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Effect of stacking sequence on the performance of hybrid natural/synthetic fiber reinforced polymer composite laminates

Subrata C. Das [a,b,c,*], Debasree Paul [b,c], Sotirios A. Grammatikos [a], Md. A.B. Siddiquee [c], Styliani Papatzani [d], Panagiota Koralli [e], Jahid M.M. Islam [b,f], Mubarak A. Khan [b,g], S.M. Shauddin [b], Ruhul A. Khan [b], Nectarios Vidakis [h], Markos Petousis [h]

[a] *ASEMlab – Advanced and Sustainable Engineering Materials Laboratory, Group of Sustainable Composites, Department of Manufacturing and Civil Engineering, Norwegian University of Science and Technology, Gjøvik 2815, Norway*
[b] *Institute of Radiation and Polymer Technology, Atomic Energy Research Establishment, Bangladesh Atomic Energy Commission, Savar, Dhaka 1349, Bangladesh*
[c] *Department of Textile Engineering, Mawlana Bhashani Science and Technology University, Tangail 1902, Bangladesh*
[d] *Department of Mathematics and Engineering Sciences, Hellenic Army Academy, Evelpidon Avenue, 16672 Vari Attika, Greece*
[e] *Institute of Chemical Biology, National Hellenic Research Foundation, 48 Vassileos Constantinou Avenue, Athens 11635, Greece*
[f] *School of Science, Monash University Malaysia, Sunway Campus, Selangar 47500, Malaysia*
[g] *Bangladesh Jute Mills Corporation, Ministry of Textiles and Jute, Dhaka 1000, Bangladesh*
[h] *Department of Mechanical Engineering, Hellenic Mediterranean University, 71410 Heraklion, Greece*

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ABSTRACT

Here, the effect of stacking sequence on the mechanical and thermomechanical properties of composites using natural fiber (jute), synthetic fiber (glass) and unsaturated polyester resin, is presented. The fabricated composite laminates were neat jute/polyester, neat glass/polyester, and hybrid jute/glass/polyester. It was revealed that neat glass/polyester laminate showed better mechanical performance than the other laminates, and glass fiber hybridization significantly affects the properties of the hybrid laminates. Furthermore, three selected composites were studied using Dynamic Mechanical Analysis (DMA) and Scanning Electron Microscopy (SEM) imaging. Lastly, to improve the mechanical properties of the developed composites, 1 kGy dose of γ-irradiation was applied. As a result, the tensile strength, bending strength, tensile modulus, and bending modulus was increased 10.7, 26.7, 21.5, 36.5% for neat jute/polyester composites; 6.2, 10.9, 50.3, 18.0% for neat glass/polyester composites; and 8.9, 11.9, 21.7, 19.9% for hybrid composites, respectively.

1. Introduction

Nowadays, there is an emerging trend of using natural or renewable resources in composite materials such as natural fibers, bio-based polymer matrices, and bio-based fillers [1–5]. The use of natural fibers in composite materials is considered promising due to a number of advantages such as availability, low cost (raw fibers), reusability, low density, higher specific properties, biodegradability, and non-toxicity. On the other hand, a number of limitations of bio-based composites have been reported, including lower mechanical properties compared to synthetic fibers and hydrophilicity [1,6–9]. In the composite industry, glass fiber reinforcements are largely used due to (among others) their increased mechanical properties, corrosion resistance, non-biodegradability, and low cost [10,11]. Therefore, hybrid fiber-reinforced composites are preferred in order to synergistically employ natural and synthetic fiber reinforcements to exploit the benefits of both [12,13].

The stacking sequence effects on hybrid composites reinforced by jute and glass fiber reinforcements have been studied by several researchers [14–20]. Sanjay et al. has reported experimental findings on the effects of stacking sequence on the tensile, flexural and hardness properties of hybrid laminates made of jute, kenaf and E-glass fiber reinforcements, revealing that glass fiber reinforcement layers generally improve the mechanical properties of hybrid laminates. Also, they reported that glass/kenaf reinforcement layers as skin and jute reinforcement layers as core, exhibited optimum performance among other hybrid laminates [14]. Ramesh et al. studied different hybrid composite laminates made of sisal, jute and glass fiber reinforced

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* Corresponding author at: ASEMlab – Advanced and Sustainable Engineering Materials Laboratory, Group of Sustainable Composites, Department of Manufacturing and Civil Engineering, Norwegian University of Science and Technology, Gjøvik 2815, Norway.

E-mail addresses: subrata.c.das@ntnu.no, scdas.fibers@gmail.com (S.C. Das).

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polyester reporting tensile strength of 176.2, 229.5 and 200.0 MPa and impact strength of 18, 10 and 12 J, for glass/sisal/polyester, glass/jute/polyester and glass/jute/sisal/polyester, respectively [15]. The addition of glass fabric layers as skin/outer layer of the laminate has revealed improved mechanical properties in woven jute/glass/polyester hybrid composites [19]. In jute/glass hybrid composites, it has been reported that the stacking sequence played an important role in the fracture and mechanical properties of the composite laminates [16,20]. Selver et al. studied the effect of stacking sequence on the performance of hybrid composites fabricated with flax, jute, and glass fiber reinforcements and an epoxy matrix. It was revealed that with the addition of natural fiber reinforcements in a glass/epoxy system, the density of the hybrid composite laminates reduces from 1.81 g/cm^3 to 1.61 g/cm^3 and 1.48 g/cm^3, respectively. In the same, lower mechanical properties were reported for hybrid and natural fiber reinforced composites in comparison to glass/epoxy ones. It was also found that glass fiber reinforcement in the outer layers (i.e. glass/flax/glass or glass/jute/glass) contributed to higher bending strength compared to using glass fiber layers in the middle part of the laminates (flax/glass/flax or jute/glass/jute). Dynamic mechanical analysis (DMA) revealed higher damping for natural fiber reinforced composites than glass fiber reinforced composites, whilst tan δ of jute and flax fiber reinforced composites found to be approximately 13% and 16% higher than glass fiber reinforced composites [17]. Recently, Selver et al. also reported findings on the impact and post-impact behavior of glass fiber reinforced polymer composite (GFRP) laminates and hybrid glass/natural fiber reinforced polymer composite laminates made of various layering sequence [18]. GFRP showed higher impact resistance than hybrid laminates. In the case of hybrid laminates, glass fabric layers positioned in the laminate's outer surface (skin) resulted in higher impact strength than when positioned in the interior (core) of the laminates. In the same study, higher energy absorption was exhibited by the natural fiber composite laminates and hybrid composite laminates than for the GFRP composite laminates. The effects of glass/natural fiber-reinforcement stacking sequence have also been studied in other hybrid formulations such as banana/glass [21], cotton/glass [22], coir/glass [23], bamboo/glass [24], areca sheath fibers/jute/glass [25], revealing improved mechanical properties.

Given the relatively lower performance of natural fiber reinforced composites, compared to glass fiber reinforced ones, various techniques have been studied so far to increase the mechanical properties of the former at comparable levels to the latter [26,27]. Gamma (γ) radiation treatment is one of them. Gamma radiation is an ionizing radiation, known to deposit energy on solid cellulose by Compton scattering, realizing an improvement in the final properties of natural fiber reinforced composites, via the production of macro-cellulosic radicals. The radicals generated are responsible for changing the physical, chemical and biological properties of cellulosic fibers [8,28,29]. The benefits of γ-radiation for the tuning of composite materials are: continuous and fast processing, low atmospheric pollution, processing at ambient temperatures, adaptability to various manufacturing processes, etc. [30–33]. Khan et al. studied the effect of γ-radiation (2.5–10 kGy) on jute/polypropylene composites, reporting increased mechanical properties for the irradiated composites. In this work, an attempt was carried out to achieve higher mechanical properties when using 5 kGy dose of γ-radiation [28,29,34]. In specific, tensile strength (TS), tensile modulus or stiffness (TM), bending strength (BS), bending modulus (BM) and impact strength (IS) were improved by 16, 45, 12, 38 and 62%, respectively [34]. The effects of γ-radiation on the mechanical properties of hybrid composites have also been investigated by Refs [35,36].

Jute is an abundant plant in Bangladesh, therefore, utilizing this readily available and economically viable natural resource into composite materials for multi-purpose applications, impacts positively sustainability at national and international levels. Although, when considering harsh service conditions and structural applications, there are still major limitations pertaining to relatively low mechanical properties and very poor water resistance [7,9,37,38]. Therefore, GFRPs are preferred in demanding applications due to the higher mechanical properties compared to jute fiber reinforced polymer composites [39,40], but also due to the long-term stability and performance, low cost, high availability, processability, etc. Thus, until the development of natural fiber composites (NFCs) that possess comparable properties and cost to GFRP composites, the combination of glass and jute fiber reinforcement layers to produce hybrid polymer composite laminates can be a viable solution to achieve desired physico-mechanical properties in order to increase market uptake of jute NFCs. To the best of the authors‘ knowledge, the application of γ-radiation is very limited in hybrid composites, as it has mostly been applied for the strengthening of thermoplastic-based composites, whilst very limited applications have been reported for thermosetting polymer composites. Hence, the present research work sheds light to the usage of γ-radiation in natural jute/glass fiber reinforced thermosetting polymer matrix (polyester) hybrid composites, which can offer a sustainable and chemical-free post-treatment for the enhancement of the mechanical properties of polymer composite structures.

2. Materials and methods

2.1. Materials

The reinforcement materials were woven jute fabric (1 × 1 plain weave) and E-glass fiber mat (400 g/m^2 of areal density). The matrix material was unsaturated polyester resin, and the employed hardener was MEKP (methyl ethyl ketone peroxide). The jute fabric was purchased from the local market of Dhaka, Bangladesh; E-glass mat and polyester resin system were procured from SHCP, Singapore. Table 1 represents the general properties of jute fibers and glass fibers. The properties of the unsaturated polyester resin matrix is tabulated in Table 2 according to the company data sheet.

2.2. Methodology

2.2.1. Fabrication of composites

First, the reinforcement such as jute fabric and E-glass mat were cut into the size of 40 × 40 mm^2. To remove moisture, jute fabric was first oven-dried at a temperature of 100 °C for 60 min. In the sequel, the desired amount of polyester resin was added to a plastic pot, mixed with MEKP (1–2%), mixed gently with a stirrer for a few minutes, before the final use of the resin system for fiber impregnation. The composite laminates were wet laid-up, and then hot-pressed in a hydraulic press to facilitate curing, fiber impregnation and laminate consolidation at a temperature of 90 °C for a period of 10 min. This process was then followed by room temperature post-curing for 24 h. Three types of laminates with varying stacking sequences were prepared, such as a five-layer neat jute/polyester composite laminate (S0), five-layer neat glass/polyester composite (S7) and also hybrid composite laminates via combinations of various layers of jute fabric and glass non-woven mats in a polyester matrix (S1–S6). All stacking

Table 1
Properties of jute and glass fibers [41].

Properties	Jute fibers	E-glass fibers
Density (g/cm^3)	1.3–1.49	2.5–2.59
Diameter (μm)	20–200	< 17
TS (MPa)	320–800	2000–3500
TM (GPa)	8–78	70–76
Specific modulus (approx.)	30	29
Elongation (%)	1–1.8	1.8–4.8
Moisture content (wt.%)	12.5–13.7	–

Table 2
Properties of unsaturated polyester resin.

Properties	Values
Appearance	Opaque, blue color change resin solution
Viscosity at 30 °C	4–5
Water absorption (%) (7 day value)	0.35
Heat distortion temperature (°C)	67.3
Elongation at break (%)	3.2
BS (kgf/mm^2)	8.4
BM (kgf/mm^2)	536.1
TS (kgf/mm^2)	3
IS (kgf-cm/cm)	3.9

sequences are tabulated in Table 3. Here, J and G stand for jute and glass fiber reinforcement laminas/layers, respectively.

2.2.1.1. Irradiation of the composites. The irradiation of the composite specimens was performed at the Atomic Energy Research Establishment, Savar, Dhaka, employing a dose of 1 kGy, using a Co-60 gamma source (25 kci).

2.2.2. Mechanical properties testing

2.2.2.1. Tensile strength. Tensile tests of the composites were conducted according to the ASTM D638-03 [42] using a Universal Testing Machine (Model: H50KS-0404, Hounsfield Series S, UK) with a crosshead speed of 10 mm per min at a span distance of 50 mm. The plane dimensions of the test specimens were 120 (l) × 15 (w) mm^2.

2.2.2.2. Flexural strength. Static flexural tests were carried out according to ISO 14125 [43] using the same testing machine mentioned above with a crosshead speed of 60 mm per min at a span distance of 25 mm. The specimen dimensions were 60 (l) × 15 (w) mm^2.

2.2.2.3. Impact testing. Impact tests were conducted on unnotched mode composite specimens according to ASTM D 6110-97 [44] using a Universal Impact Tester (HUNG TA INSTRUMENT CO. LTD, Taiwan). A hammer mass of 2.63 kg at a gravity distance of 30.68 mm and lift angle of 150° was implemented.

2.2.2.4. DMA. DMA (Dynamic mechanical analysis) of neat jute/polyester, neat glass/polyester and hybrid (jute/glass/polyester) composites were performed using a dynamic mechanical analyzer (DMA 850, Discovery, USA). The test was conducted as per ASTM D 5023 with the specimen dimension of 50 (l) × 13 (w) mm^2. The temperature increased at a rate of 2 °C per minute from room temperature to 160 °C at 1 Hz frequency. Liquid nitrogen was employed as the cooling agent.

2.2.2.5. SEM. A Schottky Field Emission ultra-high resolution Scanning Electron Microscope (FE-SEM, JEOL 7610F, Japan) was used to examine the fracture surfaces of the composite specimens. The specimens were chosen from the tensile tests series and were sputtered with Pd/Au prior to examination. The sputter/coater used was a Quorum SC7620 (Quorum Technologies Ltd, UK). Imaging was carried out at a maximum operating range of 5 kV, and the surface micrographs were taken at increasing magnifications and at a scale of 100 and 10 μm.

3. Results

3.1. Mechanical properties

The tensile strength (TS) of the hybrid laminates S1, S2, S3, S4, S5 and S6 were 91.3, 76.7, 123.1, 132.8, 125.8 and 137.6 MPa, respectively, whilst the bending strength (BS) values recorded were 166.6, 114.4, 313.0, 173.6, 163.3 and 252.4 MPa, respectively, as shown in Fig. 1. For neat jute/polyester composite (S0), TS and BS were found to be 64.6 and 127.2 MPa, respectively, and for neat glass/polyester composite (S7), TS and BS were found to be 159.1 and 340.4 MPa,

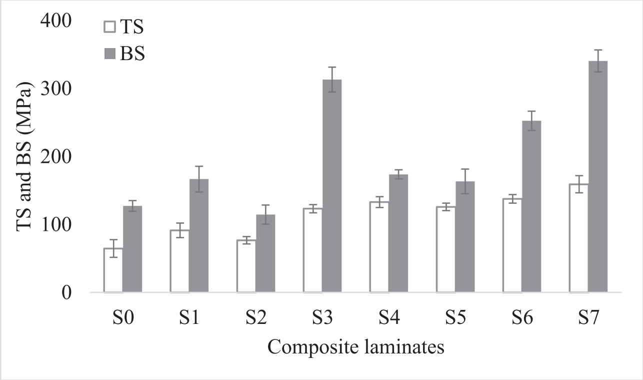

Fig. 1. Tensile and flexural strength of the composites.

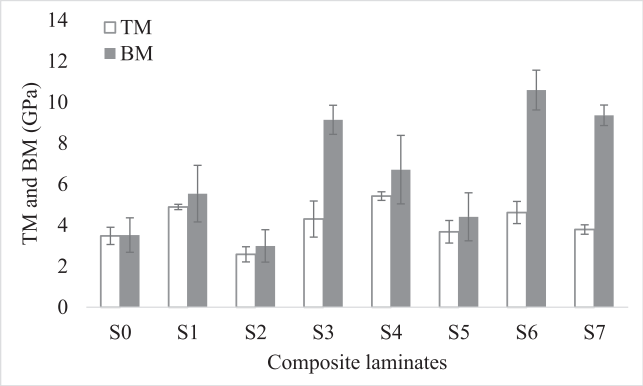

Fig. 2. Tensile and flexural modulus of the composites.

Table 3
Formulation of different composites.

Symbol	Stacking or layering sequence	Jute fiber content (wt. %)	Glass fiber content (wt. %)	Jute fiber volume fraction (%)	Glass fiber volume fraction (%)	Laminate thickness (mm)
S0	J + J + J + J + J	61.9	–	55.5	–	1.95 ± 0.07
S1	J + G	29.3	30.1	35.6	25.4	0.71 ± 0.02
S2	J + G + J	27.4	12.3	25.8	8.6	1.66 ± 0.07
S3	G + J + G	23.6	47.7	38.7	43.3	1.43 ± 0.03
S4	J + G + J + G	37.6	29.2	46.5	28.8	1.11 ± 0.08
S5	J + G + J + G + J	40.3	24.9	47.0	24.8	1.62 ± 0.03
S6	G + J + G + J + G	24.5	30.8	29.6	24.1	1.80 ± 0.07
S7	G + G + G + G + G	–	48.7	–	30.4	2.17 ± 0.09

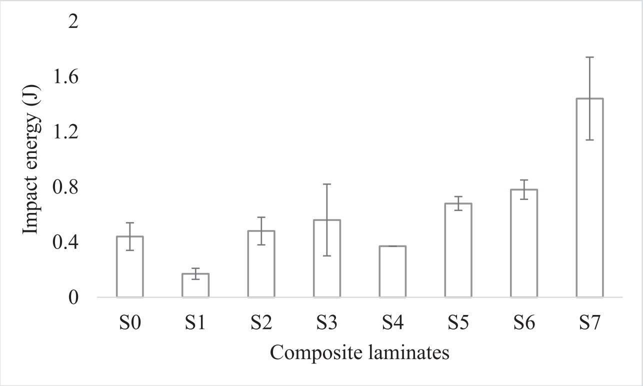

Fig. 3. Impact energy of the composites.

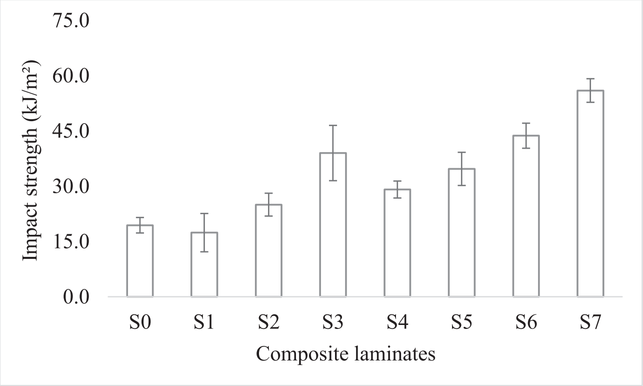

Fig. 4. Impact strength of the composites.

respectively. The S0 laminate (neat jute/polyester composite) exhibited the lowest BS value, whilst S7 (neat glass/polyester composite) the highest value of BS compared to all other composite laminate formulations. Fig. 2 shows that for S0, S1, S2, S3, S4, S5, S6 and S7 composites, tensile modulus (TM) values were recorded to be 3.48, 4.89, 2.58, 4.30, 5.42, 3.68, 4.62 and 3.79 GPa, respectively, and bending modulus (BM) values 3.52, 5.54, 2.99, 9.14, 6.71, 4.41, 10.59 and 9.36 GPa, respectively. S7 exhibited higher bending stiffness than S0 and the hybrid laminates.

The TS is positively influenced by the addition of glass fiber laminas, as in the case of hybrid laminates [23,45]. This is due to the higher strength and stiffness of glass fiber reinforcement compared to jute fiber reinforcement. On top of that, fiber–matrix adhesion is reported to be improved for glass fibers/polyester than jute/polyester, so it is more difficult for the prevalent forces to pull-out glass fibers from the polyester matrix than natural fibers [12–14]. As such, failure of the composites reported to occur due to the fracture of glass fibers in the case of glass/polyester compared to fiber pull out, in the case of natural fibers/polyester. Similar hybridization effects were also observed by Refs [14,40]. With respect to the bending of hybrid laminates, a non-linear behavior was exhibited, revealing the higher BS and BM for the case of S3 and S6. This indicates that both the placement of glass fiber layers as skin and the glass fiber content (wt%) within a hybrid laminate influence positively the bending performance [14,17,19,46].

For S0, S1, S2, S3, S4, S5, S6, S7 composites, the impact energy values were found to be 0.44, 0.17, 0.48, 0.56, 0.37, 0.68, 0.78, 1.44 J, respectively, as shown in Fig. 3; and the impact strength values recorded were 19.4, 17.4, 25.0, 39.0, 29.1, 34.7, 43.7, 56.0 kJ/m^2, respectively, for S0, S1, S2, S3, S4, S5, S6 and S7, respectively, as shown in Fig. 4. The highest value of impact energy and impact strength was reached by S7, which was expected. Interestingly enough, the hybrid laminates showed higher impact strength values than neat jute/polyester due to the presence of glass fibers layers. The impact strength of FRPs is dependent on various factors such as properties of the reinforcing fibers used in a composite; more importantly, the fiber–matrix interfacial adhesion as well as the interlaminar adhesion [14,19,40,46–48].

Three types of composite laminates were selected for further investigation, namely: neat jute/polyester (S0), neat glass/polyester (S7) and jute/glass/polyester hybrid (S6). Two regimes of further testing were carried out:

a) The performance of the three composite laminates was interrogated further, employing DMA and SEM.
b) The effect of γ-ray radiation on the mechanical properties of the three composite laminates was assessed after irradiation under Co-60 of γ-ray of 1 kGy dose.

3.2. DMA

Fig. 5(a) shows the storage modulus (E') curve as a function of temperature at 1 Hz frequency of neat jute/polyester, neat glass/polyester and hybrid laminates. E' is the amount of energy absorbed by compos-

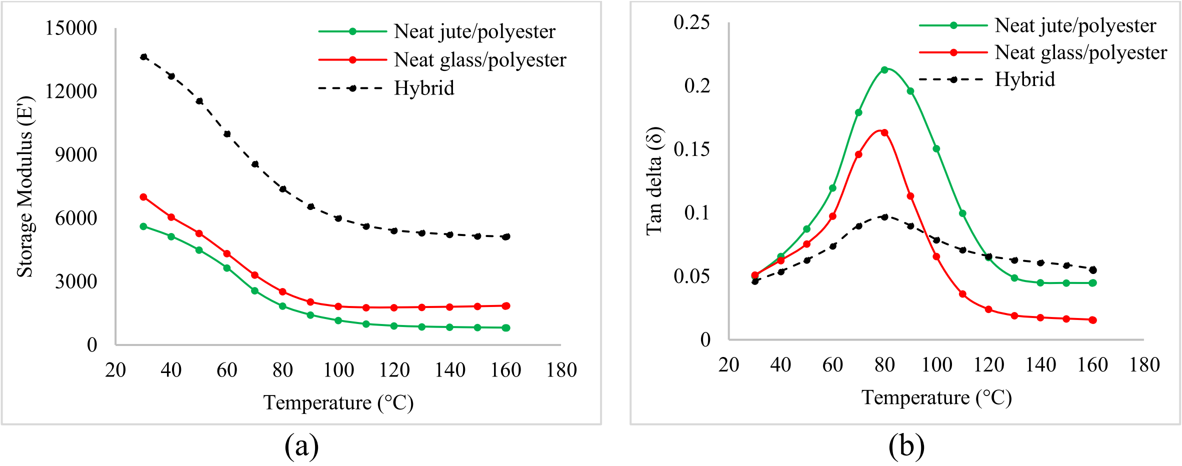

Fig. 5. Storage modulus (E') vs temperature (a), and damping (Tan δ) vs temperature (b) graphs for the composites.

Table 4
Peak height of tan δ curve, Tg (°C) from loss modulus (E") and tan δ curve.

Composites	Peak height of tan δ curve	Tg (°C) from E" curve	Tg (°C) from tan δ curve
Neat jute/polyester	0.213	68.64	80.64
Neat glass/polyester	0.168	71.35	76.85
Hybrid (jute/glass/polyester)	0.097	72.77	78.66

ites per cycle of oscillation and a measure of composite stiffness. In this work, improved E' was found for hybrid laminate than neat glass/polyester and neat jute/polyester composites, which is confirming the BM values illustrated in Fig. 2. With the increase of temperature, the E' of all composites decreased due to temperature-related stiffness loss in fibers [49,50]. The value of E' decreased gradually due to the rise in temperature in the transition region because of the increase in molecular mobility at temperature above Tg.

The damping (tan δ) curve as a function of temperature at 1 Hz frequency of neat jute/polyester, neat glass/polyester and hybrid laminates is shown in Fig. 5(b). It mainly comprises the ratio of E" (loss modulus) to E' that depends on the adhesion between fibers and matrix. The maximum peak of tan δ found for neat jute/polyester composite revealed better damping than that of other composites. Table 4 shows the peak of tan δ curve and Tg (°C) obtained from tan δ curve for all the composites.

3.3. SEM

Micrographs of neat jute/polyester, neat glass/polyester and jute/glass/polyester hybrid laminates are shown in Fig. 6 at various magnifications. Imaging was carried out near the fractured surface of tensile

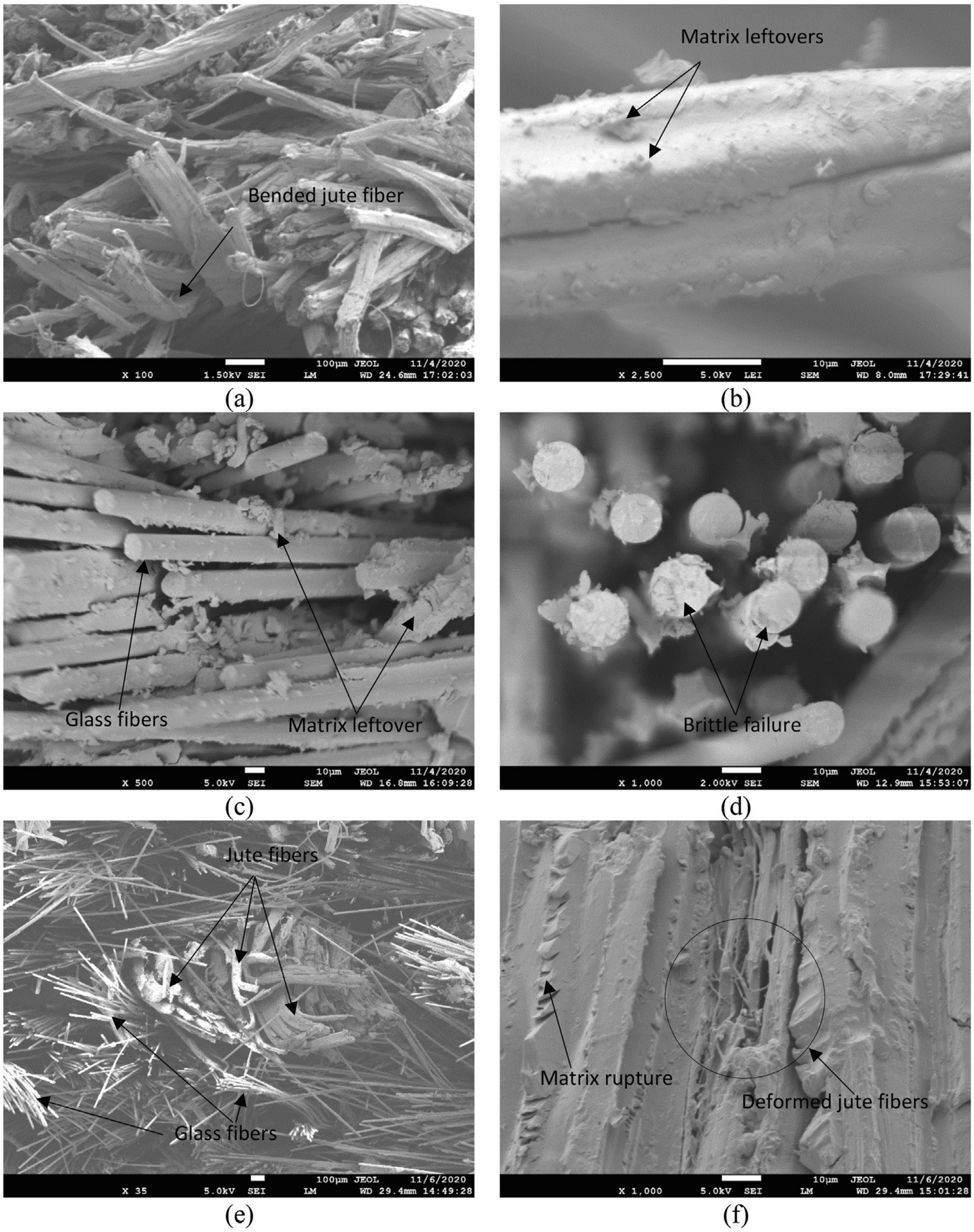

Fig. 6. SEM images of neat jute/polyester (a, b), neat glass/polyester (c, d) and jute/glass/polyester hybrid (e, f) composite laminates at various magnification.

tested specimens. Fig. 6 (a, b) shows the micrograph of neat jute/polyester composites, and from the figures it can be seen that jute fibers pulled out, leading to fiber entanglement, whilst the orientation of the fibers after fracture shows to be random, denoting a somewhat non-brittle failure. Interface cracks or voids are also found on the jute fiber surface after fiber pulled out during tension (Fig. 6 b). On the other hand, glass fibers exhibit brittle failure with the fibers‘ breakage surface being flat and smooth (Fig. 6 c, d). Also, there can be distinguished matrix left overs on the surface of the fibers, which is a sign that the fiber–matrix interface was strong. In the case of hybrid composites, as indicated in Fig. 6 (e, f), brittle failure occurred for glass fibers and fiber pull-out for jute fibers [14,25]. The surface morphology of the matrix can also be seen in Fig. 6 (f), revealing the affinity of both jute and glass fibers with the polyester matrix.

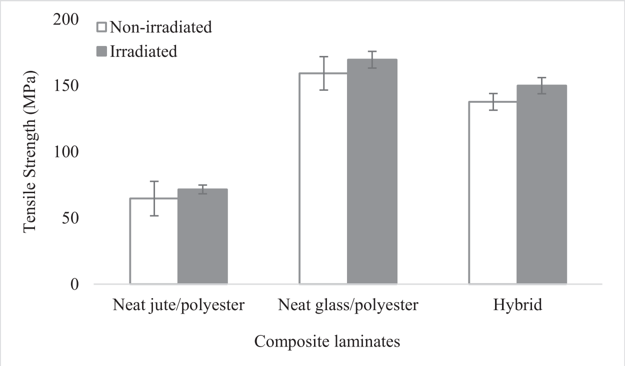

Fig. 7. Effect of γ-radiation on the TS of the composites.

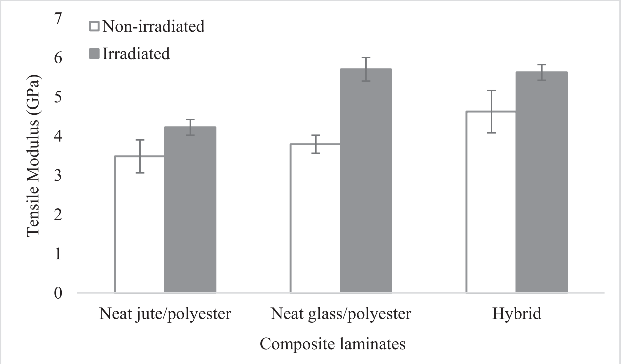

Fig. 8. Effect of γ-radiation on the TM of the composites.

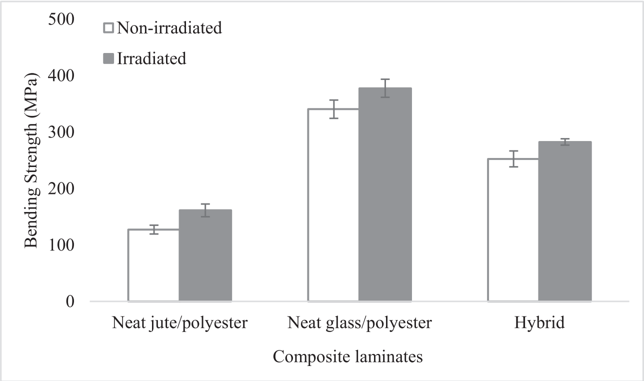

Fig. 9. Effect of γ-radiation on the BS of the composites.

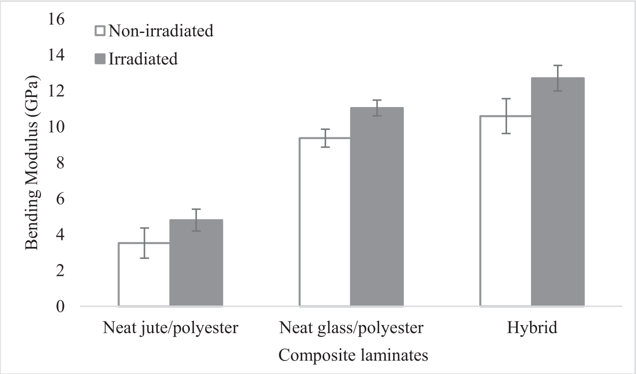

Fig. 10. Effect of γ-radiation on the BM of the composites.

3.4. Mechanical properties of selected irradiated laminates

The effect of radiation is studied in this section. Figs. 7 and 8 show the TS and TM of neat jute/polyester, neat glass/polyester and hybrid composite laminates upon the γ-radiation treatment (1 kGy dose), respectively. After irradiation, the TS of neat jute/polyester, neat glass/polyester and hybrid composites were measured to be equal to 71.5, 169.4, 149.8 MPa, respectively, as shown in Fig. 5. It is found that at 1 kGy dose, the mean values of TS marginally increased by 10.7, 6.2 and 8.9% for neat jute/polyester, neat glass/polyester and hybrid composites, respectively, compared to that of untreated composites (non-irradiated). Fig. 8 shows the variation of TM of the different composites after γ-radiation revealing mean TM values increase of 21.5, 50.3 and 21.7% for neat jute/polyester, neat glass/polyester and hybrid composites, respectively. Similar to tensile properties, the BS and BM improved with the γ-radiation treatment as shown in Figs. 9 and 10, respectively. For neat jute/polyester, neat glass/polyester and hybrid laminates, the recorded improvement of mean BS and BM values was found to be 26.7, 10.9, 11.9%, and 36.5, 18.0, 19.9%, respectively.

All types of composite laminates showed improvement in their mechanical properties by γ-irradiation compared to untreated composites. The reason behind the improvement of mechanical properties by γ-irradiation exposure is well elaborated in previous works of Khan et al. [8,28,34,35], revealing that reinforcement and matrix are affected by γ-irradiation, which may produce active sites that can contribute to better fiber and matrix bonding and is confirmed by the present work. At low radiation dose, bond scission and cross-linking occur, but at a higher dose, bond scission is prevalent. Additionally, γ-irradiation may also remove moisture from a composite, which in turn contributes to higher performance.

4. Conclusions

The findings of the study can be summarized as follows:

In the case of mechanical properties, the neat glass/polyester (S7) exhibited the highest mechanical properties expectedly among others. The tensile, bending, and impact strength of S7 found 146.3, 167.6, and 188.7% higher than that of neat jute/polyester (S0) composite, respectively. The stacking sequence has found a predominant influence on the mechanical properties of hybrid composites, where glass fiber content and the position of glass fiber layers in the composite laminates play an important role. Among various hybrid composites, S6 revealed higher mechanical properties, i.e., 32.8 and 299.9% higher tensile and bending modulus, respectively, than S0; and 21.9 and 13.1% higher tensile and bending modulus, respectively, than S7.

From DMA study, the adequate thermal stability of neat glass/polyester composites and hybrid composite laminates was found. The hybrid laminate revealed the highest storage modulus values, among others, confirming the flexural test results. Furthermore, the hybrid composite laminate case exhibited the highest damping effect among all, whilst Tg remained expectedly unaffected by the stacking sequence and type of fabric.

SEM micrograph revealed a strong affinity between both fiber reinforcements and the polyester matrix, as well as the profoundly brittle failure of glass fiber reinforcement compared to jute.

The post-treatment by ionizing (γ) radiation showed evidence of mechanical properties enhancement for all composites. Other doses could be examined in future research to maximize the improvement of γ-irradiated composites, a potentially interesting sector for larger scale applications by industry.

The hybrid composites studied in this work can be used in low to moderate load-bearing structural and semi-structural applications in automotive, household or furniture and packaging industries, etc., due to the performance delivered.

In the future, a life cycle assessment (LCA) study to assess the environmental impact of hybrid composite laminate, could yield interesting results.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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