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Influence of Ionizing Radiation on the Mechanical Properties of a Wood-Plastic Composite

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Abstract

The focus of this study was to examine the potential benefits of irradiating polyethylene (PE)-based wood-plastic composites (WPCs) in order to enhance the mechanical properties of the WPC. The PE-based WPCs were irradiated, post extrusion, at dose levels of 0, 50, 100, 150, 200, and 250 kGy with an electron beam (EB). The irradiated WPCs were then evaluated using a third point bending test (ASTM D4761) along with scanning electron microscopy (SEM). It was found that ultimate strength and modulus of elasticity (MOE) increased with increasing dose level. Examination of the fracture surfaces of polyethylene revealed a distinct difference in failure between irradiated and non-irradiated surfaces.

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Keywords: Electron Beam; Radiation Crosslinked Polyethylene; Wood Plastic Composite

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1. Introduction

Wood-plastic composites (WPCs) are a mixture of wood in a polymer matrix. The wood can be of any form (Clemons 2002) and the polymer matrices can either be thermoplastic, which can be thermally altered after processing, or thermoset, which cannot be altered after processing. Thermosets used in thermoset polymer matrices generally consist of polyesters, epoxies, and phenols (George et al. 2001). Thermoplastics used in WPC polymer matrices include polyethylenes (PE), polystyrenes (PS), and polypropylenes (PP) (George et al. 2001). Of these matrices, PE-based WPCs have the highest volume of use in the manufacture of building materials, such as decking and deck railings (Clemons 2002). PP-based WPCs are commonly used in automotive products (Clemons 2002).

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Development of materials used in construction often focuses on increasing the materials' strength-to-weight ratio. When less material is used, consumer and transportation costs will decrease and the value of the product will increase. This is due to the wider range of applications a stronger material has. In order to increase a strength-to-weight ratio, strength must be increased and/or weight must be decreased. WPCs have a low strength-to-weight ratio (Wolcott and Smith 2004) because of the polymer matrix. Decreasing the amount of polymer used in the matrix material would directly decrease the cost of the material. The specific gravity of WPCs used in decking is approximately twice that of solid lumber (FPL 2010). Hollow or shaped cross-sections have been used to decrease the weight of PE-based WPCs (Wolcott and Smith 2004), however, using less material does not increase the strength of the material. Wolcott and Smith (2004) stated that although reductions in weight are needed, they must be done in a manner that does not reduce mechanical properties like toughness, creep, and strength.

A large amount of research has been put into increasing the strength of PE-based WPCs. Past research has focused on the use of coupling agents, and more recently nano-reinforcements, to increase mechanical properties of PE-based WPCs (Faruk and Matuana 2008, Lu et al. 2005). While both methods have presented successful results in the enhancement of mechanical properties, the investigation into finding a cost effective, high performance method of enhancement is ongoing (Cai et al. 2013). Another method that has gained attention in recent literature is to increase the strength and stiffness of the WPC by crosslinking the matrix material, thereby creating a thermoset WPC (Bengtsson et al. 2006, Kuan et al. 2006, Janigova et al. 2001, Reyes et al. 2001). Reinforcement of PE by forming crosslinks using peroxide-initiated or vinyl silane-grafting crosslinking methods has improved mechanical properties of WPCs (Bengtsson et al. 2006, Kuan et al. 2006, Janigova et al. 2001). Reyes et al. (2001) irradiated blends of recycled PP/PE-based WPCs with a cobalt 60 gamma radiation source. It was found that the crosslinking reaction of the PE outweighed any scission reactions of PP upon irradiation, and mechanical properties were increased.

Radiation crosslinking of PE has been researched since the 1950s (Charlesby 1952, Dole et al. 1954). Polyethylene crosslinking occurs after the cleavage of carbon-hydrogen (C-H) bonds by accelerated electrons, forming free radicals. The atoms along the molecular chain combine with the unpaired electrons from the free radicals to form the crosslinked, three-dimensional networks. The resulting polyethylene thermoset is stiffer and harder than the thermoplastic (Bharat 2000). Electron beam (EB) processing of PE has carved out established markets in wire jacketing, hot water tubing, closed cell foams, and joint replacement (Atkinson and Cicek 1983, Berejka and Cleland 2011, Manning et al. 2005). The idea that an EB can be used as a tool to alter an existing commercial product, such as the composite decking in this study, was a result of these pre-existing crosslinked PE markets.

When cellulose, the primary structural component of wood, is exposed to EB radiation, the dominant reaction is chain scissioning (Berejka and Cleland 2011, Mclaren 1978, Smith and Mixer 1959). Therefore, the resulting degradation of the wood fibers should occur, to some extent, in a radiation-crosslinked wood thermoplastic composite. High amounts of wood fiber degradation could impact the overall strength of the material. This paper presents the results of a comparative evaluation of a PE-based WPC subjected to different doses of radiation. Results are presented in terms of the flexural strength properties to evaluate the impacts of EB on the PE-based WPCs.

2. Materials

PE-based WPCs were purchased from a local supply store in 3.6 m (12') lengths, cut into 0.9 m (3') sections. The products purchased were deck boards, rated for a 0.4 m (16") on center span at 4.8 kPa (100 PSF). The boards were 14 cm (5.5") wide, 2.54 cm (1") thick with an average density of 0.98 g/cm. The deck boards were an extruded composite blend of PE (40-50%), wood fiber (50-60%), and less than 1% carbon black by weight. The PE was mostly linear low density polyethylene (LLDPE), derived from recycled grocery bags and stretch film, as listed in Table 1. The wood fibers were waste material from furniture makers or recycled pallets.

Table 1. Composite Information

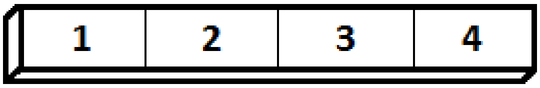
WPC Constituent	Quantity (wt.)	Source
PE	40-50%	Recycled/Reclaimed Grocery Bags and Stretch Film
Wood Fiber	50-60%	Furniture Manufacturers and/or Waste Pallets
Carbon Black	<1%	

3. Methods

3.1. Electron Beam

The 2.54 X 13.97 X 91.44 cm (1 X 5.5 X 36”) PE-based WPC specimens (56 total) were irradiated with a Dynamitron 3.0 MeV, 90 kilowatt EB accelerator at IBA Industrial, located in Edgewood, NY. Nine replicates were treated at five different dose levels: 50, 100, 150, 200, 250 kGy; and compared against a control (0 kGy). The samples were irradiated in air and to assure the most uniform dose distribution, irradiated from both the front and back of each sample. A study by Gheysari and Behjat (2001) used the same doses, with a 5 MeV EB, and found declining tensile strength after 200 kGy. Therefore, 0-250 kGy seemed to be a reasonable range for absorbed dose level. The process sampling matrix represented in Table 2 explains the designation of dose by board section. The doses were assigned in this way to assure that each dose had sample specimens from both inner and outer sections of the board. This was done in case the boards were stressed in the middle section due to improper handling. The total dose to each board was applied in fractions of 50 kGy in order to avoid rapid temperature increases.

Table 2. Dose Designation by Board Section for Each Dose



(Board by Section)

0 kGy	50 kGy	100 kGy	150 kGy	200 kGy	250 kGy
1	2	3	4	1	2
3	4	1	2	3	4
1	2	3	4	1	2
3	4	1	2	3	4
1	2	3	4	1	2
3	4	1	2	3	4
1	2	3	4	1	2
3	4	1	2	3	4
1	2	3	4	1	2

3.2. Storage and Moisture

Once purchased, the specimens were transported and stored in a conditioning chamber at 21^{0}C (70^{0}F) and 50% relative humidity (RH) at SUNY-ESF in Syracuse, NY. The specimens were weighed before and after storage, which resulted in a weight change of less than 1%. Wafers 140 X 25 X 12.5 mm (5.5 X 1 X 0.5”) in size were cut from an inside section of each of the original 0.9 m (12’) deck boards. The wafers were placed in an oven at 103^{0}C (217^{0}F), until weight loss discontinued, then placed in a desiccator to cool, before a final mass measurement.

Conditioning and moisture content parameters were in accordance with ASTM 4442. The moisture content of the specimens were estimated to range from 1.0-1.3%, consistent with values published by the manufacturer.

3.3. Flexural Test

Third-point (4 point) bending tests were conducted using a Tinius Olsen 54,431 kg (120,000 lb) Electomatic Universal Testing Machine, in accordance with ASTM D4761. A 2,268 kg (5,000 lb) load cell was used as well as a constant 1.7 cm/min (0.75”/min) crosshead speed to failure. A linear variable differential transformer (LVDT) conditioner was in place to detect deflection. The modulus of elasticity MOE (also call flexural modulus) was derived from the most linear region of the stress-strain curve, estimated to be between 345 and 690 KPa (500 and 800 PSI), based on the high R-value of the trendline.

3.4. Scanning Electron Microscopy

Images of the fractured surfaces of the PE-based WPC specimens were taken using a JEOL JSM-5800 LV scanning electron microscope (SEM). Secondary electrons were detected and a 2.0 kV accelerating voltage was used. The images were taken to monitor the fracture surface of the PE and wood fibers after EB treatment. The samples were sputter-coated in a 30 nm layer of platinum before micrograph analysis.

4. Results

4.1. Mechanical Properties

Figure 1 is a typical stress strain curve for the WPC in a flexural test. The MOE was determined using the straightest part of the curve between 2 and 9 MPa. The relationship between dose and average ultimate strength, in bending, for the PE-based WPC specimens tested is shown in Figure 2. Each point shown in Figure 2 is an average of ultimate strength tests. The error bars indicate +/-1.0 standard deviation from the mean. An overall increase of 10% in average ultimate strength resulted from the 250 kGy EB dose compared to the 0 kGy control. A plot of the average values for the doses tested indicates a direct linear relationship as dose increases.

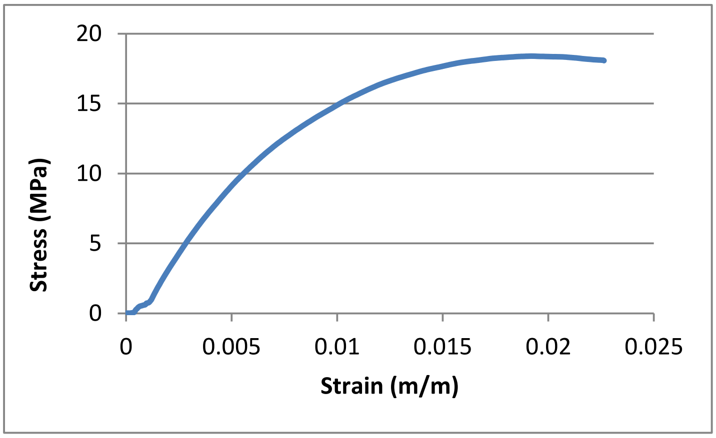


Figure 1. Typical Stress Strain Curve for a WPC

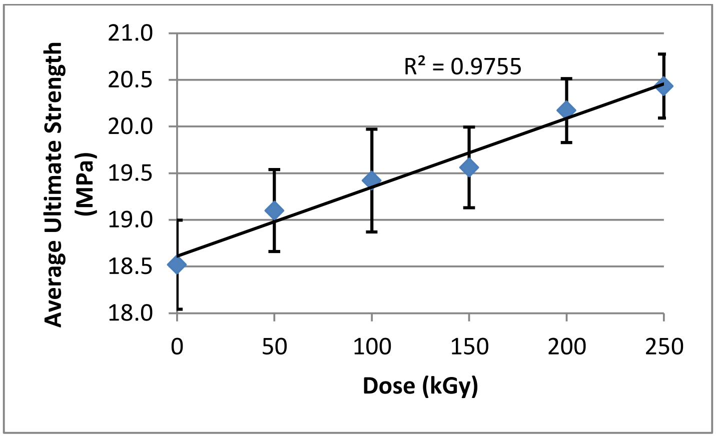


Figure 2. EB Dose vs. Average Ultimate Strength

A simple linear regression analysis of the trendline for the dose versus average ultimate strength relationship is shown in Table 3. Both the R^2 and adjusted R^2 values demonstrate a predictive power greater than 96%. The overall significance of the simple linear regression analysis is greater than 99%, which is equal to the reliability of the ultimate strength coefficient. These statistical characteristics validate the regression analysis for the dose versus average ultimate strength relationship.

Table 3. Average Ultimate Strength Regression Analysis

Regression Statistics	
Multiple R	0.988
R Square	0.976
Adjusted R Square	0.969
Standard Error	16.366
Observations	6

ANOVA

	df	*SS*	*MS*	*F*	*Significance F*
Regression	1	42678.628	42678.628	159.342	< 0.001
Residual	4	1071.372	267.843		
Total	5	43750			

	Coefficients	*Standard Error*	*t Stat*	*P-value*
Intercept	-2454.365	204.447	-12.005	< 0.001
Average Ultimate Strength	< 0.001	< 0.001	12.623	< 0.001

Figure 3 shows the relationship between dose and average MOE for the PE-based WPC specimens tested. An overall increase of 8% in average MOE resulted from the 250 kGy EB dose compared to the 0 kGy control. Similar to the dose relationship with ultimate strength, a plot of the average MOE values for the doses tested indicates a direct linear relationship between dose and average MOE.

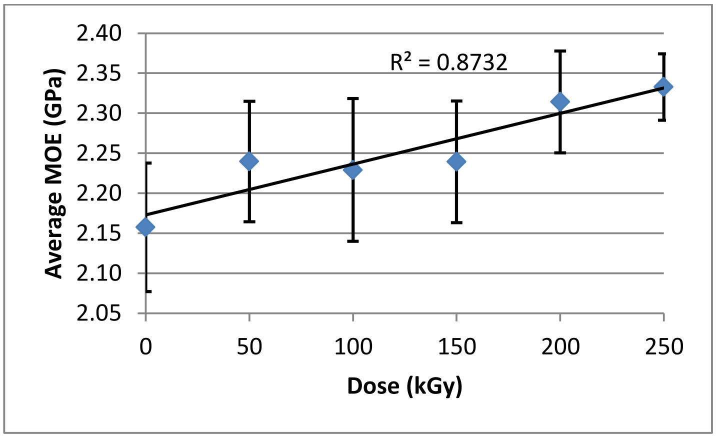


Figure 3. EB Dose vs Average MOE

A simple linear regression analysis for the dose versus average MOE trendline is shown in Table 4. The R^2 and adjusted R^2 values are 87% and 84%, respectively, which demonstrates the predictive power of the model. The overall significance of the regression analysis is approximately 99%, which is equal to the reliability of the average MOE coefficient. These characteristics validate the regression analysis for the dose versus average MOE relationship.

Table 4. Average MOE Regression Analysis

Regression Statistics	
Multiple R	0.934
R Square	0.873
Adjusted R Square	0.842
Standard Error	37.234
Observations	6.000

ANOVA

	df	*SS*	*MS*	*F*	*Significance F*
Regression	1	38204.454	38204.454	27.557	0.006
Residual	4	5545.546	1386.386		
Total	5	43750.000			

	Coefficients	*Standard Error*	*t Stat*	*P-value*
Intercept	-2973.029	590.357	-5.036	0.007
Average MOE	< 0.001	< 0.001	5.249	0.006

5. Discussion

Figure 4 is an SEM micrograph of the non-irradiated LLDPE fracture surface resulting from the bending tests. The long, fibrous strands are evident upon examination of the fractured surface. The ductile fracture mechanism indicated by the white arrows in Figure 5 shows a stretched (tension) region of large plastic strain in the WPC. Figure 5 is the irradiated PE after the 250 kGy EB radiation dose, identifiable by the short, brittle, elastic failure points. Some small amounts of inelastic stretching is indicated by the white arrows in Figure 6, including a void most likely formed by stretching. There did not appear to be any difference in the compression region.

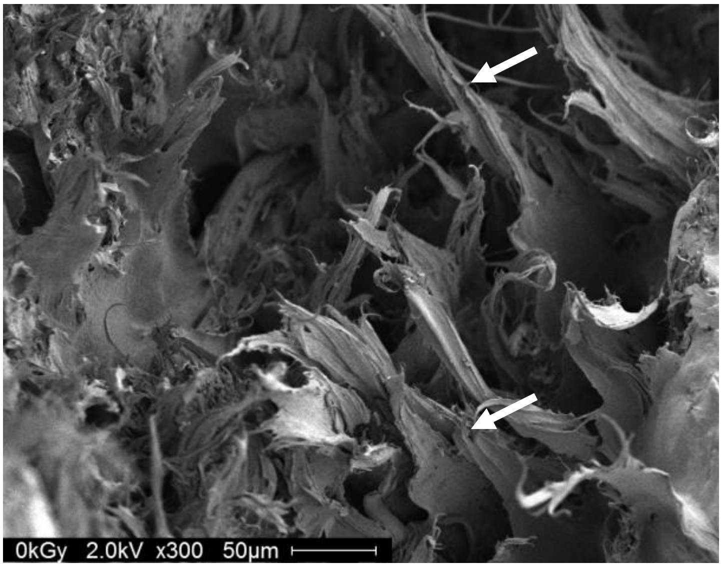


Figure 4. Non-irradiated LLDPE from WPC in the Stretched (Tension) Region.

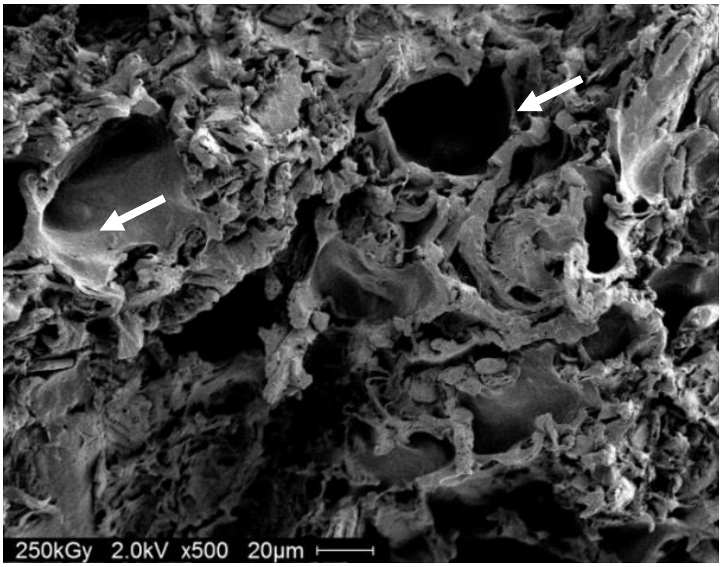


Figure 5. Irradiated PE from the WPC Stretched (Tension) Region.

The dose vs Average MOE relationship in Figures 1 and 3 show that increasing the radiation dose on the deck boards appears to cause a direct linear relationship in terms of ultimate strength; whereas MOE increases from 0 kGy to 50 kGy, then steps, or plateaus, between 50 kGy and 150 kGy, and then increases from 150 kGy to 250 kGy. The "step" could possibly be explained by the percentage of wood fiber in the WPCs. The crosslinking reaction may simply be increasing the strength of the weaker portion of the composite (matrix); therefore, WPCs with varying filler contents may have more uniform strength characteristics with crosslinked matrices. In other words, the increasing stiffness (which is directly related to the MOE) is seen at higher dose levels as larger portions of the matrix are crosslinked, while the results of stiffness are much more unpredictable at lower dose levels. At lower dose levels the crosslinked content is less and the wood fibers have more influence. The white arrows in Figure 6 indicate an example of the voids created by the separation of the crosslinked and non-crosslinked phases within the matrix.

Another possible explanation source of variation may be related to the type of PE in the matrix. While the material safety data sheet claims that the PE-based WPC is manufactured with plastics obtained primarily from reclaimed/recycled grocery bags and stretch film, there is a chance that other types of PE are mixed in with the LLDPE. In this case, the crosslinkability could vary, because low-density PE and high-density PE have G-values (events per 100 eV) ranging from 0.5 to 1.09 (Dawes et al. 2007).

Despite the possibility of variation caused by PE type, or wood content, the stepped behavior is believed to be a result of crosslinking. Gheysari and Behjat (2001) had similar stepped results at the 50-150 kGy range with LLDPE when tensile tests were performed. The stepped result could easily be a product of a larger coefficient of variation for MOE when compared to the ultimate strength for this material (FPL 2010), however, some ductile thermoplastic materials exhibit a rubber toughening effect under similar circumstances (Aly 2012, Decarli et al. 2005). With this theory, between the dose range of 50 and 150 kGy, the crosslinked and non-crosslinked portions form separate phases within the polymer. Voids are formed and shear yielding is increased when these phases are stressed in tension, which requires more energy to break the material (Aly 2012, Decarli et al. 2005). At 200 kGy, a large enough portion of the polymer matrix is in the brittle, crosslinked phase where fewer voids are formed, shear yielding is reduced, and the stiffness begins to increase linearly again.

6. Conclusion

Based on the test results and statistical analyses presented in this paper, EB treatment of the PE matrix in WPCs increased the average ultimate strength and average MOE of the PE-based WPC. The use of EB to increase strength and stiffness could lead to a decrease in the thickness of WPCs or to an increase in joist spacing. Less construction material would have beneficial economic and environmental impacts for both the manufacturer and consumer.

It is recommended that work be conducted at dose levels greater than 250 kGy. Further research at doses higher than 250 kGy would provide useful information about the potential for further strength enhancement. Lastly, a wider variety of mechanical testing, such as creep or hardness testing, would be relevant to the product's in-service performance.

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