Effect of Gamma-Irradiation on Properties of Polymer/Fibrous/Nanomaterials Particleboard Composites

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Summary: This investigation aimed to study the role of different contents of nano-slag, as well as various doses of gamma-irradiation on physical and mechanical properties of rice husk-polyvinyl chloride particleboard composites. Equal proportions of rice husk fibers and polyvinyl chloride polymer were used. The treatment of rice husk fibers with silane coupling agent showed a significant improvement in both mechanical and physical properties of the prepared particleboard composites as compared to those containing untreated rice husk fibers. Moreover, the partial replacement of polyvinyl chloride with different percentages of nano-slag namely 5, 10, 15, and 20% by the weight of polymer manifested a good effect on the properties of the resulting particleboard composites precisely at 10%nano-slag. In addition, the effect of different gamma-irradiation doses on the properties of the particleboard composite specimens that contain 10% nano-slag showed an enhancement in the physical (thickness swelling %) and mechanical (flexural strength, and hardness) properties. In addition, the results elaborated that the irradiated particleboard composites had a good thermal stability.

Keywords: Gamma-irradiation, Rice husk, Nanofiller, PVC, Particle board composite.

Introduction

Noteworthy, the increased demand for wood consumption as an inevitable consequence of increased population growth is leading to an increased rate of deforestation around the world which is accompanied by a negative impact on the environment. With increasing awareness of the importance of sustainable development in the worlds, many attempts are made to achieve it. Sustainable development has an effective role in rationalizing energy consumption, environmental protection, and natural conservation. Therefore, there has been a great attention to the production of particleboards from other biomasses such as grass, straw, plant, and agricultural residues. Recycling processes and utilization of these agricultural wastes in a positive manner offer one of the best ways of creating wealth from waste [1]. Particleboard is a panel fabricated from lignocellulosic fibers with an adhesive material under pressure. One of the most important and inexpensive natural fiber used in particleboard materials is the rice husk fiber which is a by-product of the rice milling process. The global production of rice husk is approximately in the range of 80 million tones which are readily available for disposal every year [2, 3]. Rice husk fibers are commonly used for energy generation or for animal food while their industrial applications are still not used.

Over the recent years, natural fiber-reinforced polymer matrixes revealed more alternatives in the materials market due to their unique advantages. Both thermoplastic and thermosets are widely used as matrix materials in the production of composite materials. Polyethylene, polypropylene, polystyrene, and polyvinyl chloride are considered important types of thermoplastic polymers used as matrix materials for composites. Some of the advantages of thermoplastic matrix composites are their flexible design, low processing costs, and ease of molding complex parts. [4].

Furthermore, it has been emphasized that the strong surface adhesion between the fiber and the polymer plays an important role in the carriage of stress from matrix to the fiber which participates in improving the performance of the composite. As a result, the chemical modification of the fibers with coupling agents can provide many benefits to the resulting particleboard composites [5-7]. Indeed, silane coupling agent is effective in modifying the natural fiber/polymer interface and exhibits an improvement in the interfacial region between fiber and polymer and consequently offers perfect bonding [8- 10].

Nowadays, fillers have received a great interest for application in the field of polymer particleboard composites due to their reducing cost, improving processing, density control, electrical properties, and thermal conductivity [11]. Nanofillers

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lists increased within years (nano-oxides, nanocaly, and carbon nanotubes) and give some unique features to polymer composites. The large surface area of nanofillers promotes better interfacial interactions with the polymer, leading to a good property enhancement [12-16]. In polymer composites, different types of nanofillers are used for improving the mechanical and thermal properties. Among them is ground granulated blast-furnace slag (GGBS) which represents useful filler in the fabrication of plastics because of its superior dispersibility, heat resistance, and stability. It is also used as good filler for making polymeric natural fiber composites to improve its mechanical properties such as impact properties, abrasion resistance, and hardness [17].

Radiation processing has been demonstrated on a large commercial scale to be a very effective method to improve end-use properties of various polymers. It is a well-established and economical method of precisely modifying the properties of bulk polymer resins and formed polymer components. The radiation technology is preferred over the other processes due to many advantages when compared with other conventional methods. In radiation processing, no catalyst or additives are required to initiate the reaction. Also, it is a continuous operation, having a minimum time requirement, less atmospheric pollution, curing at ambient temperatures, increased design flexibility through process control, etc. In short, radiation plays an important role in the polymer processing that is used in the composite field [18, 19].

In the present study, the influence of different gamma-irradiation doses as well as the effect of chemical treatment of rice husk fibers with silane coupling agent on the physical, mechanical and thermal properties of rice husk-polyvinyl chloride particleboard composites that containing different percentages of nano-slag is studied.

Experimental

Materials

The thermoplastic polymer used in this study was polyvinyl chloride (PVC) supplied by Cedasa Company Egypt. It had a chemical formula $(C_2H_3Cl)_n$, and its density was 1.3-1.45g/cm^3. Rice husk (RH) fibers were firstly washed with tap water to remove residues and dried at 75 ºC for 24 h. Then they were sieved to obtain particle size ranged between 0.5 mm and 0.8 mm. Ground-granulated blast-furnace slag was obtained by quenching of molten iron slag (a by-product of iron and steel-making) supplied by iron and steel company Helwan, Egypt. It had a particle size of 68 nm measured by a Transmission electron microscopy (TEM) as shown in Fig.1. It was abbreviated as NS, and its chemical oxide composition was given in Table-1. While coupling agent 3-aminopropyltrimethoxysilane with a chemical formula $H_2N(CH_2)_3Si(OCH_3)_3$ supplied by Sigma-Aldrich Company was used for the chemical treatment of rice husk fibers. It was a colorless liquid, 97% concentration, and its density was 1.03 g/cm^3.

Table-1: Chemical oxide composition of nano-slag (NS).

Filler type	Chemical oxide composition (%)								
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	TiO_2	K_2O	MnO
Nano-slag	34.5	6.4	1.0	45.2	3.3	2.4	1.3	-	5.8

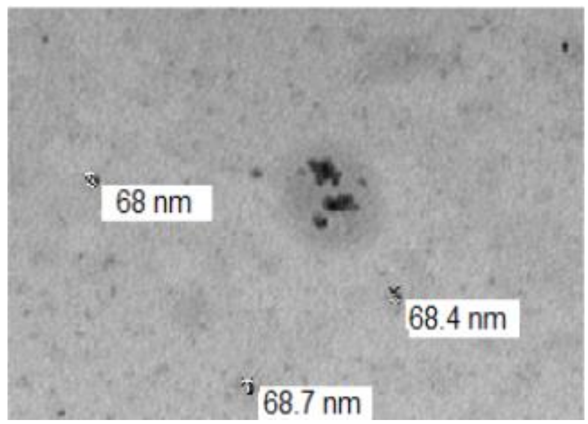


Fig. 1: Transmission electron microscopy of nano-slag.

Treatment of RH fibers with silane coupling agent

Rice husk (RH) fibers were treated with silane coupling agent, 3-aminopropyltrimethoxysilane delivered in a liquid form; the applied amount of coupling agent was 2% by the weight of rice husk (RH) fibers. Before application; it was diluted with a mixture of methanol/distilled water (80/20) and acetic acid was used to adjust pH value at 4. The fibers were soaked in this solution for 2 h. and then dried in a drying oven at 70 °C.

Preparation of particleboard composites

Firstly, both untreated and treated rice husk fibers (RH) were mixed with polyvinyl chloride (PVC) to prepare particleboard composites 50%RH:50%PVC. Then a partial substitution of polyvinyl chloride with different ratios of nano-slag (NS) namely, (5%, 10%, 15%, and 20%) by the weight of the polymer was carried out. Next, all mixtures were pressed into a mould of dimensions 16 x 16 x 0.8 cm^3, using an electric hot press type Carver-M-154; hot pressing was performed at 120 ºC (Pre-heating for 5 minutes at 20000 PSI was carried out). The resulting specimens were cooled at the same pressure for another 5 minutes, and the prepared particleboard composites (PBNSC) were shown in Table-2. Finally, different gamma-irradiation doses were applied for the particleboard composites' specimens containing 10%NS (PBNSC-10) ranged from 5 - 30 kGy. The irradiation process was carried out in National Center for Radiation Research and Technology using the cobalt-60 source Cell type 4000 A at a dose rate of 5.9 kGy/h supplied by Bhabha Atomic Centre, Mombey, India.

Table-2: Mix proportions of untreated and treated particleboard composites.

Nano-slag %	Mix composition (w/w)%	Symbol code
0	50%RH:50%PVC	
5	50%RH: (45%PVC+5%NS)	
10	50%RH: (40%PVC+10%NS)	PBNSC
15	50%RH: (35%PVC+15%NS)	
20	50%RH: (30%PVC+20%NS)	
10	50%RH: (40%PVC+10%NS)	PBNSC-10

Physico-mechanical measurements

Flexural strength

Three individual rectangular shaped samples of dimensions (15 cm in length, 2.5 cm in width, and 0.8 cm in thickness) were subjected to testing and the arithmetic of the result values had been recorded using flexural machine (ADR-Auto 25KN) supplied from ELE international (UK). The flexural strength properties were determined according to ASTM standard (D1037-11, 1987) [20] as in the following equation.

$$Flexural\ strength = \frac{3FL}{2bd^2} \quad (1)$$

where;
b: the width of the specimen in cm,
d: thickness of specimen in cm,
L: length of the specimen in cm, and
F: maximum load (kg/cm^2)

Hardness

The hardness test was carried out by using shore durometer, supplied by Baxlo instruments De Medida Precision S.L., Percelona, Spain. Three rectangular shaped specimens were subjected to static loading having dimensions of (15cm in length, 2.5 cm in width, and 0.8 cm in thickness) according to (ASTM D 2240, 2000).

Thickness swelling (%)

This test was done according to ASTM standards D1037 specification [21]. The thickness was measured at four points midway along each side 2.5 cm. After 24 hours of submersion in distilled water, the thickness was measured at the same four points and the average was obtained. The following calculation could then be made:

$$Thickness\ swelling\ (\%) = \left(\frac{T_1 - T_2}{T_2}\right) x\, 100 \quad (2)$$

where,
T_1 = wet thickness
T_2 = initial thickness

TEM, SEM, XRD and TGA Techniques

TEM analysis revealed the particle size of nano-slag (NS), cryogenically microtome sections were observed using a TEM (JEOL JSM-100CX, Shimadzu. Co., (Japan) with an acceleration voltage of 80 kV. The morphology of the fractured surface of some selected specimens was investigated by scanning electron microscopy (SEM). The SEM micrographs were taken with a JSM.5400 (JEOL/Japan). XRD analysis was used to identify the changes in the crystalline. The samples after being dried, a freshly fractured surface of the samples are mounted on a stub of metal with adhesive, coated with 40–60 nm of Gold. The XRD were recorded on Shimadzu Corporation X-ray diffractometer, coupled with automatic data recording DP-DI system (ver.1.1) with a wavelength of 0.15425 nm. (Tyoto, Japan) and Ni-filtered Cu-Kα radiation was used during scanning, and patterns were recorded at a scanning speed of 8°/min. And the thermal behavior of the specimens was determined by using thermogravimetric analyzer (TGA) model Schimadzu TGA-50 (Kyoto, Japan); by raising the applied temperature from 25°C to 900°C with a flow rate of nitrogen gas 20ml/min and a heating rate of 10ºC/min.

Results and Discussion

Flexural strength

The influence of nano-slag content on the flexural strength values of particleboard composites; containing untreated and treated rice husk fibers is given in Fig. 2. It is clear that the flexural strength values of both untreated and treated particleboard composites (PBNSC) increase with increasing nano-slag content up to a certain value (10% of nano-slag) and then decrease. The partial substitution of polyvinyl chloride (PVC) with 10% nano-slag (NS) results in the highest flexural strength values for both untreated and treated particleboard composites. This is due to the fact that with a lower loading of nano-slag up to 10%, the potential of the formation of micro-voids is less, and the dispersion of nano-slag particles in the polymer matrix is more uniform leading to an enhancement in the strength [22]. With the increase of nano-slag content above 10% a marked decrease in flexural strength is clearly observed due to the poor dispersion of the nano-slag particles inside the polymer forming platelet agglomerations which act as stress concentrators.

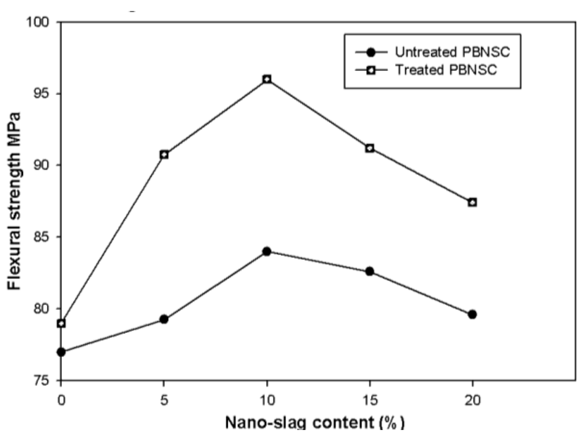
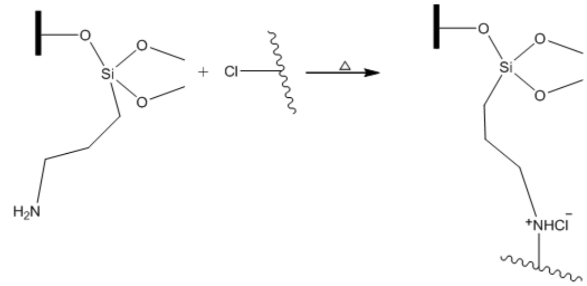


Fig. 2: Effect of nano-slag content on the flexural strength values of both untreated and treated particleboard composites (PBNSC).

As expected, the chemical treatment of rice husk fibers with 3-aminopropyltrimethoxysilane exhibited better flexural strength values for the particleboard composites (PBNSC) as compared to the untreated ones at any nano-slag content. This improvement has been proposed to be due to the strong affinity of the amino group towards the hydroxyl groups of rice husk fibers and the formation of a cage-like interpenetrating polymer network (IPN) composed of the polysiloxane structures [23, 24]. The IPN network can entrap the polyvinyl chloride molecules thereby anchoring the thermoplastic matrix to the treated fiber surface. In addition, acid-base interactions may also play a role in improving interfacial adhesion of composites composed of the amino silane-treated rice husk fibers and polyvinyl chloride matrix by forming a bridge of chemical bonds between fiber and polymer and as a result, the flexural strength is improved (scheme-1).



Scheme-1: Interaction between treated rice husk fiber and polyvinyl chloride.

Radiation chemistry implies the chemical effects of ionizing radiation (high energy) with materials. The ionizing radiation penetrating the matter causes an excitation and ionization of the molecules in the medium. When gamma ray interacts with a polymeric material, its energy is absorbed by the polymer and active species such as radicals are produced, thereby initiating various chemical reactions. The relation between flexural strength of the untreated and treated particleboard composites containing 10% nano-slag (PBNSC-10) as a function of gamma-irradiation doses is shown in Fig. 3. In general, both polymer cross-linking and degradation by chain scission occur during irradiation process, but one of these effects may be predominant. The obtained experimental data indicates that the development in the flexural strength of irradiated particleboard composite at a lower absorbed dose is essentially related to the cross-linking process during the gamma irradiation of the samples, which leads to a dense structure of the specimens. Beyond a dose of 5kGy, a chain scission process may occur which accompanied with a decrease in the flexural strength values.

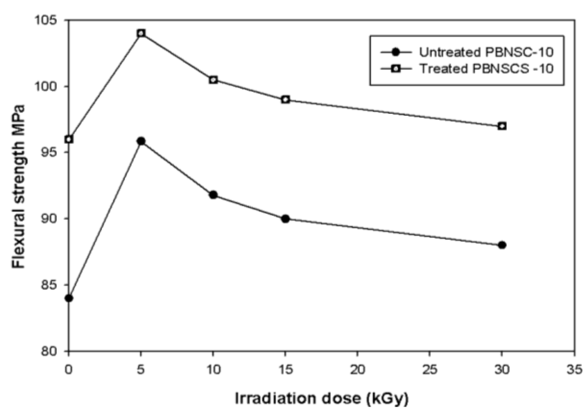


Fig. 3: Effect of gamma-irradiation doses on flexural strength values of both untreated and treated particleboard composites containing 10% nano-slag (PBNSC-10).

Hardness

The effect of different percentages of nano-slag content on the hardness values of both untreated and treated particleboard composites (PBNSC) is graphically represented in Fig.4. It is observed from the Fig that the hardness values of all particleboard composites increase with increasing nano-slag content up to 10% and then started to decrease beyond this percentage. Also, at any nano-slag content the hardness values are higher for particleboard composites containing treated rice husk fibers than those containing untreated fibers. This behavior can be explained on the basis of the treatment of rice husk fibers with amino silane improved the interfacial adhesion between the polymer and rice husk fiber as mentioned earlier, leading to less micro-voids and filler-polymer debonding in the interphase region. Moreover, the enhancement in the hardness values of the particleboard composites with the incorporation of small amounts of nano-slag (up to10%) may be attributed to the fact that the addition of nano-slag will restrict the chain mobility of the polymer and consequently increases the stiffness properties of the composites. The incorporation of nano-slag improved the interfacial interaction between the polymer and natural fibers in the composites, resulting stiffer and tougher particleboard composites leading to higher hardness values. However, after this optimal limit of nano-slag the hardness values are dropped with the continuous increase of the nano-slag content (15 and 20%). This may be due to the size of the clusters reaching a crucial limit and therefore the reinforcing function of the nano-slag decreases [25].

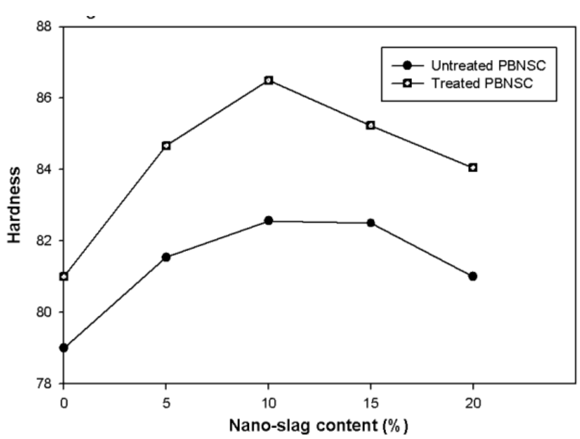


Fig. 4: Effect of nano-slag content on the hardness values of both untreated and treated particleboard composites (PBNSC).

On the other hand, the effect of different doses of gamma-irradiation on hardness values of particleboard composites containing 10% nano-slag (PBNSC-10) is graphically represented in Fig. 5. It is clearly observed that, the hardness values of gamma-irradiated composites show an improvement with a dose of 5kGy due to the formation of free radicals under the effect of ionizing radiation and crosslinking process takes place with the formation of three dimensional net-work structure which improves the mechanical properties.

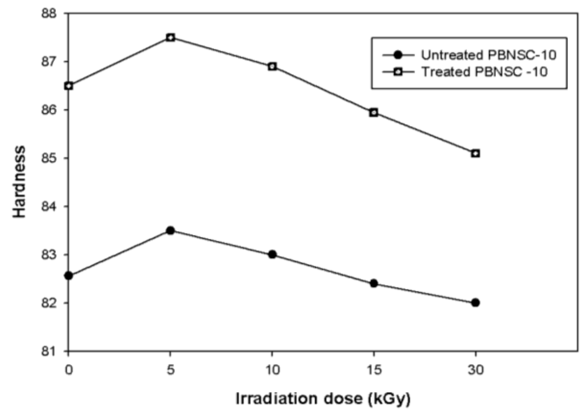


Fig. 5: Effect of gamma-irradiation doses on the harness values of both untreated and treated particleboard composites containing 10% nano-slag (PBNSC-10).

Thickness swelling percentage

Thickness swelling percentages of both untreated and treated particleboard composites are plotted against nano-slag content as shown in Fig. 6. It is clearly seen that, thickness swelling values decrease with the increase in nano-slag loading up to 10% which may be explained on the basis of the barrier properties of nano-slag filler which inhibit the water permeation into the particleboard specimens. This barrier property hinders water from going into the inner part of the particleboard composites. At higher nano-slag percentages (15% and 20%) the thickness swelling values increase probably due to the imperfect dispersion of nano-slag particles in the polymer. The results also indicate that the thickness swelling (%) values of the composites containing aminosilane treated rice husk fibers are lower than those containing untreated fibers at the same nano-slag content. This may be due to the fact that thickness swelling percentage is affected by the strength properties of the composites and the degree of water absorption; i.e, the composites with high strength properties and low water absorption percentages exhibit the lowest thickness swelling values [26- 29].

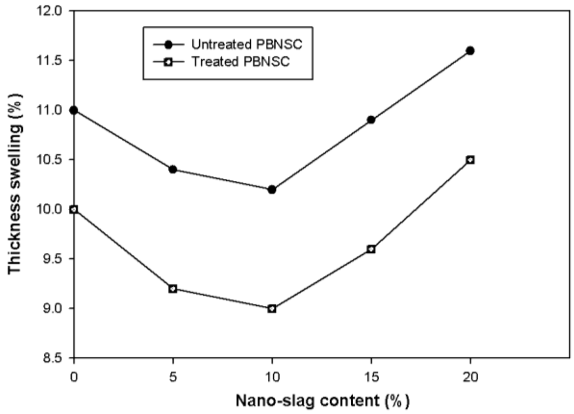


Fig. 6: Effect of nano-slag content on the thickness swelling percentage of both untreated and treated particleboard composites (PBNSC).

Fig 7 illustrates the variation of the thickness swelling (%) of both untreated and treated particleboard composites containing 10%nano-slag (PBNSC-10) with different gamma-irradiation doses. It is observed that, gamma-irradiated composites show a reduction in the thickness swelling values at a dose of 5 kGy due to crosslinking and formation of a net-work structure as mentioned above.

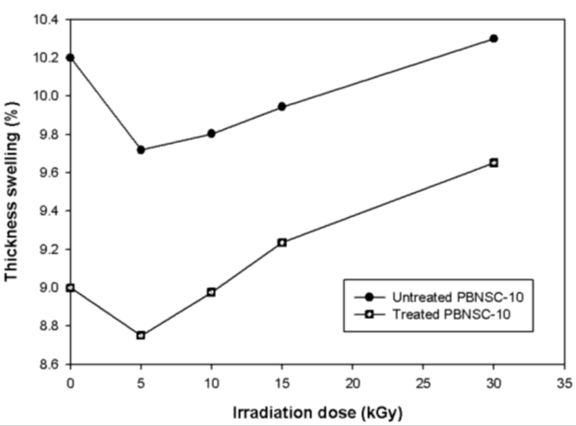


Fig. 7: Effect of gamma-irradiation doses on the thickness swelling percentage of both untreated and treated particleboard composites containing 10% nano-slag (PBNSC-10).

Thermogravimetric Analysis (TGA)

A thermogravimetric analysis study was undertaken in order to obtain information about the thermal decomposition behavior of untreated and treated particleboard composites containing 10% nano-slag filler (PBNSC-10) before and after irradiation with a dose of 5 kGy of gamma rays as shown in Fig. 8. The thermal decomposition of these composites was significantly related to thermal stability of its constituents materials. TGA results of particleboard composites (PBNSC-10) showed a slight loss of weight at a temperature range 100-250ºC due to the elimination of physically absorbed water in the RH, superficial or external water bounded by surface tension, and loss of light volatiles. While in a temperature range of 250-360°C a majority of weight loss took place due to dehydrochlorination of PVC and evolution of HCl. A further weight loss between 360-630 °C was observed due to the evolution of the volatile compounds generated during decomposition of primary hemicellulose and cellulose of rice husk fibers, lignin conversion [30], cracking and decomposition of the dehydrochlorinated polyvinyl chloride. Finally the particleboard composites left residues approximately 13.9, and 21.6 for untreated particleboard composites specimens at 0kGy and 5kGy respectively. While, the remaining weight percentages of treated specimens at 0kGy and 5kGy were about 19.8, and 22.6, i.e., the thermal stability of the treated particleboard composite was improved after silane treatment [31].

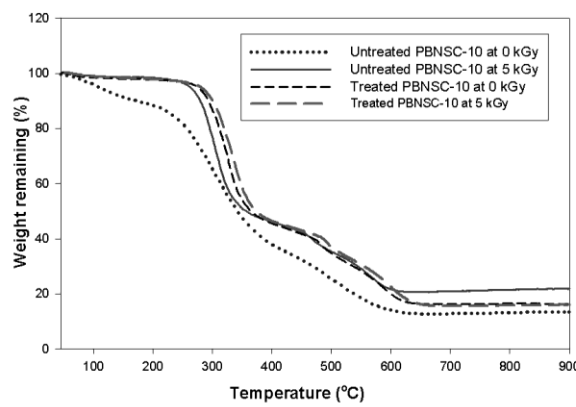


Fig. 8: TGA thermograms of both untreated and treated particleboard composites containing 10% nano-slag (PBNSC-10) at 0 and 5 kGy of gamma rays.

Fig. 9 illustrated the maximum rate of thermal decomposition reaction (dw/dt) of all particleboard composites' specimens (PBNSC-10) as a function of temperature. This Fig showed that, the first maximum rate of thermal decomposition reaction was located at 291, and 293°C for untreated particleboard composites at 0kGy and 5kGy respectively. While in the case of treated particleboard composite specimens the first maximum rate of thermal decomposition reaction was located at 301 and 303 °C before and after irradiation. The results also indicated that, the second rate of maximum thermal decomposition values was lied around 490°C and 540°C for the untreated at 0kGy and 5kGy respectively. While its values for the treated specimens were 545 and 565 °C before and after irradiation. Based on the above it is clear that the treated particleboard composite containing 10% nano-slag and irradiated at a dose of 5kGy is thermally more stable as compared to the other particleboard composites due to the crosslinking and the formation of a net-work structure during the irradiation process.

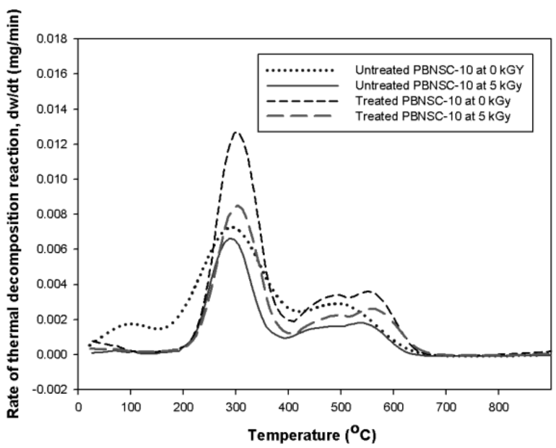


Fig. 9: Rate of thermal decomposition reaction (dw/dt) of both untreated and treated particleboard composites containing 10% nano-slag (PBNSC-10) at 0 and 5 kGy of gamma rays.

XRD analysis

The XRD diffraction patterns of rice husk fiber (RH) and polyvinyl chloride (PVC) were shown in Fig. 10. From this Fig it was observed that there was a semi-crystalline peak situated at 2θ=21.7° (d-spacing 4.0 Å) which corresponded to the major structure of cellulose in rice husk (RH). Also the spectrum of polyvinyl chloride (PVC) showed a very low broad peak appeared around 2θ=23.4° [32]. Fig. 11 represented the XRD diffraction patterns of both untreated and treated particleboard composites containing 10% nano-slag (PBNSC-10) before and after irradiation with a dose of 5kGy. Generally, it was clearly observed that the characteristic peak of the treated particleboard composite irradiated at a dose of 5kGy was located at 2θ=19.94° and it was more intense than those of the other composites (2θ=21.34°). This meant that the chemical treatment of the particleboard composite with silane removed part of the amorphous material covering the rice husk fibers and thus exposing the cellulose. Along with the effect of gamma-irradiation which exhibited an improvement in the intensity of the peaks of the irradiated specimens due to the cross-linking process.

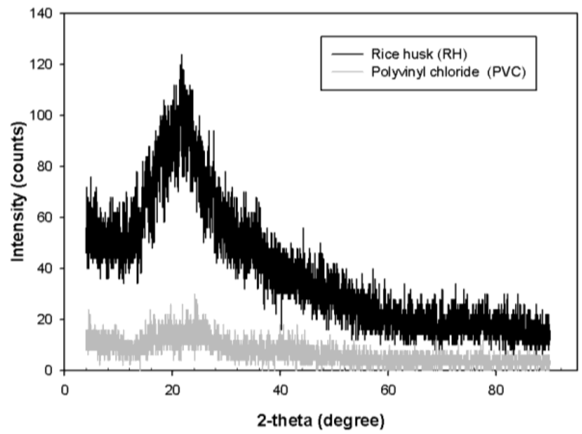


Fig. 10: XRD diffraction of both rice husk fiber (RH) and polyvinyl chloride (PVC).

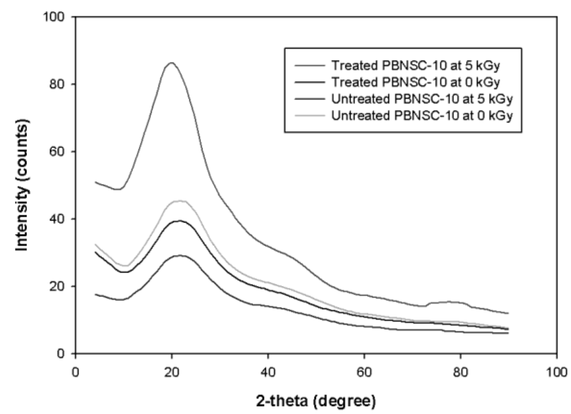


Fig. 11: XRD diffraction of untreated and treated particleboard composites containing 10% nano-slag (PBNSC-10) at 0 kGy and 5 kGy.

Scanning Electron Microscope (SEM)

Fig. 12 (a and b) showed SEM analysis of the fractured surfaces of the untreated and treated particleboard composites with the incorporation of 10% nano-slag before they had irradiated with gamma-irradiation (0kGy). The microstructure of the untreated PBNSC-10 displayed poor adhesion between fiber and polymer matrix which facilitated fiber pull-out and as a result voids and cavities were showed in the micrograph. While the particleboard composites made with treated fibers showed an improvement in polymer/fiber adhesion, avoiding fiber pull-out and the gap between fiber surface and polymer was reduced. This indicated that the fiber treatment contributed to the improvement in surface adhesion between fibers and polymers. Moreover, enhanced properties were observed for the treated particleboard composite irradiated at 5kGy (Fig.12c) depending on the cross-linking between the constituents of the particleboard composite and a denser structure with fewer voids or cavities was distinguished.

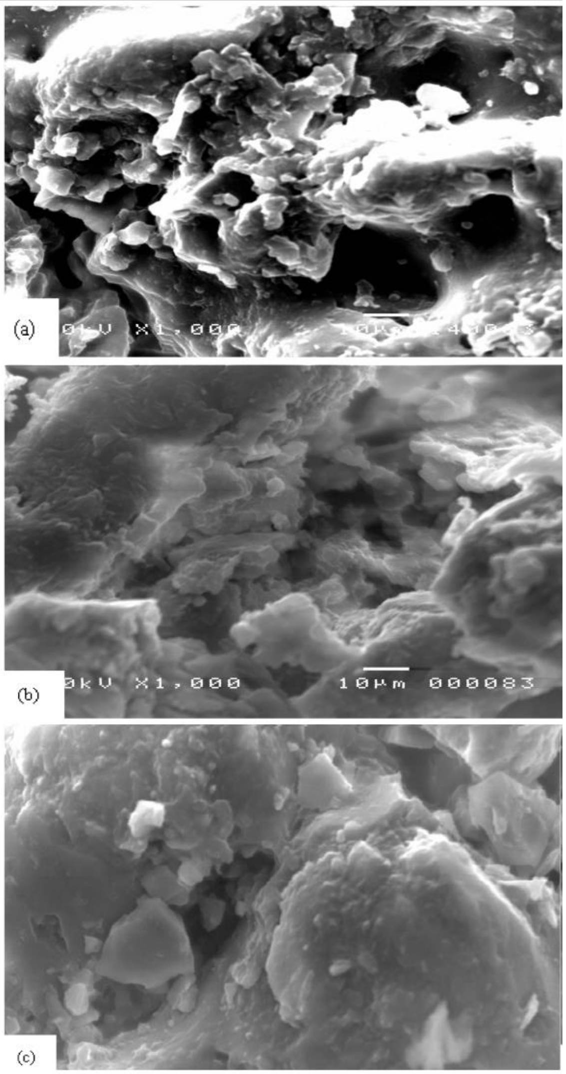


Fig. 12: SEM micrographs for (a) untreated particleboard composite (PBNSC-10) at 0 kGy, (b) treated particleboard composites (PBNSC-10) at 0 kGy (c) treated particleboard composites (PBNSC-10) at a dose of 5 kGy.

Conclusion

From the obtained results in the present investigation, the following conclusions may be deduced:

The particleboard composites treated with silane coupling agent significantly improved the interaction between rice husk fibers and polymer matrix and exhibited better physical and mechanical properties than that of the untreated particleboard composite.

Nano-slag used as filler up to 10% by the weight of the polymer of the particleboard composited significantly affected the physical and mechanical properties.

Mechanical properties such as flexural strength, and hardness improved and the resulting particleboard composites become more rigid with the increase of nano-slag content up to 10% NS in both untreated and treated particleboard composites.

Thickness swelling percentage showed an affective improvement for the particleboard composite treated with silane coupling agent and loaded with 10% nano-slag as compared to the untreated ones.

Gamma-irradiation process enhanced the physical and mechanical properties of both treated and untreated particleboard composites (PBNSC-10) at 5kGy and then the properties of the particleboard composites were decreased.

Thermally, the treated particleboard composite (PBNSC-10) that irradiated at a dose of 5 kGy showed a better thermal stability than the un-irradiated one. Whereas, the remaining weight percentages of the treated specimens at 0kGy and 5kGy were about 19.8, and 22.6 respectively.

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