 (3)

where, W_1 and W_2 represent the weight of the sample prior to testing and at any given temperature, respectively [32].

2.2.4. Scanning electron microscopy images analysis

SEM images on the tensile fracture surface were compared to the effects of gamma irradiation on the composite using a JSM-6490LA, JEOL, Japan, under an accelerating voltage of 20 kV. SEM images (ranging from \times 30 to \times 1000) of the sample LP4J6 (non-irradiated, 5.00, 7.50, and 10.00 kGy irradiated) were taken under consideration for analysis.

3. Results and discussion

3.1. Mechanical performance

In 50% LLDPE composites, reinforced PALF was blended with jute fibres ranging from 30 to 80% in weight fraction. In addition, mechanical performance is influenced by the compatibility of the matrix, PALF, and jute fibre in the composites [33]. The influence of fibre loading and gamma irradiation on the mechanical properties of the samples was investigated. Figures 1, 2, 3, 4, 5, and 6 show the average results of three tests for each sample strategized with a standard deviation error bar.

3.1.1. Tensile strength

Composites' tensile performance demonstrates their ability to endure a given amount of stress during axial loading without deformation or failure. It is noticed from Figure 1 that the TS of the composite samples increased up to 7.5 kGy radiation. With the further increment of irradiation dose, TS decreased at 10.00 kGy irradiation. In the non-radiated and irradiated states, the maximum TS of sample LP4J6 were 26.01 (± 0.31) MPa and 33.36 (± 0.59) MPa, respectively. The sample LP4J6 had 50% weight fraction LLDPE, 40% PALF, and 60% jute fibre in the remaining 50% weight fraction of the composite. The most favourable ratio of lignocellulosic PALF and jute fibres was observed in LLDPE composite. The best bonding was achieved in that combination (LP4J6) among PALF, jute, and LLDPE. The composite using 80% PALF and 20% jute fibre (LP8J2) had minimum TS (19.20 \pm 0.36 MPa). This research also revealed a reduction in the weight fraction of PALF in jute fibre up to a specific percentage increased the TS. The best TS of sample LP4J6 was achieved by adding more jute fibre to the LLDPE matrix and PALF.

3.1.2. Tensile modulus

The sample LP8J2, which contained 80% PALF and 20% jute, had the lowest TM (722.57 \pm 6.36 MPa), as shown in Figure 2. The aggregation

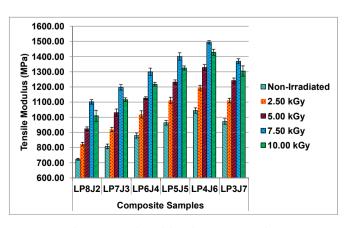


Figure 1. Tensile strength of composite samples.

H. Rahman et al.

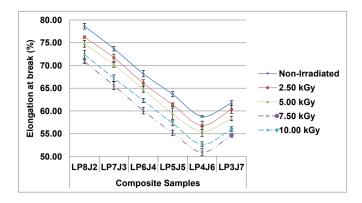


Figure 3. Elongation at break (%) of composite samples.

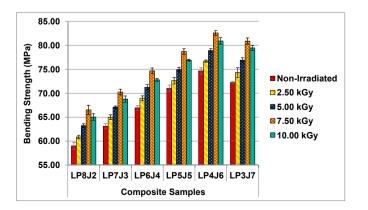
of fibers in matrix decreased the interaction between PALF, jute fiber, and LLDPE. At 7.5 kGy irradiation, the composite sample LP4J6, which contained 40% PALF and 60% jute fibre, had the highest TM (1494.41 \pm 10.94 MPa). Additionally, the composite sample LP3J7 showed a reduced TM than the sample LP4J6 for having a lower PALF (30%) and a higher amount of jute fibre (70%). The compatibility of reinforcement with matrix and the suitable fibre-polymer fraction determine the tensile properties of lignocellulosic fibre-reinforced polymer composites [2]. The bond formation of the fabricated hybrid composite is influenced by the varying PALF and jute fibre proportions in the same LLDPE matrix, as well as the gamma irradiation dosages.

3.1.3. Elongation percentage at break

Figure 3 shows that when the jute fibre percentage was smaller, the composites (LP8J2) had a higher EB% because PALF has a higher extensibility than jute fibres. Similar results were reported up to sample LP4J6, because to worse adhesion between the PALF, jute, and LLDPE. Sample LP3J7 had a greater EB% than LP4J6. However, due to the improved adhesion between PALF-jute and LLDPE, a minimum EB% (50.92 \pm 0.77) was discovered for the optimum percentage of PALF (40%) and jute fibre (60%) in LLDPE composite at 7.50 kGy irradiation. At a specific dose of up to 7.50 kGy, gamma irradiation improved the interfacial contact between PALF-jute fibre and LLDPE, resulting in enough active sites for cross-linking [11]. On the other hand, higher jute fibre and lower PALF content decreased fibre-matrix adhesion, resulting in lower load distribution capability in LLDPE reinforcement.

3.1.4. Bending strength

The highest BS of a 60% jute-containing composite irradiated at 7.50 kGy radiation was 82.58 (\pm 0.49) MPa, while it was 58.98 (\pm 0.74) MPa for a 20% jute-containing composite that was not irradiated (Figure 4). At 7.50 kGy irradiation, the BS was increased to 60% jute content in the composite. Due to the presence of lignin, a higher amount of jute



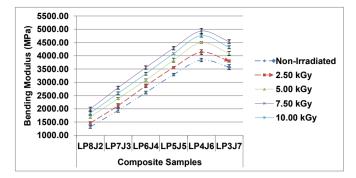


Figure 5. Bending modulus of composite samples.

hardened the structure of a composite. Furthermore, at 10.00 kGy irradiation, BS dropped in composite sample LP3J7, which contained 30% PALF and 70% jute fibre. The composite's reinforcing fibres and LLDPE matrix chain scission was caused by higher dosed gamma irradiation than the optimal 7.50 kGy. Consequently, stress gathered in a definite position in the composites and confined to a small area. As a result of the localised strains, the bending strength was decreased.

3.1.5. Bending modulus

At 7.50 kGy irradiation, the sample LP4J6 showed the highest BM (4932.46 \pm 96.12 MPa) (Figure 5). Due to a lower percentage of PALF and a higher percentage of jute fibre, the BM was lower. The higher amount of jute fibres in sample LP3J7 could be the responsible for crack propagation in the composite. The additional accumulation of the reinforcement fibres in the polymer matrix also increased the chance of bulkiness. This formed the stress centralization regions and trends to instigate the cracks in the composite for a lower amount of impending energy. Increased gamma doses ranging from 2.50 to 7.50 kGy enhanced the bending modulus of the developed samples. Furthermore, as the gamma dose was increased (10.00 kGy), the BM dropped due to the reverse effect of cross-linking, known as photo-degradation. Overdosed gamma irradiation causes this to happen [34].

3.1.6. Impact strength

The IS of irradiated samples improved as the gamma irradiation dose was increased from 2.50 to 7.50 kGy, as shown in Figure 6. The maximal IS of LP4J6 irradiated at 7.50 kGy dosage was reported to be 34.38 (\pm 0.42) kJ/m². When the gamma irradiation dose was increased to 10.00 kGy, the impact strength dropped (31.63 \pm 0.32 kJ/m²). Overdosed irradiation causes crack propagation in the composite. The IS was substantially influenced by the inadequacy of adhesion between the LLDPE matrix and reinforced PALF-jute fibres. At 7.50 kGy irradiation, the stress transmission between the LLDPE matrix and PALF-jute fibres composition in composite sample LP4J6 was excellent. At 7.50 kGy irradiation, the balanced stress transfer prevented crack growth in the composite, and the IS of sample LP4J6 enhanced (Table 3). The ideal hybridization effect of PALF, jute fibre, and LLDPE matrix resulted in this. The cross-linking between molecules of PALF-jute fibre reinforced LLDPE matrix hybrid

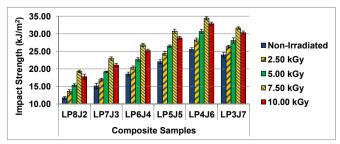


Figure 4. Bending strength of composite samples.

Figure 6. Impact strength of composite samples.

Irradiation dose (kGy)	Sample LP4J6: 50%	Sample LP4J6: 50% LLDPE and 50% fibres (40% PALF and 60% jute fibre)						
	TS (MPa)	TM (MPa)	EB (%)	BS (MPa)	BM (MPa)	IS (kJ/m ²)		
Non-irradiated	26.01 ± 0.31	1044.06 ± 17.61	58.82 ± 0.19	74.65 ± 0.68	3842.19 ± 65.37	25.53 ± 0.50		
2.50	28.02 ± 0.19	1194.35 ± 16.86	56.82 ± 0.94	76.70 ± 0.24	4133.68 ± 91.08	28.25 ± 0.55		
5.00	30.72 ± 0.58	1328.00 ± 19.63	55.47 ± 1.06	78.92 ± 0.42	4495.86 ± 33.28	30.66 ± 0.55		
7.50	33.36 ± 0.59	1494.41 ± 10.94	50.92 ± 0.77	82.58 ± 0.49	4932.46 ± 96.12	34.38 ± 0.42		
10.00	$\textbf{32.10} \pm \textbf{0.34}$	1428.21 ± 19.61	52.67 ± 0.55	80.91 ± 0.70	4762.89 ± 57.87	32.85 ± 0.47		

Table 3. Tensile strength (TS), tensile modulus (TM), elongation percentage at break (EB%), bending strength (BS), bending modulus (BM) and impact strength (IS) of the sample LP4J6 at different irradiation.

composites enhanced after irradiation till 7.50 kGy irradiation. As a result, the intermolecular distance shrank under certain circumstances [35]. After a 7.50 kGy dosage, greater intensity gamma irradiation lowered the IS of composites, causing the matrix chains breaking.

As a result, it can be concluded that gamma irradiation generates enough active sites in produced hybrid composites of PALF-jute fibres and LLDPE to form new bonding till 7.50 kGy. By applying force on the components of composites via Compton scattering, high-energy gamma irradiation creates free radicals in the fibres and matrix [36]. Quick localization of energy created the restricted macro-cellulosic radicals in molecules. As a result of the cross-linking among produced radicals, the characteristics of the composite varied [36]. Up to a certain dose (7.50 kGy irradiation), enough chain scission and cross-linking among the molecules of PALF, jute fibre, and LLDPE in composite occurred, as well as mechanical performance was improved. As the gamma radiation was increased, a large amount of chain scission occurred, and the desired characteristics began to deteriorate [2, 11, 22, 34, 35].

The moduli (tensile and bending) of natural fibre reinforced polymer composites depends greatly on the gamma irradiation dose [36]. The natural fibre reinforced composites may have various tensile and bending moduli in different states, according to a prior study [2, 36]. The maximum bending and tensile moduli were obtained for the sample LP4J6 at 7.50 kGy irradiation, 4932.46 (\pm 96.12) MPa and 1494.41 (\pm 10.94) MPa, respectively. Randomly orientated PALF and jute fibres (5–6 inches) were used to reinforce the composite samples. This considerable fluctuation in moduli could be responsible for the orientation, length, and hybridization impact of fibres in composites, as well as modifying the crystalline structure to impose varied gamma irradiation dosages. It will be taken into consideration for the next area of research.

3.2. Thermal performance

The thermal performance of the composite samples was investigated using TGA. It also indicated the effects of different gamma irradiation dosages on composite weight loss at various temperatures. The physicomechanical properties of the composite sample LP4J6, which contained 40% PALF and 60% jute fibre in an LLDPE matrix, were the best. This sample's thermal performance may also be better for optimal PALF and jute fibre hybridization in LLDPE composites [9]. The thermal stability of three LP4J6 samples irradiated at 5.00, 7.50, and 10.00 kGy was investigated, 6.151 mg, 6.315 mg, and 7.392 mg, respectively, were the initial sample weights. The amount of hydrophilic lignin in lignocellulosic fibres determines their thermal stability [37]. The early decomposition temperature of lignocellulosic fibres is usually around 50 °C, while the leading decomposition temperature is over 300 °C [38].

Figure 7(a) indicated that for a 5.00 kGy irradiation sample, the noteworthy stages of remaining weight after deterioration were 89.78%, 47.89%, 7.80 %, and 0.49 % at 291.71 °C, 410.87 °C, 524.87 °C, and 599.52 °C, respectively. At 304.37 °C, 408.18 °C, 513.45 °C, and 599.63 °C, the visible phases of the weight of the 7.50 kGy irradiation sample were 95.26%, 71.41%, 4.80%, and 0.95%, respectively, as shown in Figure 7. (b). Furthermore, the residual weight steps for a 10.00 kGy irradiation sample were 92.00%, 59.41%, 4.98%, and 0.88 at 320.20 °C, 417.09 °C, 512.26 °C, and 599.67 °C, respectively, as shown in Figure 7.

(c). The omission of wax and lignin was considered in the breakdown region (370 °C–550 °C) [39]. Moisture, hemicellulose, cellulose, lignin, and ash are all present in PALF and jute fibres. As a result, the weight

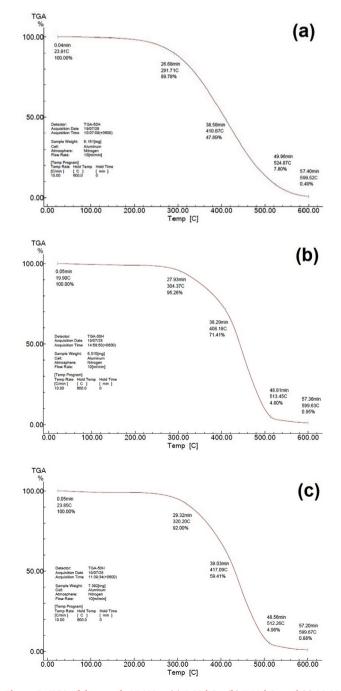


Figure 7. TGA of the sample LP4J6 at (a) 5.00 kGy, (b) 7.50 kGy and (c) 10.00 kGy irradiation.

reduction phases of the composite samples differed [36, 40]. Lignocellulosic fibres begin to disintegrate around 260 °C and show a rapid increment at 300 °C [41].

At roughly 410 °C (410.87–417.09 °C), the weight of the 5.00, 7.50, and 10.00 kGy irradiated samples was 47.89%, 71.41%, and 59.41 %, respectively. At roughly 200 °C, the first slope in Figure 7 represents the breakdown of certain residual moisture in composite materials. Hemicelluloses and lignin degraded up to 350 °C after that, and the tailed slope reflects PALF-jute fibre cellulose degradation [42]. Figure 7's residual slope revealed the breakdown of inorganic components included in samples at temperatures about 500 °C.

The 5.00 kGy irradiation sample degraded faster than the 7.50 kGy and 10.00 kGy irradiated samples, with the weight loss of the 7.50 kGy irradiated sample being the lowest. Around the 600 $^{\circ}$ C temperature, the same findings were observed. The thermal stability of the 7.50 kGy irradiation sample was found to be the best among the three LP4J6 samples studied in this study. By providing active sites in the composite, gamma irradiation promotes adhesion between the fibres and the

polymer matrix. Thus, the physico-mechanical properties of irradiation samples were improved until a dose of 7.50 kGy was reached, resulting in significant cross-linking between reinforced fibres and matrix [43, 44]. This study investigated the effects of gamma irradiation, and found that the thermal stability of produced hybrid composites improved when exposed to specified levels of gamma irradiation.

3.3. Scanning electron microscopy images

In both non-irradiated and irradiated states, the optimal rational jute-PALF reinforced LLDPE hybrid composite LP4J6 exhibited the best physico-mechanical characteristics. SEM images ranging from \times 30 to \times 1000 were used to examine the sample morphology (LP4J6) at the tension fractured surfaces, as shown in Figure 8. Because of the varied amounts of gamma irradiation, the images indicated significant differences in interfacial adhesion between fibres and matrix. The PALF and jute fibres' breakages were readily visible. The different irradiation doses ranging from 5.00 to 10.00 kGy on the composite sample were used to make these

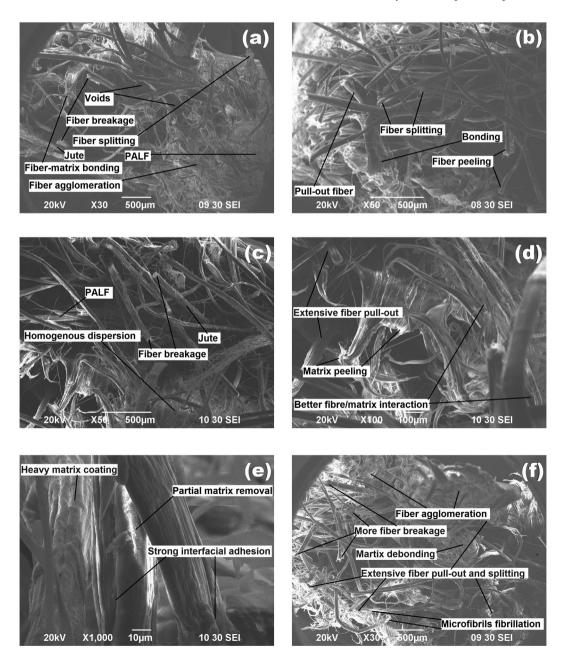


Figure 8. SEM images of the sample LP4J6 on tensile fracture surface irradiated at (a) non-irradiated, (b) 5.00 kGy, (c-e) 7.50 kGy, and (f) 10.00 kGy.

distinctions. Figure 8 (a) shows that the fracture surface of the nonirradiated sample LP4J6 was not regular. The aggregation, fracture, splitting, and voids between the reinforcing fibers and matrix were all readily visible. However, after irradiation, it showed a more stable tensile fractured surface, as shown in Figure 8 (b). It also highlighted the rupture, peeling, and uneven reinforcing allocation of the fibres in the LLDPE matrix. After applying external load, certain voids were discovered that acted as fracture initiators. As a result, at lower tensile strength, the composite failed [42]. Furthermore, the tensile fracture of the surface at the ductile state was caused by fibrillation and crazing in the reinforcement fibres and polymer matrix of the composite [45].

The physical configuration of the optimal rational PALF-jute fibre reinforced LLDPE composites improved significantly after gamma irradiation. The cohesive force between the reinforced fibres and the LLDPE matrix was increased. As a result, better fibre-matrix linkages were formed in the composite, resulting in improved physico-mechanical properties over non-irradiated samples, as shown in Figure 8 (b-f). At 7.50 kGy irradiation, the fractured surface of composite samples showed stable and homogeneous dispersion with excellent bonding between the fibres and matrix, as shown in Figure 8 (c-d). The images indicated a better distribution of PALF and jute fibres in LLDPE, particularly in the matrix-rich region. The amount of reinforcing fibre breakage and partial matrix detachment owing to tensile fracture stress was displayed in Figure 8 (e). The reinforced PALF-jute fibres' fibrils were coated with a thick layer of LLDPE. At a dose of 7.50 kGy, the fibres' rupture and presence of matrix coating around the fibres indicate a strong and adequate contact and interfacial adhesion between PALF-jute fibres and LLDPE. Between the fibres and the matrix, enough active sites and crosslinking were created. The stress transfer was also improved, and the propagating cracks did not spread throughout the composites at the same time. Thus, with 7.50 kGy irradiation, the best physico-mechanical properties developed.

The tensile fracture surface of a 10.00 kGy irradiation sample is shown in Figure 8 (f). Fibre-matrix debonding, agglomeration, microfibrils fibrillation, and an uneven fracture surface were all present to some extent. As a result, at 10.00 kGy irradiation, the physicomechanical characteristics deteriorated.

4. Conclusions

This study looked at the physico-mechanical characteristics and thermal performance of PALF-jute reinforced LLDPE hybrid composites. According on the findings, the following conclusions can be drawn:

- In LLDPE composites, naturally abundant agro waste PALF and jute fibre reinforcement demonstrated a remarkable hybridization effect. The physico-mechanical characteristics of the composite sample having 40% PALF and 60% jute fibre in a 50% (weight fraction) LLDPE matrix were the best. As a result, using agro-waste PALF with jute fibre to reinforce LLDPE hybrid composites will be a very effective strategy.
- At 7.50 kGy irradiation, the most optimum rational composite sample improved TS, TM, BS, BM, and IS by up to 28.26 %, 43.13 %, 10.62 %, 28.38 %, and 34.67 %, respectively, compared to non-irradiated samples. On the other hand, The EB%, reduced by 13.43 % due to improved reinforcement-matrix bonding. This will be a suitable substitute for conventional thermoplastics.
- TGA also revealed that among the 5.00, 7.50, and 10.00 kGy irradiated composites, the thermal stability of the 7.50 kGy irradiated sample was the best. An ideal dose of gamma irradiation creates a stronger intra-chain link between reinforcement fibres and matrix. As a result of the 7.50 kGy gamma irradiation, the mechanical properties and thermal stability of the developed hybrid composite increased.
- Further investigation into the effects of various chemical and physical treatments on PALF-jute fibres is possible. As a result, the compatibility of PALF-jute fibres with LLDPE will be improved. Another topic

of investigation might be the effect of gamma irradiation on the chemical and structural modifications of produced composites in order to gain a better understanding of how different attributes emerge.

Declarations

Author contribution statement

Habibur Rahman: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Farjana Yeasmin: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Tarikul Islam: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Mahmudul Hasan, Muhamad Borhan Uddin: Performed the experiments; Wrote the paper.

Ruhul Amin Khan: Conceived and designed the experiments; Wrote the paper.

Funding statement

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Data availability statement

Data included in article/supplementary material/referenced in article.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

References

- [1] M. Hemath, S. Mavinkere Rangappa, V. Kushvaha, H.N. Dhakal, S. Siengchin, A comprehensive review on mechanical, electromagnetic radiation shielding, and thermal conductivity of fibres/inorganic fillers reinforced hybrid polymer composites, Polym. Compos. 41 (2020) 3940–3965.
- [2] T. Islam, R.A. Khan, M.A. Khan, M.A. Rahman, M. Fernandez-Lahore, Q.M.I. Huque, R. Islam, Physico-mechanical and degradation properties of gamma-irradiated biocomposites of jute fabric-reinforced poly(caprolactone), Polym. Plast. Technol. Eng. 48 (11) (2009) 1198–1205.
- [3] Y.G. Thyavihalli Girijappa, S. Mavinkere Rangappa, J. Parameswaranpillai, S. Siengchin, Natural fibres as sustainable and renewable resource for development of eco-friendly composites: A comprehensive review, Front. Mater. 6 (2019) 1–14.
- [4] N. Akter, J. Saha, S. Das, M. Khan, Effect of bitumen and polyester resin mixture on the physico-mechanical and degradable properties of jute fabrics, Fibres 6 (3) (2018) 44.
- [5] R.A. Khan, H.U. Zaman, M.A. Khan, F. Nigar, T. Islam, R. Islam, S. Saha, M.M. Rahman, A.I. Mustafa, M.A. Gafur, Effect of the incorporation of PVC on the mechanical properties of the jute-reinforced LLDPE composite, Polym. Plast. Technol. Eng. 49 (7) (2010) 707–712.
- [6] B.K. Goriparthi, K.N.S. Suman, N. Mohan Rao, Effect of fibre surface treatments on mechanical and abrasive wear performance of polylactide/jute composites, Compos. Appl. Sci. Manuf. 43 (10) (2012) 1800–1808.
- [7] M. Fagone, H. Kloft, F. Loccarini, G. Ranocchiai, Jute fabric as a reinforcement for rammed earth structures, Compos. B Eng. 175 (2019) 107064.
- [8] S.S. Todkar, S.A. Patil, Review on mechanical properties evaluation of pineapple leaf fibre (PALF) reinforced polymer composites, Compos. B Eng. 174 (2019) 106927.
- [9] M.R. Sanjay, P. Madhu, M. Jawaid, P. Senthamaraikannan, S. Senthil, S. Pradeep, Characterization and properties of natural fibre polymer composites: A comprehensive review, J. Clean. Prod. 172 (2018) 566–581.
- [10] F.Z. Arrakhiz, M. El Achaby, M. Malha, M.O. Bensalah, O. Fassi-Fehri, R. Bouhfid, K. Benmoussa, A. Qaiss, Mechanical and thermal properties of natural fibres

H. Rahman et al.

reinforced polymer composites: doum/low density polyethylene, Mater. Des. 43 (2013) 200-205.

- [11] H.U. Zaman, A.H. Khan, M.A. Hossain, M.A. Khan, R.A. Khan, Mechanical and electrical properties of jute fabrics reinforced polyethylene/polypropylene composites: Role of gamma radiation, Polym. Plast. Technol. Eng. 48 (7) (2009) 760–766.
- [12] N. Graupner, J. Müssig, A comparison of the mechanical characteristics of kenaf and lyocellfibre reinforced poly(lactic acid) (PLA) and poly(3-hydroxybutyrate) (PHB) composites, Compos. Appl. Sci. Manuf. 42 (12) (2011) 2010–2019.
- [13] D.K.K. Cavalcanti, M.D. Banea, J.S.S. Neto, R.A.A. Lima, L.F.M. da Silva, R.J.C. Carbas, Mechanical characterization of intralaminar natural fibre-reinforced hybrid composites, Compos. B Eng. 175 (2019) 107149.
- [14] K. Yorseng, S.M. Rangappa, H. Pulikkalparambil, S. Siengchin, J. Parameswaranpillai, Accelerated weathering studies of kenaf/sisal fibre fabric reinforced fully biobased hybrid bioepoxy composites for semi-structural applications: morphology, thermo-mechanical, water absorption behavior and surface hydrophobicity, Construct. Build. Mater. 235 (2020) 117464.
- [15] M. Rafiquzzaman, M. Islam, H. Rahman, S. Talukdar, N. Hasan, Mechanical property evaluation of glass-jute fibre reinforced polymer composites, Polym. Adv. Technol. 27 (10) (2016) 1308–1316.
- [16] M. Asim, M. Jawaid, M.T. Paridah, N. Saba, M. Nasir, R.M. Shahroze, Dynamic and thermo-mechanical properties of hybridized kenaf/PALF reinforced phenolic composites, Polym. Compos. 40 (10) (2019) 3814–3822.
- [17] P. Uawongsuwan, Y. Yang, H. Hamada, Long jute fibre-reinforced polypropylene composite: Effects of jute fibre bundle and glass fibre hybridization, J. Appl. Polym. Sci. 132 (15) (2014) n/a-n/a.
- [18] S.K. Nayak, S. Mohanty, Sisal glass fibre reinforced PP hybrid composites: Effect of MAPP on the dynamic mechanical and thermal properties, J. Reinforc. Plast. Compos. 29 (10) (2009) 1551–1568.
- [19] M. Ghasemzadeh-Barvarz, C. Duchesne, D. Rodrigue, Mechanical, water absorption, and aging properties of polypropylene/flax/glass fibre hybrid composites, J. Compos. Mater. 49 (30) (2015) 3781–3798.
- [20] M. Boopalan, M. Niranjanaa, M.J. Umapathy, Study on the mechanical properties and thermal properties of jute and banana fibre reinforced epoxy hybrid composites, Compos. B Eng. 51 (2013) 54–57.
- [21] I. Aji, E. Zainudin, K. Abdan, S. Sapuan, M. Khairul, Mechanical properties and water absorption behavior of hybridized kenaf/pineapple leaf fibre-reinforced highdensity polyethylene composite, J. Compos. Mater. 47 (8) (2012) 979–990.
- [22] A.Y.M.A. Azim, S. Alimuzzaman, F. Sarker, Optimizing the fabric architecture and effect of γ-radiation on the mechanical properties of jute fibre reinforced polyester composites, in: ACS Omega, American Chemical Society (ACS), 2022.
- [23] H. Rahman, S. Alimuzzaman, M.M.A. Sayeed, R.A. Khan, Effect of gamma radiation on mechanical properties of pineapple leaf fibre (PALF)-reinforced low-density polyethylene (LDPE) composites, Int. J. Plast. Technol. 23 (2) (2019) 229–238.
- [24] H. Noura, B. Amar, D. Hocine, Y. Rabah, C. Stephane, E.H. Roland, B. Anne, Effect of gamma irradiation aging on mechanical and thermal properties of alfafibre–reinforced polypropylene composites, J. Thermoplast. Compos. Mater. 31 (5) (2017) 598–615.
- [25] A.Y.M.A. Azim, Effect of gamma radiation on the properties of jute reinforced polyester matrix composites, J. Textil. Sci. Eng. 7 (2) (2017) 1–3.
- [26] H.A. Raslan, E.S. Fathy, R.M. Mohamed, Effect of gamma irradiation and fibre surface treatment on the properties of bagasse fibre-reinforced waste polypropylene composites, Int. J. Polym. Anal. Char. 23 (2) (2017) 181–192.
- [27] M.A. Khan, R.A. Khan, Haydaruzzaman, A. Hossain, A.H. Khan, Effect of gamma radiation on the physico-mechanical and electrical properties of jute fibrereinforced polypropylene composites, J. Reinforc. Plast. Compos. 28 (13) (2008) 1651–1660.

- [28] R. Jafari, Effect of Gamma and electron beam irradiation on PAN-carbon fibre composite, Braz. J. Radiat. Sci. 4 (1) (2016).
- [29] Sahadat Hossain, Md, Md. Razzak, M.B. Uddin, A.M.S. Chowdhury, R.A. Khan, Physico-mechanical properties of jute fibre-reinforced LDPE-based composite: Effect of disaccharide (sucrose) and gamma radiation, Radiat. Eff. Defect Solid 175 (5–6) (2019) 516–528.
- [30] Q.T.H. Shubhra, A.K.M.M. Alam, M.A. Khan, M. Saha, D. Saha, M.A. Gafur, Study on the mechanical properties, environmental effect, degradation characteristics and ionizing radiation effect on silk reinforced polypropylene/natural rubber composites, Compos. Appl. Sci. Manuf. 41 (11) (2010) 1587–1596.
- [31] M. Truong, W. Zhong, S. Boyko, M. Alcock, A comparative study on natural fibre density measurement, J. Textil. Inst. 100 (6) (2009) 525–529.
- [32] M. Yang, F. Wang, S. Zhou, Z. Lu, S. Ran, L. Li, J. Shao, Thermal and mechanical performance of unidirectional composites from bamboo fibres with varying volume fractions, Polym. Compos. 40 (10) (2019) 3929–3937.
- [33] N. Saba, M.T. Paridah, M. Jawaid, Mechanical properties of kenaffibre reinforced polymer composite: A review, Construct. Build. Mater. 76 (2015) 87–96.
- [34] J. Sahari, S.M. Sapuan, E.S. Zainudin, M.A. Maleque, Mechanical and thermal properties of environmentally friendly composites derived from sugar palm tree, Mater. Des. 49 (2013) 285–289.
- [35] C.V. More, Z. Alsayed, Mohamed.S. Badawi, Abouzeid.A. Thabet, P.P. Pawar, Polymeric composite materials for radiation shielding: A review, Environ. Chem. Lett. 19 (3) (2021) 2057–2090.
- [36] Haydaruzzaman, R.A. Khan, M.A. Khan, A.H. Khan, M.A. Hossain, Effect of gamma radiation on the performance of jute fabrics-reinforced polypropylene composites, Radiat. Phys. Chem. 78 (11) (2009) 986–993.
- [37] J. Jain, S. Jain, S. Sinha, Characterization and thermal kinetic analysis of pineapple leaf fibres and their reinforcement in epoxy, J. Elastomers Plastics 51 (2018) 224–243.
- [38] T. Ganapathy, R. Sathiskumar, P. Senthamaraikannan, S.S. Saravanakumar, A. Khan, Characterization of raw and alkali treated new natural cellulosic fibres extracted from the aerial roots of banyan tree, Int. J. Biol. Macromol. 138 (2019) 573–581.
- [39] V. Fiore, T. Scalici, A. Valenza, Characterization of a new natural fibre from Arundodonax L. as potential reinforcement of polymer composites, Carbohydr. Polym. 106 (2014) 77–83.
- [40] S.D. Wanjale, J.P. Jog, Polyolefin-based natural fibre composites, in: Cellulose Fibres: Bio- and Nano-Polymer Composites, Springer Berlin Heidelberg, 2011, pp. 377–398.
- [41] H. Burrola-Núñez, P.J. Herrera-Franco, D.E. Rodríguez-Félix, H. Soto-Valdez, T.J. Madera-Santana, Surface modification and performance of jute fibres as reinforcement on polymer matrix: An overview, J. Nat. Fibers (2018) 1–17.
- [42] S.M. Rangappa, J. Parameswaranpillai, K. Yorseng, H. Pulikkalparambil, S. Siengchin, Toughened bioepoxy blends and composites based on poly (ethylene glycol)-block-poly (propylene glycol)-block-poly (ethylene glycol) triblock copolymer and sisal fibre fabrics: a new approach, Construct. Build. Mater. 271 (2021) 121843.
- [43] H.U. Zaman, M.A. Khan, R.A. Khan, Effect of gamma radiation and bulk monomer on jute fabrics polyethylene/polyvinyl chloride composites, J. Polym. Eng. 32 (4–5) (2012) 301–309.
- [44] H. Rahman, F. Yeasmin, S.A. Khan, M.Z. Hasan, M. Roy, M.B. Uddin, R.A. Khan, Fabrication and analysis of physico-mechanical characteristics of NaOH treated PALF reinforced LDPE composites: Effect of gamma irradiation, J. Mater. Res. Technol. (2021).
- [45] F. Lapique, P. Meakin, J. Feder, T. Jossang, Relationships between microstructure, fracture-surface morphology, and mechanical properties in ethylene and propylene polymers and copolymers, J. Appl. Polym. Sci. 77 (11) (2000) 2370–2382.