Effects of Fiber Inclusion and γ Radiation on Physicomechanical Properties of Jute Caddies Reinforced Waste Polyethylene Composite

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Abstract In spite of banning thin polyethylene bag, it has been used in Bangladesh by both retailer and consumer for its convenience and affordability and most utility comes from packaging retailed goods. There is a serious concern among consumers and environmentalists about the environmental impacts on plastic bags. Due to lack of proper plastic waste management our environment is becoming more and more polluted day by day. In the present study, an eco-friendly technique was adopted for reducing such type of pollution by reusing polyethylene shopping bag as polymer matrix in composite. Jute mill wastage which is commonly known as jute caddies (JC) reinforced waste polyethylene (WPE) based low cost randomly oriented discontinuous fibrous composite was fabricated using traditional hand layup method. Fourier Transform Infrared Spectroscopy (FT-IR) was used to investigate the chemical composition of both raw jute and jute caddies. Jute caddies content in the composite was varied from 20 to 45% where, 32% JC enriched composite showed the best performance in mechanical tests. Mechanical properties such as tensile and bending of composites were evaluated following several standard tests and methods. Jute caddies reinforced low density polyethylene (LDPE) based another type composite (JC/LDPE) was also fabricated to compare with JC/WPE. In order to increase the bonding strength between fiber and matrix, both types of composites were irradiated with gamma rays of dose varied from 2.5 to 12.5 kGy where, composites irradiated with 5 kGy dose delivered the best results. Water absorption tests of the composites according to the fiber content were done successfully for knowing the water resistance properties.

Keywords: environmental pollution, polyethylene bag, jute caddies, recycling and reusing, low cost composite

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1. Introduction

Bangladesh banned the manufacture and distribution of plastic bags in March 2002 issuing on-the spot fines for consumers, and up to 10 years in jail and a fine of £9000 for manufacturers. Prior to the ban, the country consumed 9 million plastic bags a day, of which 85 per cent were littered into the waste stream [1]. The first stage of the ban applied to the capital only, to be extended nationally. But the reality shows that in Bangladesh use of polyethylene bags is rampant and increasing, whereas biodegradable packaging materials are on the decline and its use is sporadic. Due to lack of proper waste management thin poly bag is spreading all over the country. But we know that, it is one of the most dreadful elements that polluting our environment. Although, it is convenient to use poly bag but it is more harmful for environment. This enormous use of polyethylene bags led to severe environmental impacts like loss of soil fertility, blocking up of drainage and sewerage systems, causing water

logging and the spread of harmful microbes and bacteria. It obstructs the rain-water flowing to the drays. It causes low-laying areas to go under water. All these result flood and water-clogging problem and the serious flooding resulting in major loss of life. It is also a potential threat to all cultivable land as plastic bags may take between 20 and 1000 years to break down in the environment [2].

Recent survey report showed that about 10 million polyethylene bags were used every day and every year about 3000 million bags were dumped in Dhaka City alone [3]. The steadily growing use of polyethylene bags posed an ominous environmental problem in the city as well as in rural areas. The worst example of adverse effect caused due to polyethylene bags was the delaying process of recession of flood water in Dhaka city during 1998. Reducing impact of polyethylene shopping bags on environment is one of the greatest challenges for the country. From the overseas approaches it is apparent that there are two distinct techniques of reducing the impact of plastic bags on the environment. One is to decrease the amount of plastic bags used in the first place, with initiatives aimed at consumers. The Irish plastic bag levy is an example of this [4]. The second technique is aimed at the post-consumer stage, using initiatives to improve plastic bag collection, reusing and recycling facilities. One of the effective ways of management of polyethylene bags in Bangladesh is recycling/reuse of post-consumer polyethylene bags for other useful purposes. But the state of recycling in Bangladesh again posed some other environmental hazards and some means of transmission of diseases. The burning of polythene in open air produces the threat of poisonous gas like carbon mono-oxide, carbon di-oxide, hydrogen cyanide, etc. which cause health hazard to the people. In addition, people who are involved in collection and recycling of polyethylene bags are the most vulnerable group. So only reusing of polyethylene can save human as well as environment from harmful effect. One of the greatest examples of reusing plastic bags can be the fabrication of fiber reinforced polymer composite (FRPC).

Environmental concerns have resulted in a renewed interest in agro-based materials for fiber reinforced polymer composite (FRPC), and therefore issues such as recyclability and environmental safety are becoming increasingly important for the introduction of new materials and products. Nowadays there is a growing interest in the use of agro-fibers (jute, hemp, rice husk, abaca wheat straw, coir, kenaf, flax etc) as reinforcing components for thermoplastics, because they are more economical, eco-friendly and renewable than the synthetic fibers [5].

Bangladesh is one of the main jute producing countries in the world, and identified as a country of golden fiber. Jute plants both tossa jute (Corchorus olitorious L.) and white jute (C. capaularies L.) grows in large scale in Bangladesh as the temperature and the wet and humid of Bangladesh are best suited for its cultivation. The districts such as Mymensingh, Comilla, Noakhali, Faridpur, Dhaka and Pabna are famous zones of jute cultivation. About 1,743,000 tonnes of jute are produced annually in Bangladesh. It is about 50% of the total world production [6]. The world's largest jute mill namely Adamjee Jute Mills along 77 jute mills are located in the country which generate annually about 30,000 tonnes of processing waste as by-products, commonly known as jute caddies. This waste has limited use for making insulation in coach roof, electric cable, all kind of sits cushion, car false panel etc. After meeting the demand of the country, it is now exported to the foreign countries [7]. Only 10% of the total jute and jute products are consumed internally and rest are being exported. Thus Bangladesh depends largely on its foreign markets for its jute products. But the glory of the country's jute has decreased due to the increased use of artificial fiber and synthetic products that created stiff competitions for jute products. Now time has come to change this trend through development of diversified jute products worldwide. Bangladesh has taken several steps for diversification of jute products. A number of diversified jute products like geo-jute, decorative fabrics, wall coverings, curtains jute blanket, jute-plastic products, jute wool etc. are being produced and marketed both at home and abroad.

Jute caddies (JC) is chiefly nonwoven short unspinnable fiber contaminated mainly with oil, grease, remnants of bark, jute stick and clay. In recent, handmade papers have been produced from jute caddies [8]. Jute caddies may be suitable fibrous material for fiber reinforced polymer (FRP) composite as the jute based polyester composite has attracted attention of various studies [9,10] where in jute fabric, either oven or needled punched, has been used as reinforcing material. Without processing charge cost of raw material (US \$400-745/Metric Ton) of such fabric is almost three times higher in comparison to JC (US \$ 120-250/Metric Ton) [11].

Plastic bags are actually made from ethylene, a gas that is produced as a by-product of oil, gas and coal production. Ethylene is made into polymers (chains of ethylene molecules) called polyethylene or polythene which is made into pellet shape. This pellet shape polyethylene (PE) is used by plastic manufacturers to produce a range of items, including plastic bags. PE is a synthetic and nonthermoplastic polymer consisting of long polar hydrocarbon chains. It has excellent chemical inertness, stiffer and harder properties than branched polymers. The mechanical properties of PE depend significantly on variables such as the extent and type of branching, the crystalline structure and the molecular weight. Moreover, it has excellent moisture resistance and good electrical properties at high frequencies. For these reasons, it has widely been used as an insulator for high frequency currents and for cable coverings. According to sold volumes, the most important PE grades are HDPE, LLDPE and LDPE. Among them, LDPE has a wide application in industry because of it's suitable properties such as it can be processed easily and is used in different materials. It is a partially crystalline solid with a degree of crystallinity in 50-70% range, melting temperature of 100-120°C and specific gravity of about 0.91-0.94 g/cm [12]. In the present study we used LDPE as the synthetic polymer to make JC/LDPE composite. In recent several reports have also been published, where jute fibers are used as reinforcement in thermoplastics like polyethylene (PE) and polypropylene (PP) [13,14,15,16].

The aim of the present study was to reuse thin polyethylene bags and jute mill wastage for the fabrication of low cost fiber reinforced polymer composite and investigate the physico-mechanical properties.

2. Experimental

2.1. Materials

Jute mill wastage (jute caddies) was collected from Janata Jute Mills Limited, Palash, Ghorashal, Narsingdi, Bangladesh. Polyethylene shopping bags were collected from dustbins and grocery shops. Low-density polyethylene (LDPE) pellets were purchased from Polyolefin Company Ltd. Singapore.

2.2. Methods for Making Composites

2.2.1. Processes of Reusing Polyethylene

The reusable polyethylene shopping bags which are composed of low density polyethylene (LDPE) were collected from roadside dustbins or even directly from grocery shops. In this way, both post-consumer and postindustrial polyethylene was collected. After collecting, the polyethylene products are prepared for melting by being cut into small pieces with scissors. The polyethylene chips were then washed for removing all residues of products originally contained in the plastic items and various other contaminants such as paper labels, dust, dirt etc. A particular wash solution consisting of an alkaline, cationic detergent and water were used to effectively get rid of all the contaminants on the plastic material, making sure that all the plastic bits are clean and ready for the final step. Polyethylene chips were then dried in an oven for half an hour at 35° C.



Figure 1. Waste polyethylene shopping bags and jute caddies

2.2.2. JC/WPE Composite Fabrication

Polyethylene sheets of 1-1.5 mm thickness were prepared from polyethylene chips by pressing at 115°C for 5 min between two steel molds under 5 bar consolidation pressure in the heat press machine (Carver, INC, USA Model 3925). Prepared LDPE sheets were then cooled to room temperature (25°C) for 15 minutes. The resulting polyethylene sheet was cut into desired size for composite fabrication. Composites were prepared by sandwiching 2 layers of short jute fibers (jute caddies) between 3 sheets of pre-weighted polyethylene. The sandwich was then placed between two steel molds and heated at 150°C for 5 minutes to soften the polymer prior to pressing 5 bar pressure for 3 minutes and finally it was allowed to cool naturally to room temperature for about 10 minutes. JC/WPE composite panels with different fiber weight percentage were prepared and the specimens of the required dimensions were cut and used for several testing.

2.2.3. JC/LDPE Composite Fabrication

The LDPE sheets (1-1.5 mm thickness) are prepared by heating the polymer granules at 115°C for 5 min between two steel plates under a pressure of 5 ton. The molds were then cooled for 3 min in a separate press machine under 5 bar pressure at room temperature. The resulting LDPE sheet was then cut to desired size and kept in the plastic bag for composite fabrication. Jute caddies reinforced low density polyethylene composite was then prepared at 135°C and 5 ton pressure using the same heat press machine. JC/LDEP composite panels with different fiber weight contents (%) were prepared and the specimens of the required dimensions were cut and used for testing.

2.3. FT-IR Analysis

In order to identify the functional groups present in the raw jute and jute caddies samples, Fourier transform infrared spectroscopy (FTIR) studies were conducted using a SHIMADZU FT-IR spectrometer (IR Prestige-21) with a resolution of 4 cm⁻¹. The samples were oven dried at 105°C for 4-5 h, mixed with KBr in a ratio of 1:100 (w/w) and pressed under vacuum to form pellets. The FTIR spectrum of the samples was recorded in the

transmittance mode in the range of $4000-400 \text{ cm}^{-1}$ with an average of 30 scans.

2.4. Application of Gamma Radiation

The composite as well as the matrix specimens were irradiated using a Co-60 gamma source (25 kci) located at the Atomic Energy Research Establishment, Dhaka, Bangladesh. Total dose varied from 2.5 to 12.5 kGy at the rate 6 kGy/h. In gamma radiation plant, model gamma beam 650 is loaded with source GBS-98 that comprises 36 double encapsulated capsules.

2.5. Determination of Mechanical Properties

Different mechanical properties of the composites were conducted through tensile and flexural tests. For each test and type of composites, five specimens were tested and the average values (tensile and bending strength, modulus, elongation at break and bending stress at 30% strain) with standard deviation were reported.

Tensile tests were performed according to ASTM D 638-01 [17] using a Universal Test Machine (model H50KS-0404, Hounsfield series S, UK). The total load capacity was 5,000 N; efficiency was within ± 1 %. Test samples were conditioned at 25°C and 50% relative humidity for several days before testing and all the tests were performed under the same conditions. The dimensions of the test specimens were (ISO 14125): 60 mm × 15 mm × 2 mm. Specimens were placed in the grips of a Universal Test Machine at 50 mm grip separation and pulled with a cross-head speed of 10 mm/min until failure.

Static flexural tests were carried out according to ASTM standard D-790 using the same testing machine mentioned above at the same cross-head speed. Specimens of 150 mm length and 15 mm width were cut and placed between two points or supports of the apparatus and initiating a load using a third point or centre. In the present study the recommended span to depth (thickness of the specimen) ratio was 16:1. The specimens were tested at a crosshead speed of 2 mm/min. The tests were conducted on the same machine used for tensile testing.

2.6. Water Absorption Test

Water absorption tests of the composites were conducted on the basis of fiber loading. All the specimens were dried in an oven for half an hour at 105°C. After then, those were placed in a desiccators to cool and immediately upon cooling the specimens were weighted. The specimens were then soaked in a beaker having three quarters deionized water for 24 hours at temperature 23°C. Specimens were then removed, patted dry with a lint free cloth, and reweighed by a Mettler balance. The whole test was performed according to the standard ASTM D570. The water absorption quantity was measured by a weight difference methodology.

3. Results and Discussion

3.1. FT-IR Analysis

FTIR spectrum of raw jute is shown in Figure 2 which depicts information about the chemical composition on the surface of raw jute fiber. In the spectrum a short and broad

peak which are observed at 3383.14 cm⁻¹ indicate the presence of H-bonded -OH group. Another narrow pick observed at 2912.51 cm⁻¹ indicates C-H stretching vibration. There is a very narrow band observed at 1735.93 cm⁻¹ indicates C=O group stretching. Another three narrow picks viewed at 1647.21 cm⁻¹, 1373.32 cm⁻¹ and 1249.87 cm⁻¹ show the presence of -C-C- stretching, C-C stretching (in ring) or C-H bending and C-H bending respectively. Another two very short peaks observed at 601.79 and 667.37 cm⁻¹ may be O-H bend (out-of-plane).

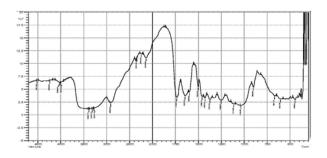


Figure 2. FT-IR spectrum of raw jute

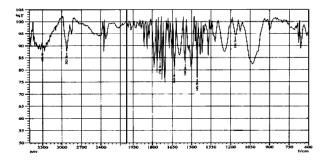


Figure 3. FT-IR spectrum of jute caddies

On the other hand, FTIR spectrum of raw jute caddies is shown in Figure 3 which illustrates information about the chemical composition on the surface of raw jute caddies. In the spectrum a short and broad peak which are observed at 3301.98 cm⁻¹ indicate the presence of Hbonded -OH group. Another narrow pick observed at 2923.70 cm⁻¹ indicates C-H stretching vibration. There is a very narrow band observed at 1742.54 cm⁻¹ shows C=O group stretching. Another three narrow picks viewed at 1634.46 cm⁻¹, 1549.54 cm⁻¹ and 1456.90 cm⁻¹ signify the presence of -C-C- stretching, C-C stretching (in ring) or C-H bending and C-H bending respectively. A very short peak observed at 1163.54 cm⁻¹ may be $-CH_2X$ or C-O or C-N stretching vibration. From the FT-IR spectra of above mentioned two results it has been observed that the absorption frequency of raw jute is not quite similar to jute caddies. The FT-IR results indicate that some chemical reactions occurred by acetylation of jute caddies during processing of raw jute in mills as the jute batching oil (JBO) along with a chemical emulsifier and water in different proportions were used for the production of fine yarns from raw jute.

3.2. Effect of Fiber Inclusion on Mechanical Properties

Figure 4 shows information about the tensile properties such as tensile strength (TS), tensile modulus (TM) and elongation at break (Eb) of the five different types of composites on the basis of jute caddies content. It is observed that tensile strength (TS) of the JC/WPE composite gradually increased from 19.7 to 21.75 MPa for the composite made up to 31.75% jute caddies (JC). And then, it decreased continuously up to 42.25% JC. Similarly, tensile modulus (TM) of the composite initially increased and reached the maximum value, 690 MPa at 31.75% JC loaded composite and then showed a continuously decreasing trend.

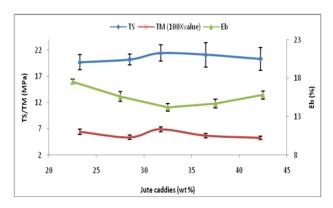


Figure 4. Effect of jute caddies content on the tensile properties of the composite (JC/WPE)

On the other hand, elongation of the composites at break was observed to decrease gradually and fall at 14.2% for the composite made with 31.75% jute caddies. Extension of the composites then started to rise regularly and continued up to 42.25% jute caddies. From the definition of strain, we know that it is a function of length extension. If the length extension is more then strain becomes more.

Materials	Fiber content (wt%)	Polymer content (wt%)	Tensile strength (MPa)	Tensile modulus (MPa)	Elongation at break (%)	Bending strength (MPa)	Bending modulus (MPa)	Stress at 3.5% strain (MPa)
LDPE sheet	0	100	14.7	415	105.85	31.55	640	14.55
WPE sheet	0	100	12.2	375	85.65	26.2	610	11.3
JC/LDPE composite	30	70	23	680	18.55	41.25	450	30.35
JC/WPE composite	30	70	19.5	595	15.35	35.45	415	25.4

Table 1. Tensile and Flexural Properties of LDPE and WPE Sheet and JC/LDPE and JC/WPE Composite

On the other hand, tensile modulus as well as tensile strength is inversely proportional to strain. Thus for low length extension, obviously value of TS and TM will be more. Figure 5 represents information about the flexural properties such as bending strength (BS), bending stress at 30% strain and bending modulus (BM) of five composites made with different jute caddies content. It can be clearly seen that both bending strength (BS) and stress at 30%

strain started up with a gently increasing trend and came to the maximum value, 35.25 and 27.5 MPa respectively for 31.75% caddies loaded composite. Bending modulus (BM) on the other hand, sharply increased initially and attained at the maximum value, 940 MPa for the same composite. And then, it declined dramatically and continued up to the final composite.

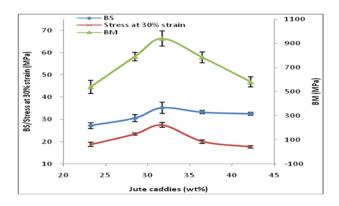


Figure 5. Effect of jute caddies content on the flexural properties of the composite (JC/WPE)

Firstly, at lower levels of jute caddies content, the composite expresses poor mechanical properties (tensile and flexural) due to poor fiber population and low load transfer capacity to one another. As a result, stress gets accumulated at certain points of the composites and highly localized strains occur in the matrix. Secondly, at intermediate levels of jute caddies loading (30%), the population of the fibers is just right for maximum orientation and the fibers actively participate in stress transfer which, result higher mechanical properties. Thirdly, high levels of jute caddies content show the increased population of fibers, which may lead to agglomeration and stress transfer becoming blocked, as a result composite delivers poor mechanical properties [18].

3.3. Effect of Gamma Radiation on Mechanical Properties

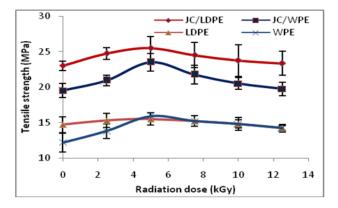


Figure 6. Effect of gamma radiation on the tensile strength of the composites and polymer matrix sheets

Figure 6 expresses information about the tensile strength of the composites (made with 30% JC) JC/LDPE and JC/WPE as well as the polymer matrixes (same thickness that was used to fabricate composite) LDPE and WPE against radiation dose. It can be clearly seen that tensile strength of JC/LDPE composite increased moderately from the initial value 23 MPa (at 0 kGy i.e.

non-irradiated) to the maximum value 25.5 MPa (at 5 kGy). After then TS of the composites constantly decreased. Similarly, TS of JC/WPE composite increased gradually from the primary value 19.5 MPa (non-irradiated) to the maximum value 23.5 MPa at 5 kGy and then decreased regularly. By contrast, TS for sheet of low density polyethylene (LDPE) had slightly increasing trend whereas sheet of waste polyethylene (WPE) had gradually increasing trend. TS value of both matrixes coincided with a value 15.2 MPa at 7.5 kGy and then slightly decreased together with the same value.

Figure 7 delivers information about the variation of bending strength (BS) of jute caddies reinforced waste polyethylene (JC/WPE) and jute caddies reinforced pure polyethylene (JC/LDPE) composite made with 30% jute caddies as well as the low density polyethylene (LDPE) and waste polyethylene (WPE) sheet matrixes (3-4.5 mm thickness) with the increase of radiation dose. It can be clearly seen from the figure that initial BS value of JC/LDPE was higher (41.25 MPa) than JC/WPE (35.45 MPa) due to the degradation fact of PE. At first both the composites showed gradually rising trend of BS from the primary value (at 0 kGy, i.e. non-irradiated) with the increase of radiation dose and gained the maximum value 45.75 and 40.5 MPa at 5 kGy. After then, BS value of the composite decreased regularly with the increase of radiation dose. On the other hand, BS value of low density polyethylene (LDPE) sheet matrix slightly rose whereas waste polyethylene (WPE) rose gradually and came to the maximum value 33.95 and 30.85 MPa respectively at 5 kGy. Then BS value of both matrixes showed a parallel decreasing trend with the further increase of radiation dose.

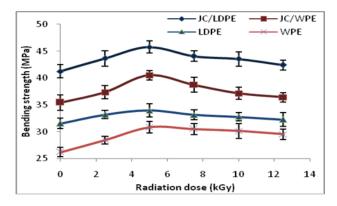


Figure 7. Effect of gamma radiation on the bending strength of the composites and polymer matrix sheets

Gamma ray has higher penetrating power and is a very strong type of ionizing radiation source. It exhibits significant effects on polymeric materials (matrix and cellulose) through producing ionization and excitation. The ionizing radiation produces three types of reactive species in the polymer matrix. These are ionic, radical, and peroxide. The peroxides species are produced only when polymers are irradiated in the presence of oxygen. In the present study, specimens were irradiated in presence of oxygen. Both the polymer matrix and cellulose may undergo cleavage or scission i.e., the polymer molecules may be broken into smaller fragments. Afterwards, rupture of the chemical bonds yields fragments of the large polymer molecules. The free radicals generated may react to change the structure of the polymer and alter the physical properties of the materials. It also may undergo

cross linking i.e., the molecules may be linked together to form a large molecules [19]. That is why, when a jute caddies reinforced polyethylene composite as well as matrixes were irradiated by gamma radiation then tensile and bending strength were increased with radiation doses and decreased after attaining a maximum value at a certain dose. As the gamma radiation doses were further increased TS and BS of polymer sheets and composites were decreased, which may be associated due to the ionizing radiation degradation of cellulose backbone at higher gamma doses [20]. During degradation, it is believed that, there will be loss in strength due to primary bond breakage in the cellulose constituents and, therefore, be related to changes taking places in the middle lamella, which reduce the ultimate cell.

3.4. Effect of Fiber Inclusion on Water Absorption Behavior

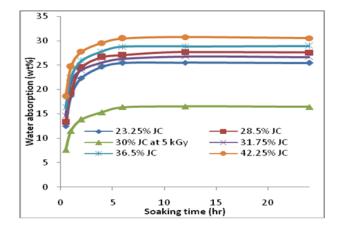


Figure 8. Effect of soaking time on water absorption of JC/WPE composites at different wt% of jute caddies

Figure 8 depicts information about the water absorption capacity of six different type composite specimens during 24 hours. The trends of water absorption by the composites were quite common except the irradiated composite made with 30% JC. The figure shows that the quantity of water absorption by the composites is increasing with the additional fiber content and soaking time. Jute fiber being a hydrophilic nature has the tendency to absorb water quickly. For this reason, all six specimens absorbed the maximum amount water within first six hours after which no considerable amount of water absorption was observed. The composite that has more jute fiber obviously exhibits more water absorption capacity. This is why, the composite made with 42.25% JC showed the greatest amount of water absorption (30.5%) while; composite made with 23.25% JC showed the lowest amount of water absorption (25.45%). That is 19.84% more water absorption was gained by the specimen made with 81.72% more JC. Water absorption property by the composite also may depend on the composite uniformity. Probably, for this reason composite made with 28.5% JC represents more water absorption than composite made with 31.75% JC. By contrast, composite made with 30% JC showed the best water resistance performance after irradiation with 5 kGy gamma dose.

The phenomenon of water absorption by the jute based composite can be explained on the basis of its hydro -d-

glucose cellulose structure of jute, specially containing three hydroxyl (-OH) groups. Presence of hydroxyl groups in the jute cellulose is due to the intermolecular hydrogen bonds with other cellulose molecules as well as with other hydrogen groups of moist air. However, the groupings of long-chain cellulose molecules in the cell contain crystalline and amorphous regions. In the crystalline region, -OH groups of adjacent cellulose molecules are mutually bonded or cross-linked. For that reason, there are no sites to occupy water within crystalline groups is accessible for absorption of water [21]. However, the decrease of water absorption by post irradiated composites attributed to the fact that gamma irradiation reduced the -OH group as well as increased crystalline regions in jute fiber through cross-linking which in turns decrease amorphous regions.

4. Conclusions

In Bangladesh, in spite of banning polyethylene shopping bag, its production and uses could not be stopped. Proper management of postconsumer plastic wastes through only reusing (since recycling is harmful) can solve the problem and it is an emergency need for the country. Moreover, being jute plenty growing country, biodegradable jute caddies is available here. Large scale low cost composite production by using wastage polyethylene and jute caddies may be a great example of beneficiary for commercial and environment. Physicomechanical tested composite fabricated with 30% jute caddies and post irradiated with 5 kGy gamma dose was found suitable in diverse fields where, higher strength is not a mandatory factor.

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