EFFECT OF ELECTRON BEAM IRRADIATION ON THE MECHANICAL AND IMPACT PROPERTIES AND WATER ABSORPTION OF RANDOM AND 2-D JUTE/PLA GREEN COMPOSITES

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1. Introduction

Cellulose-based natural fibers have been widely used as reinforcing fibers in natural fiber composites or green composites due to their advantages in lightness, cost effectiveness, environmental friendliness, carbon dioxide reduction in nature, etc. over conventional glass fibers. Green composites are now doing research and being developed in many industries, especially in automotive, building, commodity, and other applications [1,2]. Among many cellulose-based natural fibers like jute, kenaf, hemp, flax, sisal, henequen, and banana, jute fibers are attracting attention due to their low cost, natural abundance and industrial availability.

In fiber-reinforced polymer composites the mechanical properties, in general, depend on various factors like fiber and matrix types, fiber-matrix adhesion, fiber content, fiber aspect ratio, fiber orientation, fiber modification, and composite processing method as well as stress transfer efficiency at interfaces [3,4].

It has been addressed that the fiber-matrix adhesion is critically important in a composite system, particularly in green composites with natural fibers and polymer. However, the interfacial bonding between the natural fiber and the polymer is poor, leading to the reduction of the properties and performances of green composites [5]. There are two main methods of natural fiber surface modification to improve the interfacial adhesion between the natural fibers and the resin in a green composite. One is chemical modification of natural fiber surfaces by means of dewaxing, alkalization, grafting, cyanoethylation, acetylation, bleaching, and sizing with silane or other coupling agents [5]. The other is physical modification by means of plasma and electron beam treatments. A number of studies on the chemical and physical modifications have been performed to improve the interfacial, mechanical and/or thermal properties of various green composites with different natural fibers and polymer matrices [6-11]. Although a few papers studied on the effect of electron beam treatment of natural fibers on the properties of other composite systems have been reported by Cho *et al.* [10-13], the electron beam effect on green composites with PLA and jute has been scarcely reported.

Electron beam irradiation techniques have been increasingly utilized for surface modification and property enhancement of polymer materials like fibers, films, plastics and composites for many years. It may remove the surface impurities and generate functional groups on the fiber surfaces with an optimal treatment. Electron beam processing is a dry, clean and cold method with advantages such as energy-saving, high speed, uniform irradiation, and environmental friendliness.

Consequently, the research objective is to investigate and compare the influence of electron beam exposed to jute fiber surfaces on the mechanical and impact properties and water absorption of randomly aligned jute fiber/PLA and two-directionally aligned jute fabric/PLA green composites. The investigation was focused on the interlaminar shear strength, tensile and flexural properties, impact strength and water absorption behavior.

2. Experimental

2.1 Materials

Jute fiber bundles of 70 to 80 mm long were used throughout this work, supplied from Bangladeshi Jute Institute. Jute fabrics (Model B-3514) with plain weave, which were produced in India, were used. Poly(L-lactic acid) (PLA, 2002D Grade, pellet form, NatureWorks™) was used. Prior to use, jute fibers, jute fabrics and PLA pellets were sufficiently dried at 70°C for several hours in a vacuum oven.

2.2 Electron Beam Irradiation of Jute

Prior to composite fabrication, raw jute fiber bundles and jute fabrics sealed in polyethylene bags were irradiated at various electron beam dosages of 5, 10, 30, 50, and 100 kGy, respectively. The electron beam irradiation processes were performed at ambient temperature in air using ELV-4 type equipment at EB-Tech Co., Ltd., Deajeon, Korea. Raw jute fibers and fabrics without electron beam treatment (0 kGy) were also used for comparison.

2.3 Fabrication of Green Composites

Jute fiber bundles, jute fabrics, and PLA pellets were sufficiently dried at 70°C for 12 h in a vacuum oven before composite fabrication. For preparing random jute/PLA green composites, the irradiated jute fibers were chopped to about 10±0.5 mm long in average. For preparing 2-D jute/PLA green composites, the irradiated jute fabrics were cut to 150 mm × 200 mm in size. Both random and 2-D jute/PLA green composites were fabricated by a compression molding method.

2.4 Characterization

Prior to resin microdroplet formation, jute fibers with a relatively uniform fiber diameter were selected for the single fiber microbonding test because jute fibers may have different fiber diameters, depending on the fiber locations due to the irregular fiber surfaces. A universal testing machine (Instron 4467) was used for a single fiber microbonding test. The load cell was 100 N and the crosshead speed was 2 mm/min. The micro-vise grip distance was 20 mm. The average value of the interfacial shear strength for green composite was obtained from 30 specimens of each sample using the following equation.

$$\tau = F/(\pi \cdot D_f \cdot L_e)$$

Here, τ is the interfacial shear strength (IFSS). F is the force required for debonding the PLA resin microdroplet from the single jute filament while tensile loads are applied. π is the circular constant, 3.1416. D_f is the diameter of the measuring fiber and L_e is the jute fiber length embedded in the PLA resin microdroplet.

The interlaminar shear strength (ILSS) of jute fabric/PLA green composites was also measured by a short-beam shear test according to ASTM D2344 using a universal testing machine (Instron 4467). The span-to-depth ratio was 4:1. The load cell was 30 kN and the crosshead speed was 1 mm/min. The average value of ILSS was obtained from10 specimens of each sample.

The flexural properties of jute/PLA green composites with various electron beam dosages were measured using a three-point bending test method according to ASTM D790M-86 using a universal testing machine (Instron 4467). The span-to-depth ratio was 16. A load cell of 30 kN was used. A crosshead speed of 2.1 mm/min was used. The average values of flexural strength and modulus were obtained from ten test specimens.

The impacts tests for neat PLA and random-type and 2D-type jute/PLA green composites were performed at ambient temperature with an 18.54 N hammer weight and with 21.6 J impact energy according to ASTM D256 using an Izod-type impact tester (Tinius Olsen Model-892, USA). All of the samples were notched and the dimensions were 62 mm x 10.2 mm x 3.2 mm. The average impact strength of each sample was obtained from successful ten specimens.

Water absorption test was carried out at 25℃ in a water bath of 3000 ml. Distilled water was used. Each specimen was intentionally cut in the middle of the compression-molded plaque so that the jute fibers located in the thickness direction of the composite were readily exposed to water. This was to examine directly the effect of electron beam treatment on the water resistance of the jute/PLA green composites. The specimen dimensions were 135 mm × 15 mm × 5 mm. The weight change of each specimen was monitored once a day for 50 days.

3. Results and Discussion

Fig. 1 presents the effect of electron beam dosage irradiated to jute fibers on the interfacial shear strength of jute/PLA green composites. In the relatively low dosages below 10 kGy, the IFSS was slightly increased. A significant increase of the IFSS

was obtained at 10 kGy, indicating an improvement of about 22% on the interfacial adhesion between the jute fibers and the PLA in comparison to the specimen without electron beam irradiation. Above 10 kGy, the IFSS was rather decreased gradually with increasing the electron beam intensity.

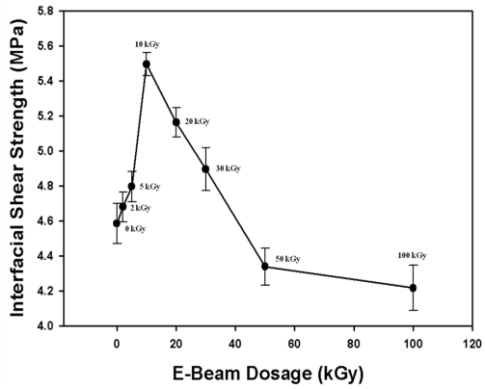

Fig. 1. Interfacial shear strength of jute/PLA green composites as a function of electron beam dosage.

Fig. 2 depicts the variation of the interlaminar shear strength of 2-D jute fabric/PLA green composites as a function of electron beam intensity. The ILSS variation exhibited a similar tendency with the IFSS, showing the maximum ILSS value at 10 kGy and the gradual reduction of the ILSS from 10 kGy to 100 kGy. The result indicated that the interfacial adhesion between the jute fiber and the PLA matrix resin was greatest at 10 kGy. It may be expected that such an enhancement of the jute-PLA adhesion can contribute significantly to improving the mechanical properties and water absorption resistance of jute/PLA green composites.

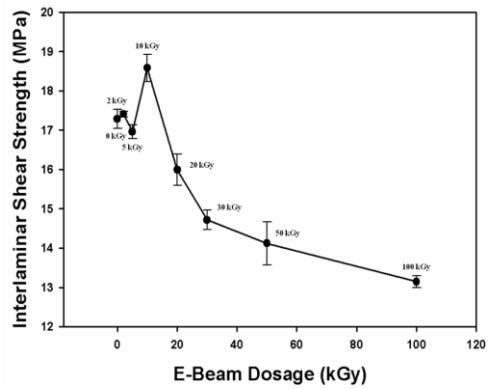

Fig. 2. Interlaminar shear strength of 2-D jute/PLA green composites as a function of electron beam dosage.

Fig. 3 shows the effect of electron beam irradiation of the flexural strength and modulus of random jute/PLA green composites with different jute loadings. With increasing jute loading the flexural strength was increased depending on the electron beam dosage. The flexural strength of un-irradiated random jute/PLA specimen was lower than that of neat PLA and it was enhanced with the irradiation, showing the maximum at 10 kGy. The irradiation higher than 10 kGy rather decreased the strength. The flexural modulus of all random jute/PLA green composites were greater than that of neat and it was significantly increased with the irradiation with the maximum value at 10 kGy and 50 wt.% loading of chopped jute fibers.

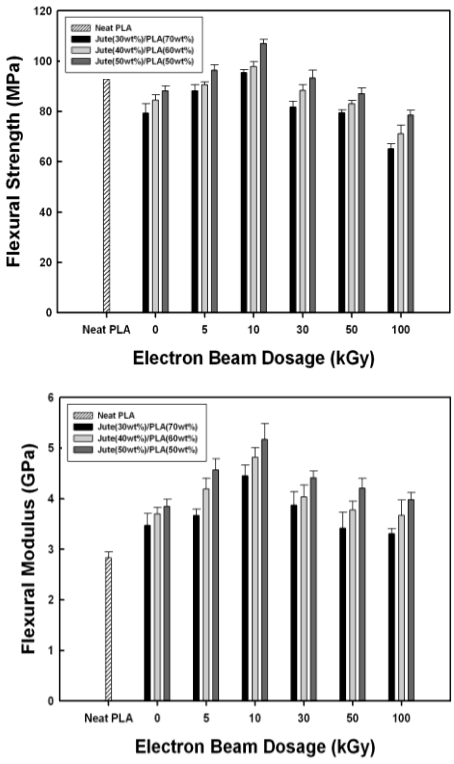

Fig.3. Flexural strength and modulus of random jute/PLA green composites with jute fibers treated at different electron beam dosages.

Fig. 4 shows the effect of electron beam irradiation of the flexural strength and modulus of 2-D jute fabric/PLA green composites with different jute loadings. The overall tendency of the variation of the flexural strength and modulus depending on the electron beam dosage and the jute loading was similar with that of the random counterpart. The

greatest flexural strength and modulus were obtained at 10 kGy. This is mostly ascribed to the jute fiber surfaces modified physically by the electron beam irradiation. The surfaces were cleaned with the removal of low molecular weight components like wax, weak boundary layers and surface impurities. The effect was greatest at 10 kGy.

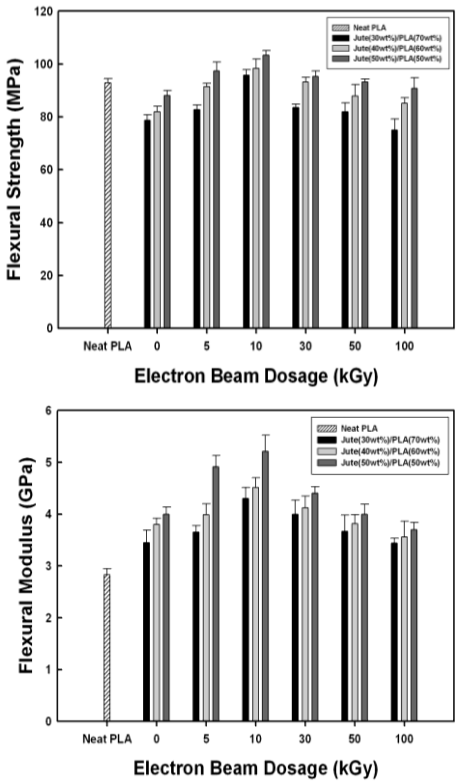

Fig.4. Flexural strength and modulus of 2-D jute/PLA green composites with jute fibers treated at different electron beam dosages.

Fig. 5 depicts the variation of the impact strength of neat PLA and random (5A) and 2-D (5B) jute/PLA green composites as a function of electron beam dosage. The jute fiber and fabric contents of each green composite were varied from 30 wt% (filled triangles) to 50 wt% (filled circles). Both show a similar tendency of the variation. The result indicates that the impact strength of neat PLA, which has been known to be low in the impact toughness, was significantly improved by incorporating either jute fabrics or chopped jute fibers into the resin. The greater jute fiber and fabric contents the greater impact strength. The 2-D jute/PLA green composites reinforced with jute woven fabrics exhibited much higher impact strength than the random jute/PLA specimens with chopped jute fibers. In the both composites, the impact strength was gradually increased with increasing the electron beam dosage to 10 kGy, showing the maximum value at 10 kGy. This was due to the enhanced jute fiber-PLA adhesion at the interfaces. In this case, the enhanced interfacial adhesion may contribute to distributing and transferring the impact energy given to the specimen to the fibers and the matrix. Above 10 kGy, the impact strength was decreased gradually down to 100 kGy. This is because the surface roughness and undulation of jute fiber can be alleviated, resulting in the decrease of the fiber-matrix adhesion. At electron beam irradiation higher than 50 kGy, the jute fiber may be somewhat damaged, leading to the poor fiber-matrix adhesion.

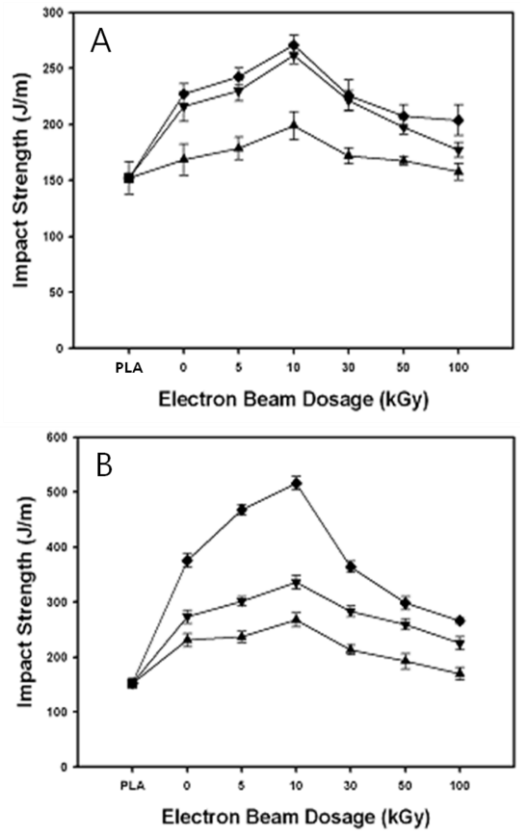

Fig. 5. Impact strengths measured for (A) random and (B) 2-D jute/PLA green composites with jute treated at different electron beam dosages with different fiber contents.

The reason for the greater impact strength of 2-D jute/PLA green composites is that the jute woven

fabrics, which have the greater aspect ratio than the chopped jute fibers used in random-type composites, can absorb the impact energy more effectively. In other words, the continuous jute fabrics can play a more effective role in reinforcing the composites and in transferring the impact energy to the neighboring fibers and the PLA matrix than the discontinuous jute fibers. The impact strength of 2-D jute/PLA with 50 wt% jute fabrics treated at 10 kGy was about 240% greater than that of neat PLA.

Fig. 6 plots the percent water absorption of neat PLA and random (6A) and 2-D (6B) jute/PLA green composites as a function of immersion time in a water bath. The jute contents were fixed to 50 wt%. In both cases, the water absorption was progressed gradually during the first ten days of the measurement, depending on the electron beam dosage irradiated to jute fibers and jute fabrics. In all cases, the water absorption was progressed very slowly after 10 days, showing the increase of the water uptake about a few percents up to 50 days. Neat PLA exhibited the water absorption about 1% after 10 days and then remained constant throughout the test. All the molded plaques were intentionally cut through the thickness direction to be exposed to water during the test. Therefore the composite specimens with jute fibers and fabrics were supposed to be readily exposed to water during immersion.

The water absorption of the composites reinforced with chopped jute fiber was in the range of 15-38% whereas that of the composites reinforced with jute fabrics was in the range of 22-31%. These values were much greater than those of other natural fiber-reinforced composites. This was because each specimen with jute was easily exposed to water and the reinforcing jute was sensitive to water. It is expected that the narrow window of the water absorption in the 2-D specimens may be due to the composite structure with jute fabrics laminated regularly. While the specimen with raw jute fibers had the water absorption about 29% after 50 days, that with raw jute fabrics had the absorption about 25%.

The water absorption of random and 2-D composites with raw jute was significantly decreased with using jute fibers and fabrics irradiated at 5 kGy and 10 kGy. The irradiation at 10 kGy exhibited the most positive effect on the water absorption resistance, indicating the minimum value of the percent water absorption. This was due mainly to the enhanced jute fiber-PLA adhesion at the interfaces. Also, it may be expected that the hydrophilic nature of jute treated with electron beam at 10 kGy was changed to more hydrophobic nature than the untreated. It has been clearly found that the increased fiber-matrix adhesion results in the reduced water uptake in a number of natural fiber-reinforced polymer composites. With the electron beam irradiation higher than 30 kGy the water absorption was rather decreased due to the decreased jute-PLA adhesion.

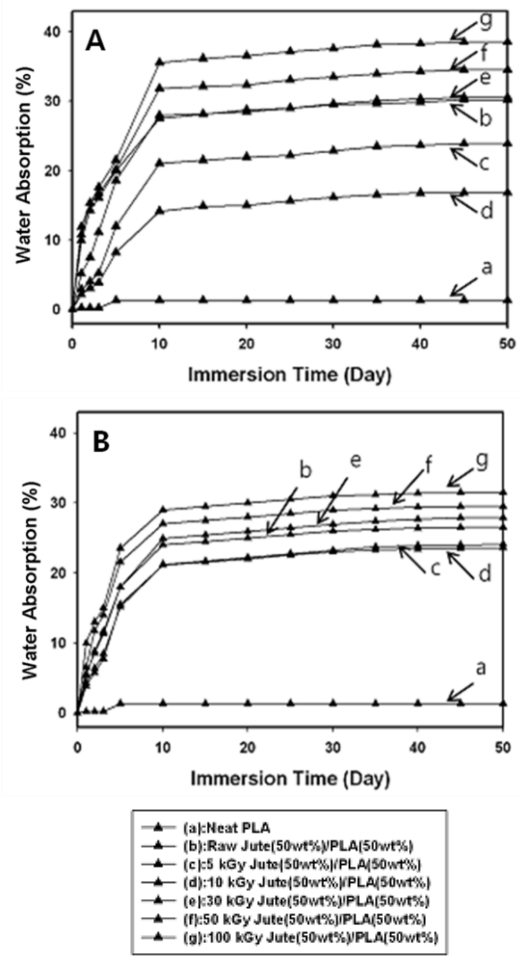

Fig. 6. Water absorption measured for (A) random and (B) 2-D jute/PLA green composites with jute treated at different electron beam dosages.

4. Conclusions

The mechanical and impact properties of neat PLA were greatly increased by reinforcing with chopped jute fibers and jute woven fabrics, depending on the jute content used. This study revealed that electron

beam irradiation at an optimal dosage of 10 kGy can effectively contribute to enhancing the interfacial, flexural and tensile properties, impact strength and long-term water absorption resistance of both random and 2-D jute/PLA green composites. The property improvement of the green composites was mainly attributed to the increased jute-PLA adhesion at the interfaces by electron beam irradiation at an appropriate dosage. The impact toughness and the water absorption resistance of 2-D jute/PLA green composites were greater than those of random jute/PLA composites and were greatest at 10 kGy, as found from the interfacial and mechanical studies.

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