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Effect of gamma radiation on the performance of jute fabrics-reinforced polypropylene composites

Haydaruzzaman [a], Ruhul A. Khan [b], Mubarak A. Khan [b,*], A.H. Khan [a], M.A. Hossain [a]

[a] Department of Physics, Jahangirnagar University, Savar, Dhaka, Bangladesh
[b] Radiation and Polymer Chemistry Laboratory, Institute of Nuclear Science and Technology, Bangladesh Atomic Energy Commission, G. P.O. Box 3787, Dhaka 1000, Bangladesh

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ABSTRACT

Jute fabrics-reinforced polypropylene (PP) composites (50% fiber) were prepared by compression molding. Composites were fabricated with non-irradiated jute fabrics/non-irradiated PP (C-0), non-irradiated jute fabrics/irradiated PP (C-1), irradiated jute fabrics/non-irradiated PP (C-2) and irradiated jute fabrics/irradiated PP (C-3). It was found that C-3 composite performed the best mechanical properties over other composites. Total radiation dose varied from 250–1000 krad and composites made of using 500 krad showed the best results. The optimized values (C-3 composites) for tensile strength (TS), bending strength (BS) and impact strength (IS) were found to be 63 MPa, 73 MPa and 2.93 kJ/m^2, respectively.

1. Introduction

Composite materials are widely used in civil, industrial and military applications mainly because of their excellent tensile and bending properties. Synthetic fiber-reinforced thermoplastic composites attracted much attention due to its better durability and moisture resistance properties. However, the manufacture, use, and removal of traditional composite structure made of glass, carbon and aramid fibers are considered negative due to growing environmental consciousness. For this reason, alternative reinforcement with natural fiber in composites has gained much attention having low cost, low density, CO_2 neutrality, biodegradability and recyclable nature (Mohanty et al., 2000a,b,c). Among all the natural fibers, jute appears to be one of the most useful, inexpensive and commercially available lignocelluloses fiber. Cellulose is the main component of jute fiber (approximately 64 wt%) (Hassan et al., 2003).The elementary unit of a cellulose macromolecule is anhydro-D-glucose, which contains three hydroxyls (–OH). This hydroxyl form hydrogen bonds inside the macromolecule itself (intra-molecular) and between other cellulose macromolecules (intermolecular) as well as with hydroxyl groups from moist air. Therefore, jute fabrics are hydrophilic in nature and their moisture content can reach 12.6% (Khan et al., 2006). Composites of hydrophilic fiber with hydrophobic or non-polar polymer matrix result in poor mechanical properties compared to pure polymer (Hassan et al., 2005). To overcome these kinds of drawbacks, many attempts, such as physical and chemical treatments, lead to changes in the surface structure and surface energy of the fibers. However, for jute to survive in the face of hard competition with synthetic products, it must be improved in its physical properties to retain its inherent status. To prevail over the drawbacks of jute fiber, high-energy gamma radiation has been employed successfully for significant physical and chemical changes as well as changes in surface structure and surface energy of the fibers. Many researchers are working on jute fibers and jute fiber-reinforced composites. Some jute reinforced composites with urethane oligomer under gamma radiation were prepared and characterized (Basher et al., 1996). Khan et al. also prepared and characterized jute–polycarbonate and jute–biopol composites and reported probable alternative of synthetic composites (Khan et al., 2003 and Khan et al., 2005). Several reports have documented where jute fibers are used as reinforcement in thermoplastics like polyethylene and polypropylene (PP) and thermo-sets like unsaturated polyester and epoxy resin (Mohanty et al., 2000a,b,c; Ali et al., 1997, Khan et al., 2001a,b; Ali et al., 1999; Mohanty and Misra, 1995; Joseph and De Carvalho, 2000; Wolcott, 1993; Czvikovszky, 1995). The influence of ionizing radiation on polymers, particularly gamma radiation, has been studied quite extensively over the past few decades. Ionizing radiation such as gamma radiation is known to deposit energy in solid cellulose by Compton scattering and the rapid localization of energy within molecules produced trapped macrocellulosic radicals. The radicals thus generated are responsible for changing the physical, chemical and biological properties of cellulose fibers (Jochen and Andrzej, 1997; Saheb and Jog, 1999; Valdez-Gonzalez et al., 1999). Polypropylene is an amorphous thermoplastic

* Corresponding author. Tel.: +880 2 7788245; fax: +880 2 7701335.
E-mail address: makhan.inst@gmail.com (M.A. Khan).

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polymer and widely used as engineering thermoplastic, because it possesses several vital and useful properties such as transparency, high mechanical strength, high heat distortion temperature, low-moisture pickup and good dielectric properties. PP is also very suitable for filling, reinforcing and blending. PP with fibrous natural polymers of biomass origin is one of the most promising routes to create natural–synthetic polymer composites (Lua and Netravali, 1999; Benedeto et al., 1989; Karmaker and Hinrichsen, 1999).In this investigation, jute fabrics and PP sheets were pretreated using gamma radiation varying total doses from 250 to 1000 krad, and then composites were fabricated by heat press. Effect of gamma treatment of reinforcing agent and matrix material on composites was investigated. Mechanical and electrical properties of the composites made of different combinations of gamma treatment were measured and tried to found the effect of gamma radiation on the composites. Simulated weathering, water uptake and electrical properties of the different types of composites were investigated and compared to the untreated composites.

2. Experimental

2.1. Materials

Jute fabrics (bleached commercial grade made of tossa jute) were obtained from Bangladesh Jute Research Institute (BJRI), Dhaka, Bangladesh. Polypropylene was collected from Polyolefin Company Ltd., Singapore.

2.2. Preparation of PP sheets

Polypropylene sheets of 0.25–0.30 mm thicknesses were prepared by heat press (Carver Inc. USA with hydraulic unit model 3925) at 190 °C for 5 min between two steel plates under a pressure of 5 tons.

2.3. Irradiation

Jute fabrics and PP sheets were irradiated using a Co-60 gamma source. In gamma radiation, a Co-60 source (25 kCi) model gamma beam 650 is loaded with source GBS-98 that comprises 36 double encapsulated capsules. Type C-252 loaded with Co-60 pellets was used. Jute fabrics and PP sheets were pretreated with gamma radiation varying different doses (250–1000 krad).

2.4. Fabrication of composite

Composites were prepared by sandwiching four layers of jute fabrics between five layers of pre-weighted PP sheets at 190 °C for 5 min between two steel plates under a pressure of 5 tons. The composite was cooled to room temperature, then cut to the desired size using band saw and packaged in a polyethylene bag and then kept in the desiccators prior to further testing.

2.5. Mechanical tests

The tensile properties of PP sheets and composites were determined directly with the help of universal testing machine (INSTRON, model 1011, UK) using gauge length of 20 mm and crosshead speed 10 mm/min. Tensile and bending properties were carried out following DIN-53455 and DIN-53452 standards, respectively. Charpy impact strength (IS) was measured by an impact tester (MT-3016) according to the DIN EN ISO 179 standard in the flat wise, unnotched mode. 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.

2.6. Accelerated weathering aging

Both treated (C-3) and untreated (C-0) composite samples were treated using simulated weathering tester from Q-panel Co (model QUV, USA). The weathering testing was performed in alternating cycles of sunshine over 4 h (65 °C ± 2 °C) and condensation for 2 h (45 °C ± 2 °C). This aging test was carried out for 600 h.

2.7. Water uptake

Both composites (C-0 and C-3) were used to determine water uptake by soaking the samples in a static water bath at room temperature. The wet sample was wiped several times using tissue papers. Water uptake was calculated from the mass gain of the samples after the treatment. The amount of water uptake was monitored periodically up to 30 h.

2.8. Electrical tests

For the measurement of dielectric properties (dielectric constant and loss tangent), composite (C-0 and C-3) samples were cut into very small pieces (rectangular shape), and then powdered and made tablets. These tablets were used for dielectric measurement. A Marconi Universal bridge (TF1313A) operating at 10 kHz was used for measuring dielectric properties. For the measurement of the temperature dependence dielectric constant, the sample was placed in the dielectric cell and the cell was placed inside a cylindrical furnace. Varying the input power to the furnace varied the cell temperature. Capacitance (C) and loss tangent ($\tan\delta$) of the samples were measured at different temperature with the help of a chromel–alumel thermocouple, which is very sensitive. Thermo electromotive force (e.m.f.) was measured by a digital multimeter (DL-706) and the corresponding temperature was found out from the calibration chart. The dielectric constant and loss tangent of the composites were measured by using bridge technique (two-electrode method) over the temperature range from 50 to 120 °C at frequency 10 kHz.

3. Results and discussion

3.1. Tensile strength (TS) and bending strength (BS)

Jute fabrics-reinforced PP composites were prepared by compression molding where jute content was about 50% which was optimized earlier (Khan et al., 2008). In order to make a systematic study of the effect of gamma radiation, three types of composites were made using: (i) non-irradiated jute fabrics/irradiated PP (C-1), (ii) irradiated jute fabrics/non-irradiated PP (C-2) and (iii) irradiated jute fabrics/irradiated PP (C-3). Radiation treatment (total gamma dose) varied from 250–1000 krad. The effect of gamma radiation on the mechanical properties (TS and BS) of PP sheet was also monitored. The results are shown graphically in Figs. 1 and 2. From the Fig. 1, it is clear that with the increase of total gamma dose, the TS of the PP sheet increased from 30.15 (indicated as 0 krad, i.e., untreated) to 38.33 MPa (500 krad) and then decreased to 34.13 MPa at 1000 krad. So, by using 500 krad of gamma dose on the PP sheet, the TS values increased about 27% compared to that of untreated PP sheet. For all the composites made of different combinations of jute and PP (C-1, C-2 and C-3), TS values increased up to 500 krad then

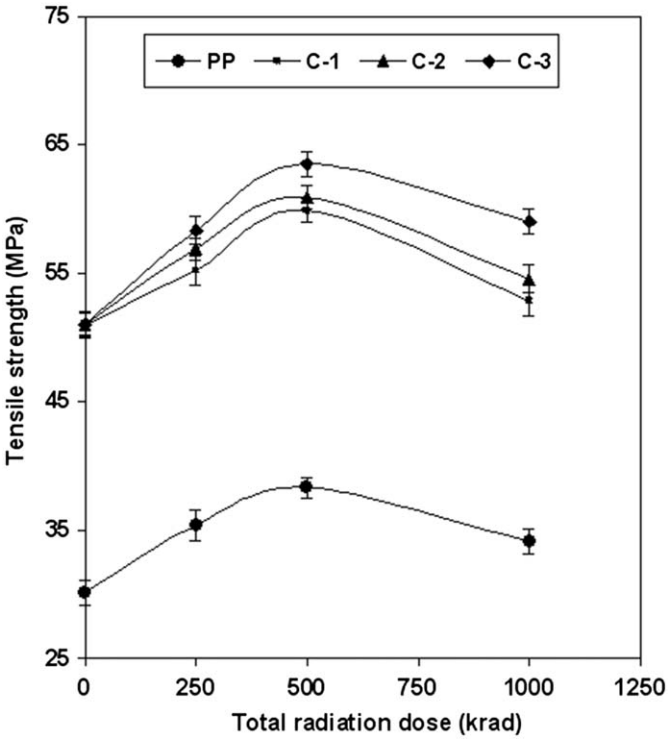


Fig. 1. Tensile strength (TS) of irradiated composite against radiation dose.

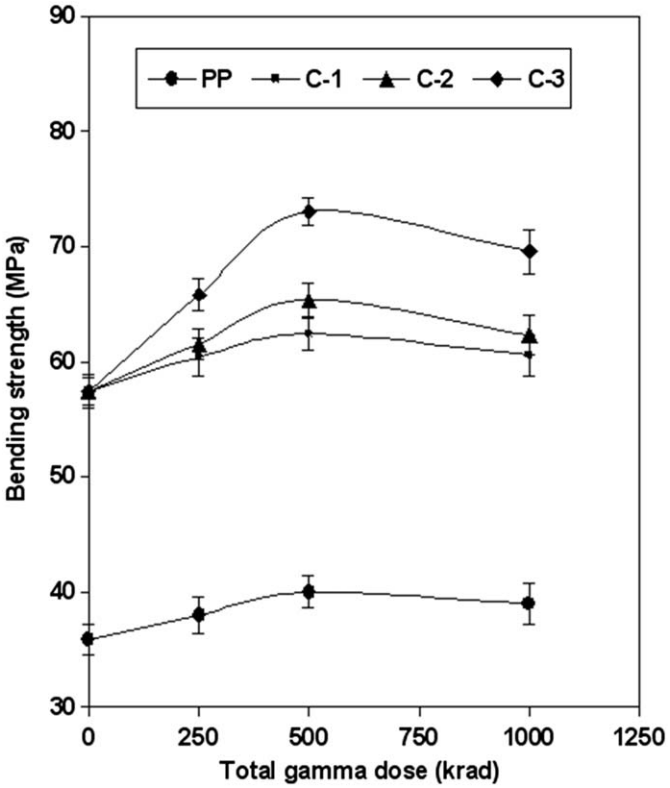


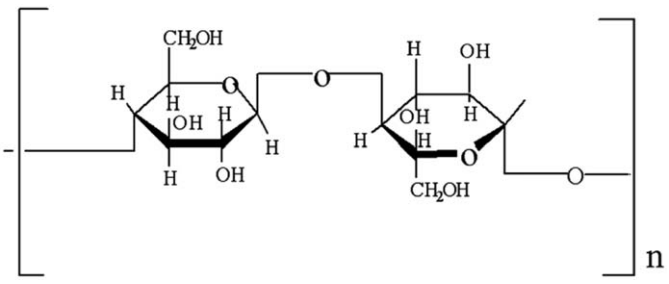
Fig. 2. Bending strength (BS) of irradiated composite against radiation dose.

decreased. The maximum TS values were found 59.85, 60.88 and 63.49 MPa, respectively, for C-1, C-2 and C-3 composites. This is about 17%, 19% and 25% enhancement of TS for C-1, C-2 and C-3 composites, compared to untreated composite.

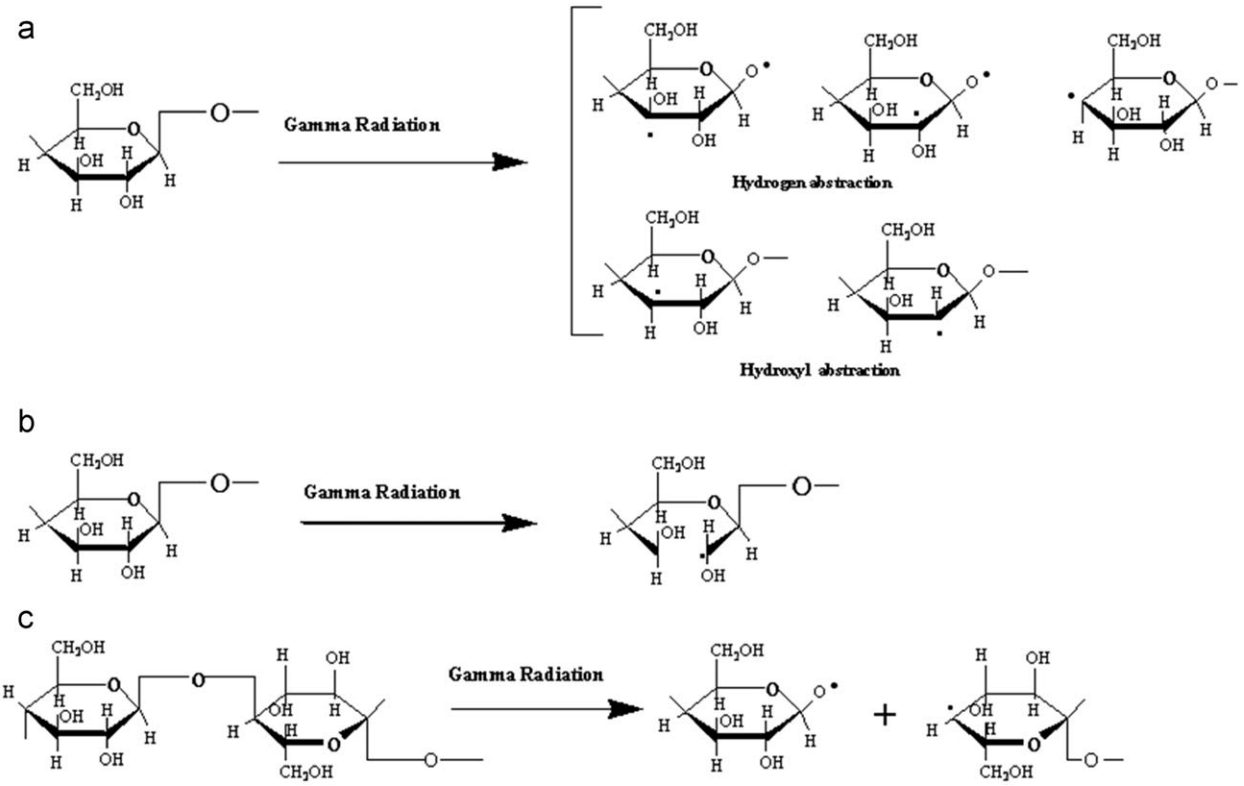
From the above findings, it can be concluded that using 500 krad of gamma radiation on PP sheet, the TS improved about 27%. But using these pretreated (500 krad) PP matrix and untreated jute, composite (C-1) did not receive the same effect. The TS values improved in C-1 composite is 17% which is slightly lower improvement than expected (27%). Using pretreated jute and pretreated PP (C-3), the improvement of TS found 25% which is much lower than our expected improvement of TS (27%+19% = 46%) in the composite. From the gamma treatment, synergistic effect did not appear. The cause behind the antagonistic effect may be attributed to the fabrication steps of the composites. But the improvement of TS is also a promising way to enhance the properties of the jute fiber-reinforced composites.

From the Fig. 2, it is clear that with the increase of total gamma dose, the BS of the PP sheet increased from 35.8 to 40 MPa (500 krad) and then slightly decreased to 39 MPa at 1000 krad. Thus, it is clear that using 500 krad of gamma dose on PP sheet, the BS values increased about 12% compared to untreated PP sheet. For composites made of different combinations of jute and PP (C-1, C-2 and C-3), BS values increased up to 500 krad then decreased. The maximum BS values were found 62.4, 65.4 and 73.1 MPa, respectively, for C-1, C-2 and C-3 composites. For C-1, C-2 and C-3 composites, about 9%, 14% and 27% increased in BS were found compared to untreated composite. Using gamma-treated jute and PP composite (C-3), the improvement of BS found 27% which is quite close to our expected BS (PP-12%+Jute-14% = 26%) of the composite. This is a significant finding in this research. Jute is a cellulosic structure with a chain of homo-polysaccharide consisting of identical monomeric units of β-D-glucopyranose, as shown in Scheme 1.

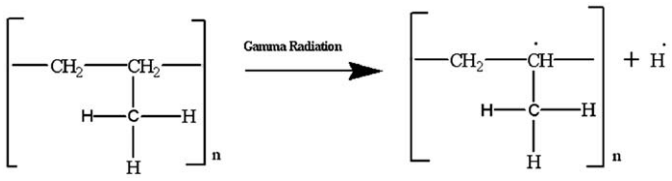
The chains are cemented together by lignin and hemicellulose, which are present at 12–14% and 22–24%, respectively. In addition to these, the structure contains other minor constituents such as wax and fats, inorganic salts, and pigments. When jute samples are subjected to high-energy radiation (gamma), radicals are produced on the cellulose chain by hydrogen and hydroxyl abstraction, as explained in Scheme 2(A). Gamma radiation also ruptures some carbon–carbon bonds and produces radicals (Scheme 2(B)). Chain scission may also take place to form other radicals (Scheme 2(C)). The ionizing radiation produces three types of reactive species in polymer (Scheme 3). These are ionic, radical and peroxide. The peroxides species are produced when polymers are irradiated in the presence of oxygen (Charlesby 1958; Charlesby and Swallow, 1959; Dole et al., 1959). In the present study, samples were irradiated in presence of oxygen. The mechanism of peroxides formation is given in Scheme 4. Peroxide reacts with cellulose and produces cellulose diperoxides (POOP) and hydroperoxides (POOH) by a radical chain reaction process. The reactions occurs in three steps: activation (1), propagation (2–3) and termination (4–7) and is given in Scheme 4. The effect of high-energy radiation on organic polymers (such as jute and PP) produced ionization and excitation, as a result some free radical produced and shown in Schemes 2 and 3.



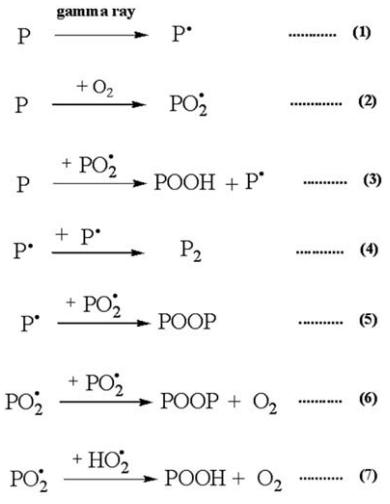
Scheme 1. Monomeric units of β-D-glucopyranose of jute.



Scheme 2. Modes of free radical generation on irradiated jute fiber. Radicals are formed after C–H, C–O or C–C bond cleavages: (A) hydrogen and hydroxyl abstraction, (B) cycle opening and (C) chain scission.



Scheme 3. Effect of gamma radiation on PP.

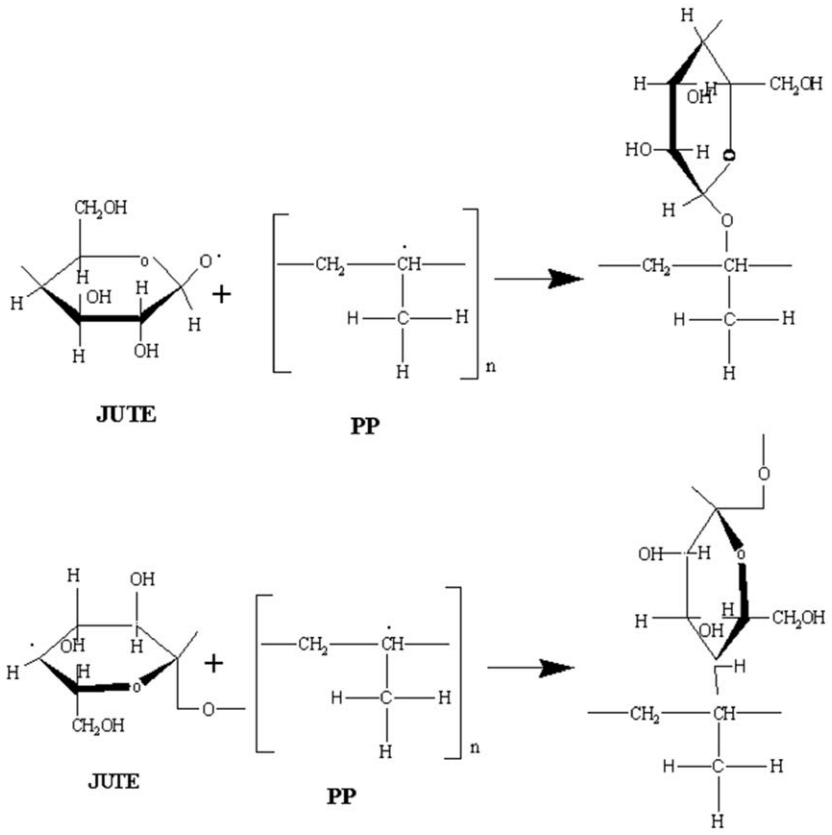


Scheme 4. Formation of peroxides and reaction between jute and peroxide (P is the jute cellulose and P corresponds to all possible radical formations according to Fig. 4).

The polymer (PP and cellulose) may undergo cleavage or scission (i.e., the polymer molecules may be broken into smaller fragment). Subsequent rupture of chemical bonds yields fragments of the large polymer molecules. The free radicals thus produced may react to change structure of the polymer and after the physical properties of the materials. It also may undergo cross-linking (i.e., the molecules may be linked together into large molecules) (Saheb and Jog, 1999; Valdez-Gonzalez et al., 1999; Wolcott, 1993). Gamma irradiation also affects the polymeric structure of the fiber and produces active site. Gamma irradiation of PP may result in cross-linking which produces higher mechanical properties up to a certain dose. Active sites inside the PP matrix might be produced by the application of gamma radiation. A probable reaction mechanism between PP and jute is given in Scheme 5. As a result, better bonding between jute and PP matrix may occur. This may be the reason behind the increased mechanical properties of pre-irradiated jute fabrics/pre-irradiated PP composites.

3.2. Tensile modulus (TM) and bending modulus (BM)

The effect of gamma radiation on tensile modulus and bending modulus of composites are shown in Figs. 3 and 4. Investigation showed that TM and BM of all samples were increased significantly compared to the non-treated one. Maximum tensile modulus and bending modulus for C-3 composite were found to be 970 MPa and 3 GPa at 500 krad dose of gamma irradiation. Maximum values of TM and BM for C-1 composites were found to be 828 MPa and 2.24 GPa. On the other hand, for C-2 composites, the TM and BM were 933 MPa, and 2.44 GPa, respectively. For C-1 composites, about 87% increased in TM and 37% increased in BM and for C-2 composite, about 110% increased in TM and 49% increased in BM compared to untreated composites were found when it was made using 500 krad of total gamma dose. The modulus values for all samples at 500 krad dose were significantly higher than the non-treated samples.



Scheme 5. A possible reaction mechanism between jute and PP due to gamma radiation.

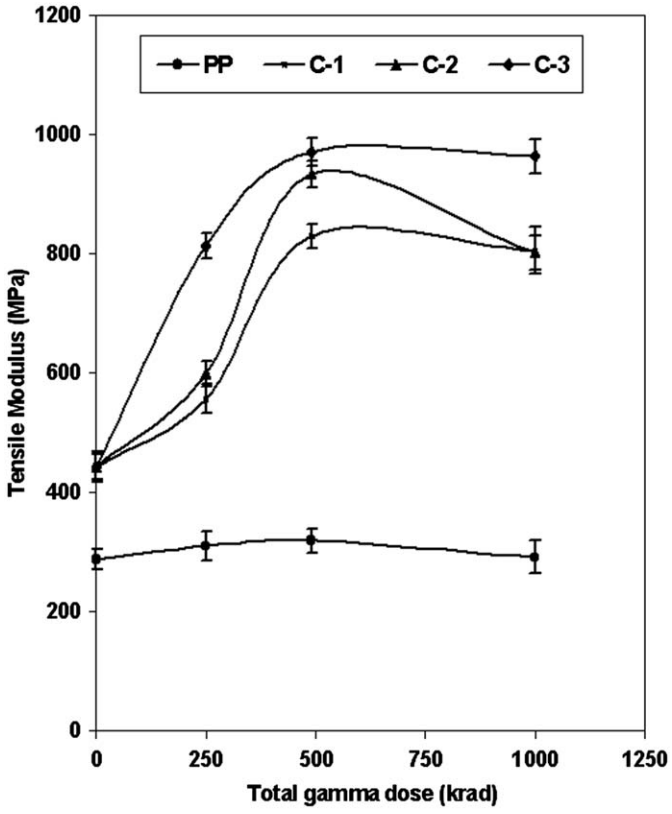


Fig. 3. Tensile modulus (TM) of irradiated composite against radiation dose.

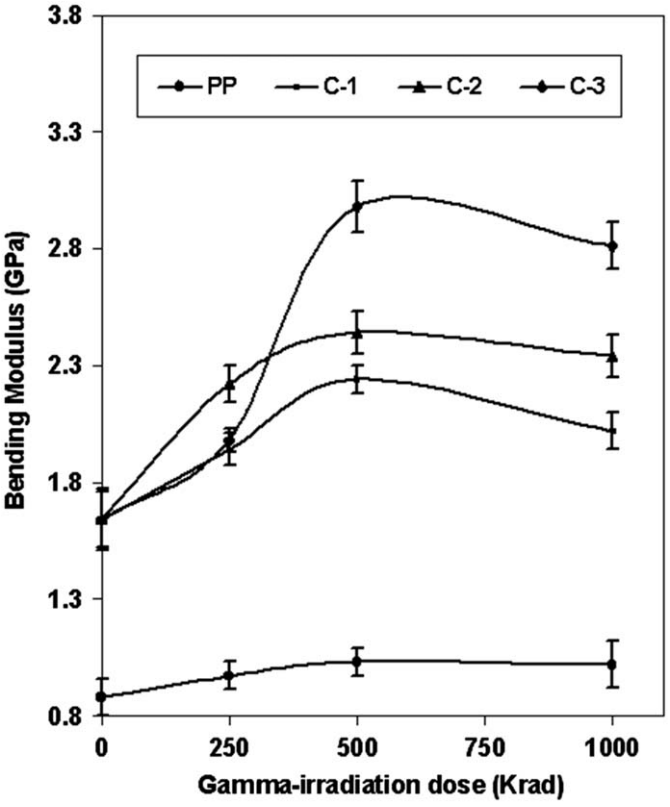


Fig. 4. Bending modulus (BM) of irradiated composite against radiation dose.

3.3. Charpy impact strength

Charpy impact strength is shown in Fig. 5. The highest impact strength values for C-1, C-2 and C-3 composites were found to be 2.76, 2.71 and 2.93 kJ/m^2 at 500 krad of total gamma dose, respectively. For C-1, C-2 and C-3 composites, about 62%, 59% and 73% increased in IS compared to untreated composites. Impact strength of the composite is influenced by the interfacial bond strength between the matrix and fiber. Gamma treatment improved interfacial bond strength by producing active sites. Irradiated jute fabrics and irradiated PP-based composites showed the better interfacial bonding.

3.4. Simulating weathering effect

Both treated (C-3) and untreated composite samples were exposed to weathering testing over a period of about 600 h of simulating sunshine and condensation at alternating cycles. In the weathering tester, high-intensity mercury or Xenon arc, water spray and humidity control were used. The loss of TS of the samples due to weathering is represented in Fig. 6. The TS loss of the untreated sample with the maximum period of observation is about 48%, whereas the treated sample lost about 32%. The weathering test demonstrated that untreated samples lost higher TS, whereas treated samples retained much of their original TS even after severe weathering test for 600 h. Among the radiation-treated samples, the treated sample with 500 krad of total dose shows more weathering resistivity that is more durable than the other treated samples.

3.5. Water uptake

Both treated (C-3) and untreated composite (C-0) samples were immersed in water at room temperature for about 30 h. The samples were taken out of water after constant time interval and their weight gain was calculated. The results of water uptake values of the treated and untreated samples are shown in Fig. 7 against the time (30 h) of soaking with respect to radiation dose. The water absorption by both treated and untreated composite samples are very quick during the initial 20 h and then water absorption decreases in the treated samples, while the untreated sample still continues to absorb water. The untreated sample attained a highest water absorption 9.85%, whereas uptake of gamma-treated composites was about 6.79% for 500 krad of total dose. Lower water uptake by the treated samples indicated that gamma radiation might have initiated a kind of grafting process of the polymer chain of jute cellulose.

3.6. Dielectric properties

Figs. 8 and 9 show the variations of dielectric constant and loss tangent with temperature for untreated (C-0) and gamma-treated composites (C-3). From the figures, it is evident that the dielectric constant and loss tangent increases sharply with increasing temperature up to transition temperature. Above these temperatures, the dielectric constant and loss tangent decreases with increasing temperature up to a certain limit and then remains almost constant. This phase transition is likely to be associated with ferro- to paraelectric transition. For these composites, the transition temperatures were observed at 80 and 65 °C, respectively. Jute is composed of cellulose whose skeletal backbone is linked by intermolecular hydrogen bonding. With increasing temperature, these bonds break up to split the chain into smaller unit of dipoles. The dipoles so formed tend to align themselves with the applied electric field and thus the dielectric constant. At the transition temperature, the formation of dipoles and alignment towards the field is maximum giving rise to maximum dielectric values, whereas above the transition

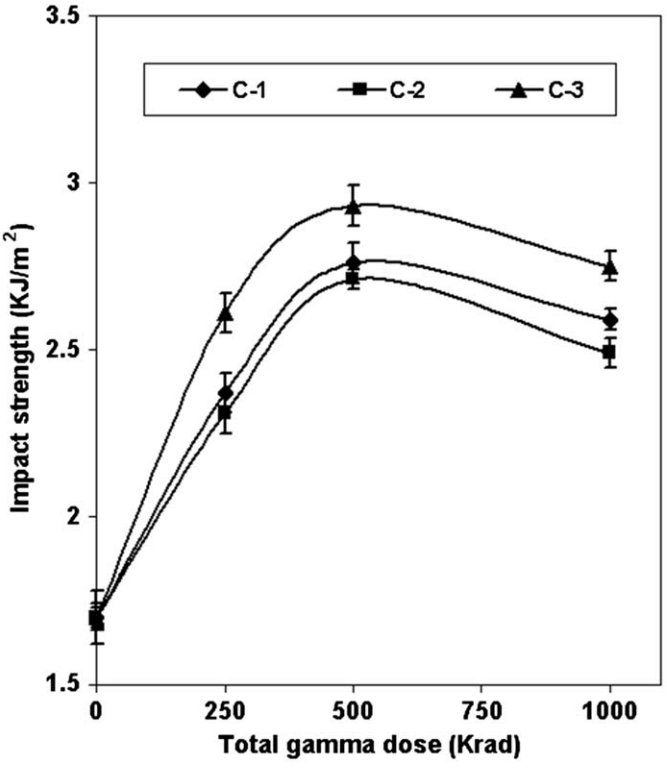


Fig. 5. Impact strength (IS) of irradiated composite against radiation dose.

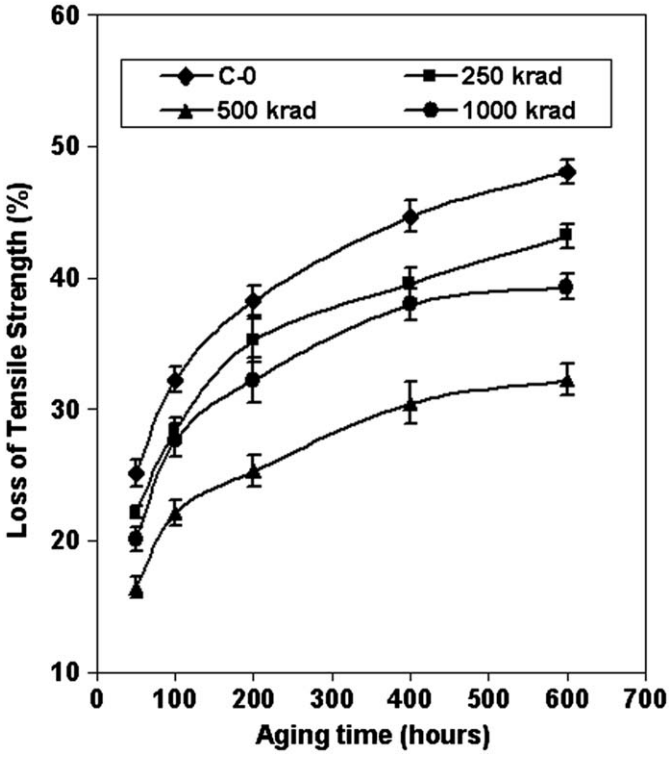


Fig. 6. Loss of tensile strength of untreated (C-0) and treated composites due to simulated weathering.

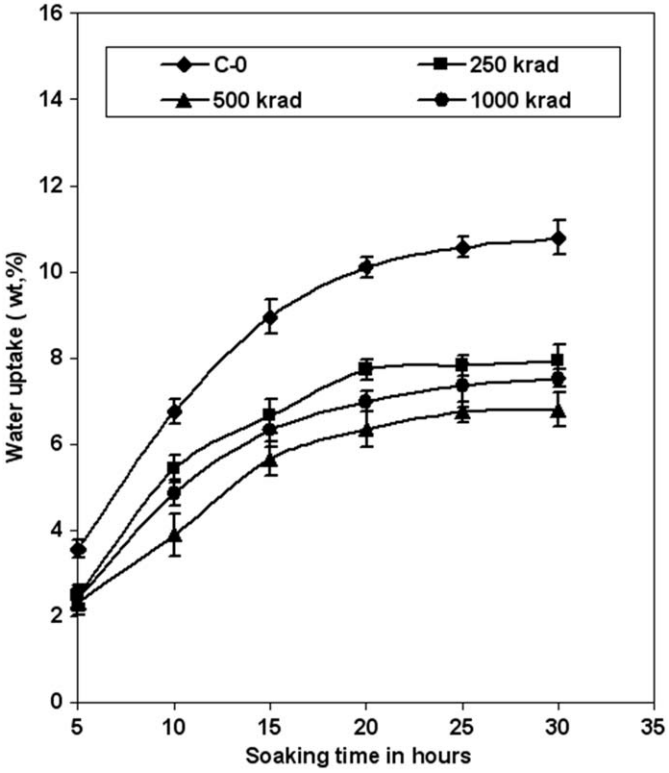


Fig. 7. Water uptakes of composites (both untreated and treated) against soaking time.

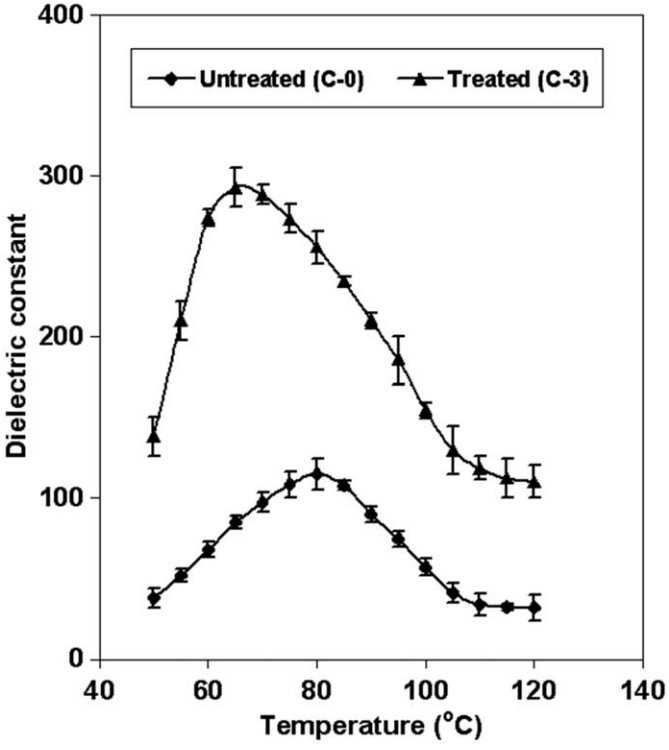


Fig. 8. Dielectric constant vs. temperature curve of untreated (C-0) and treated (C-3) composites at 500 krad of total dose at frequency 10 kHz.

temperature the dipoles tend to be oriented randomly, and hence lower the dielectric values. At an optimum temperature the dielectric values then become constant.

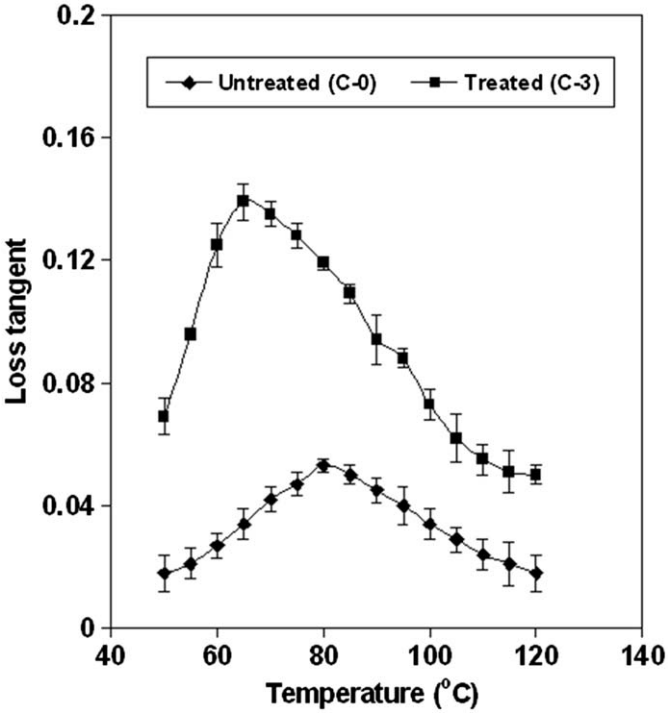


Fig. 9. Loss tangent vs. temperature curve of untreated (C-0) and treated (C-3) composites at 500 krad of total dose at frequency 10 kHz.

3.7. Scanning electron microscopic analysis

By using SEM, the surface of jute fibers (virgin and gamma treated) was examined. The SEM images of both fiber surfaces are shown in Fig. 10 where the surface topography and morphology is very clear. Untreated fiber (Fig. 10a) surface is free from blisters and seems nice looking with smooth surface. The multicellular nature of the jute fibers is clearly shown whereas rough surface with lot of fractures found on the surface of the gamma-treated sample (Fig. 10b). Thus, gamma exposure on jute results in significant change of morphology and topography of the fiber surface and this is effective for better mechanical bonding between fiber and polymer matrix. Due to better fiber matrix bonds, mechanical properties improved significantly.

4. Conclusions

Investigation showed that irradiated jute fabrics/irradiated PP-based composite produced the highest mechanical properties at 500 krad of total dose compared to the non-irradiated jute fabrics/irradiated PP and irradiated jute fabrics/non-irradiated PP-based composites. Mechanical properties such as TS, TM, BS, BM and IS of the treated composites were found higher than that of untreated composites. The dielectric constant and loss tangent of the composites is found to depend on temperature. Phase transition, likely ferro- to paraelectric transitions takes place in the composites (both treated and untreated) in the temperature range 80–65 °C. The dielectric constant and loss tangent of the composites increased as a result of irradiation. So, gamma irradiation is one of the powerful sources to improve the mechanical and dielectric properties of the hessian cloth reinforced PP composites.

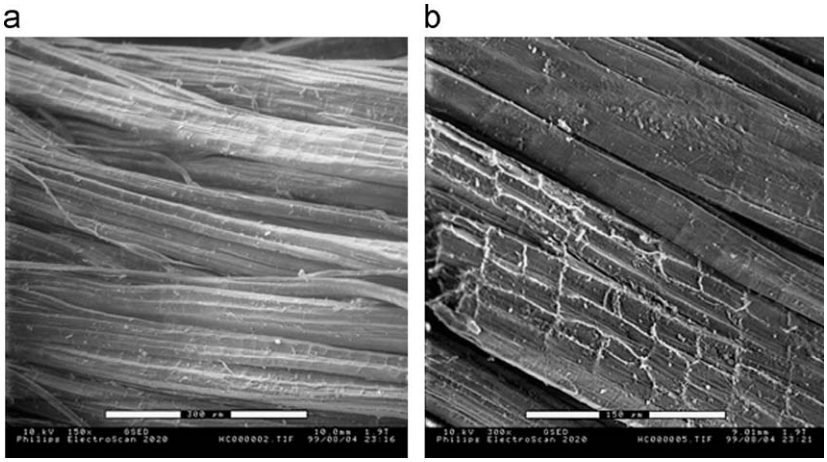


Fig. 10. SEM images of untreated (a) and 1000 krad gamma treated (b) jute fabrics.

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