Flexural Strength Properties of an Electron Beam Radiation Crosslinked Wood-Plastic Composite

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Abstract:

Wood-plastic composites (WPCs) are building materials that are nontoxic alternatives to pressure treated lumber and are a stronger, sustainable alternatives to plastic lumber. While sustainability is an advantage, concerns regarding their durability and weight have been expressed. Past research has focused on coupling agents and nanoparticles as additives to increase strength properties of WPCs. The focus of this study was to examine the potential benefits of irradiating WPCs in order to enhance the mechanical properties of thermoplastic WPCs. WPCs were irradiated, post extrusion, at dose levels of 0, 50, 100, 150, 200, and 250 kGy with an electron beam. The crosslinked composites 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. SEM images indicate greater polyethylene crosslinking when compared to cellulose chain scissioning.

Introduction:

Wood-plastic composites (WPC) are a mixture of wood particles in a polymer matrix. Wood particles are typically derived from a hardwood species, such as hard maple, oak, or ash. 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 (PET), epoxies, and phenols.

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 building materials such as decking (Clemons 2002). PP-based WPCs are commonly used in automotive products (Clemons 2002).

One of the main concerns associated with the manufacture and use of PE-based WPCs is their weight. The density of WPCs is almost twice the density of solid lumber (Bengtsson et al. 2006). Hollow or shaped cross sections have been used to decrease the weight of PE-based WPCs (Wolcott & Smith 2004). However, attempts to decrease the weight of PE-based WPCs have led to concerns regarding performance. Bengtsson et al. (2006) found a decrease in long-term load performance, because linear polymer molecules are strongly affected by time and temperature. Although reductions in weight are needed, they must be done in a manner that does not reduce mechanical properties like toughness, creep, and strength (Wolcott & Smith 2004, Bengtsson et al. 2006).

Past research has focused on the use of coupling agents and more recently nano-reinforcements to increase strength properties of PE-based WPCs (Faruk & Matuana 2013, Lu et al. 2005). While both methods have shown 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 stiffness of the matrix material by crosslinking the matrix material, thereby creating a thermoset WPC (Bengtsson & Oksman 2006, Kuan et al. 2006, Janigova et al. 2001, Reyes et al. 2001). Reinforcement of polyethylene by forming crosslinks using peroxide initiated or vinyl silane grafting crosslinking methods has been shown to improve mechanical properties in WPCs (Bengtsson & Oksman 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 strength properties were increased.

Radiation crosslinking of PE has been researched since the 1950s (Charlesby 1952, Dole et al. 1954). Polyethylene crosslinking occurs after hemolytic 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. Initiation and termination are the dominant reactions; however some propagation reactions may occur due to trans-vinylene double bond formation. The resulting polyethylene thermoset will be stiffer and harder than the thermoplastic (Bharat 2000). Electron beam processing of polyethylene has carved out established market niches in wire jacketing, hot water tubing, closed cell foams, and joint replacement (Atkinson & Cicek 1983, Manning et al. 2005, Berejka & Cleland 2011).

When cellulose, the primary structural component of wood, is exposed to electron beam radiation, the dominant reaction is cellulose chain scissioning (Berejka & Cleland 2011). The resulting degradation of the wood particles may occur in a radiation crosslinked wood thermoplastic composite. This paper presents the results of a comparative evaluation of a PE-based WPC subjected to five different doses of radiation. Results are presented in terms of the flexural strength properties.

Materials:

PE-based WPCs (3.66m lengths) were purchased from a local supply store. The specimens were cut into 0.91m sections. The PE-based WPCs composite are an extruded composite blend of PE (40-50%), wood particle (50-60%), and less than 1% carbon black by weight. According to the manufacturer, the PE is mostly linear low density polyethylene (LLDPE), derived from recycled grocery bags and stretch film. The source of the wood particles is obtained from furniture makers or waste pallets. The average weight of the specimens is 3.1 kg.

Methods:

The 3.66m PE-based WPCs specimens (56 total) were irradiated with a Dynamitron 3.0 MeV, 30 milliamp electron beam accelerator at IBA Industrial, located in Edgewood, NY. Nine replicates were treated at five different dose levels 0 (control), 50, 100, 150, 200, 250 kGy.

After irradiation and prior to testing the specimens were stored in a conditioning chamber at 21˚C and 45% relative humidity (RH) for approximately four months. 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 in size were cut from an inside section of each of the original 0.91 m purchased board, placed in an oven at 103˚C for two days, then placed in a desiccator to cool. The moisture content of the specimens was measured in accordance with ASTM 4761. The moisture content of the specimens ranged from 1.0-1.3%, consistent with values published by the manufacturer.

Third-point bending tests were conducted using a Tinius Olsen 120,000 lb. Electomatic Universal Testing Machine in accordance with ASTM D4761. A 2,268 kg load cell was applied at a constant 1.7 cm/min crosshead speed to failure. A linear variable differential transformer (LVDT) conditioner was in place to detect deflection. The linear region of the stress-strain curve was estimated to be between 345 and 690 KPa, based on the R-value of the trendline.

Images of the fractured surfaces of the PE-based WPCs were taken using a JEOL JSM-5800 low vacuum SEM.

Results and Discussion:

Mechanical Properties:

The average ultimate strength and range of results for the PE-based WPC specimens tested are shown on Figure 1. An overall increase of 10% in average ultimate strength resulted from the 250 kGy EB dose compared to the 0 kGy control. The result for each dose varied by approximately 1.7% (250 kGy) for the specimens tested. A plot of the average values for the doses tested indicates increasing linear relationship as dose increases ($R^2 = 0.9755$).

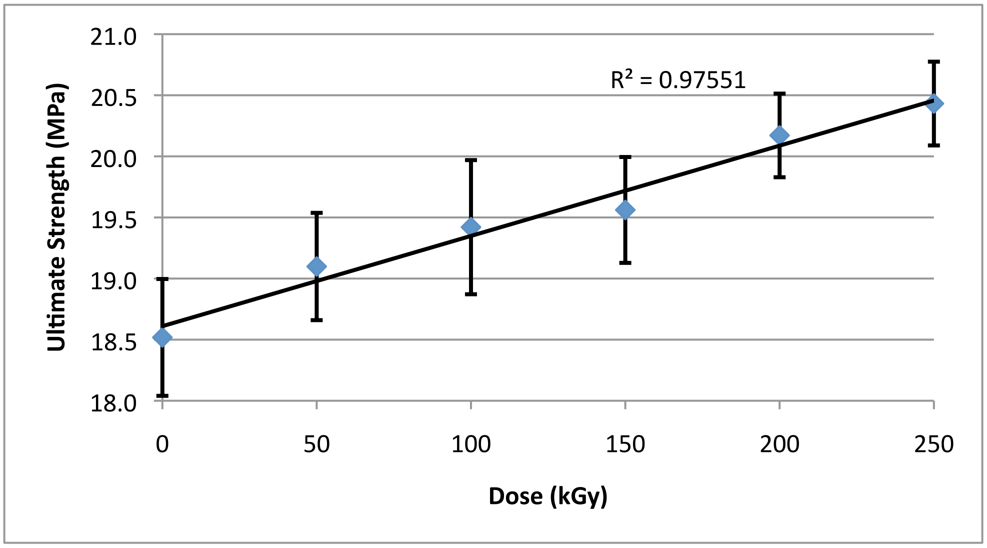


Figure 1. EB Dose vs. Average Ultimate Strength

Figure 2 shows the average modulus of elasticity (MOE) and range of results 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. The results for each dose varied by 1.8% (250 kGy) for the specimens tested. A plot of the average MOE values for the doses tested also indicates an increasing linear relationship as dose increases ($R^2 = 0.8732$).

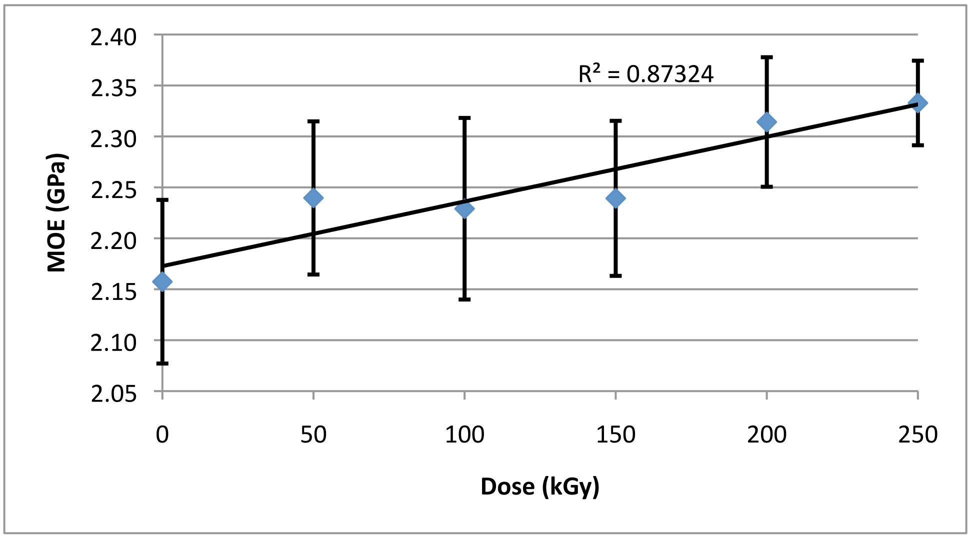


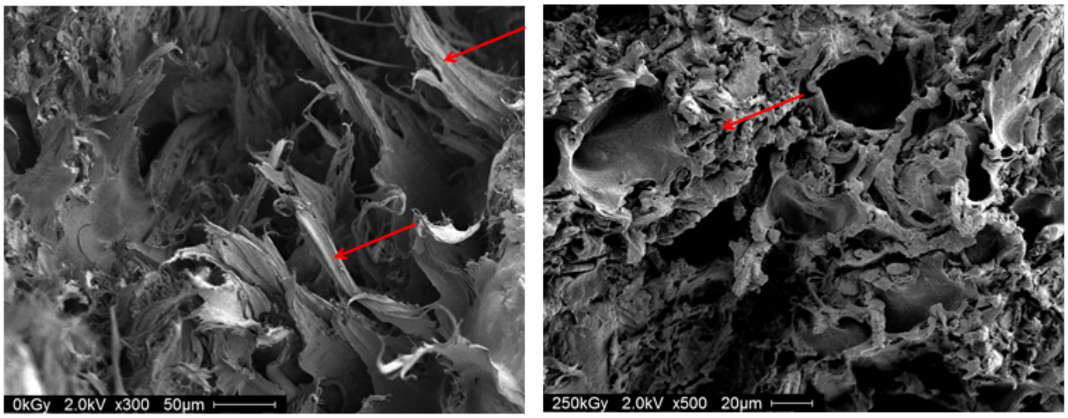
Figure 2. EB Dose vs Average MOE

The effect of increasing radiation dose of the WPCs appears linear in terms of ultimate strength, whereas MOE appears stepped between 50 kGy and 150 kGy, although an increasing trend in MOE vs does was still observed. Linear behavior was then apparent from 150 to 250 kGy. This variation could possibly be explained by the percentage of wood filler 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.

Another possible explanation, for the stepped MOE behavior may be related to the PE matrix alone. Gheysari and Behjat (2001) had similar stepped results, at the 50 kGy-150 kGy range with a LLDPE, when tensile tests were performed. When compared to high density PE (HDPE), LLDPE content has a wider overall amorphous domain distribution, a significantly larger amount of branching, and thus a greater potential for crosslinking (Wiesner 1991). Therefore, more crosslinking reactions would occur within specimens with higher LLDPE content and these specimens would increase in strength more quickly than the specimens with higher HDPE content, creating more equilibrium at higher dose levels, and therefore a more defined increase in strength in the LLDPE specimens. Wiesner (1991) discusses this in more detail, arguing that even in crosslinked PE, the crystalline state, as long as it is intact, has a heavy influence on the mechanical properties. Wiesner (1991) states it may be possible that different polymer structures require different specific amounts of crosslinks for a certain reduction of deformation. This would explain the consistent stepped behavior in crosslinked PE at the 50 to 150 kGy range.

Scanning Electron Microscopy:

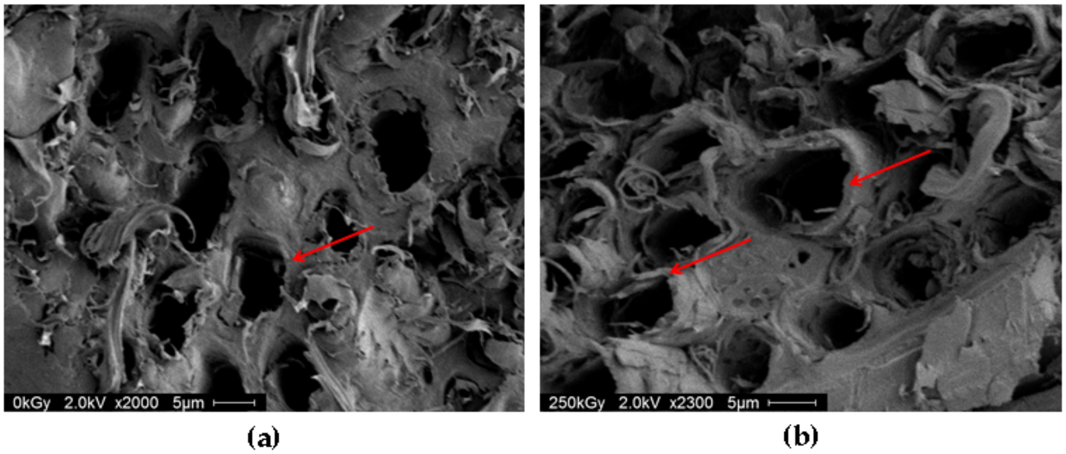
The ductile fracture mechanism indicated by the red arrows in Figure 3a (non-irradiated) shows a highly stretched region of large plastic strain in the WPC. A distinct brash failure through the brittle fracture surface of irradiated polyethylene appears in Figure 3b. This indicates a matrix stiffened by crosslinking.



(a) (b)

Figure 3. SEM images of non-irradiated (a) and irradiated (b), fractured polyethylene cross sections.

Figure 4b shows a vessel element present in a WPC specimen that was fractured during testing. Minimal difference in failure of non-irradiated (Figure 4a) verse irradiated wood particle (Figure 4b) is shown. The red arrows indicate the cell wall fracture. The fracture surface of the cell wall in Figure 4b indicates only a slight increase in brash failure.



(a) (b)

Figure 4. SEM images of non-irradiated (a) and irradiated (b), fractured wood-fiber cross-sections.

A more brash failure of the treated specimen would indicate wood degradation by electron beam. The transfer of the stress from PE to wood particle that results in the fracture of the wood particle is, therefore, a likelier result of the stiffening of the PE matrix, rather than a weakening of the wood particle.

Conclusion:

Based on the test results presented in this paper, electron beam radiation crosslinking of the polyethylene matrix in WPCs directly increased the ultimate strength and stiffness of the WPC. The degradation of wood was an initial concern; however, the less distinct brash failure through the fracture of the wood particle indicates greater polyethylene crosslinking and less cellulose chain scissioning. The use of EB to increase strength and stiffness could lead to a decrease in the thickness of WPs or increase in joist spacing to meet current load requirements.

Lower standard deviation (error bars) at high dose levels also indicates a more uniform product, but further investigation is needed to support this claim.

Further studies are needed to determine a wider variety of mechanical properties (creep resistance in particular), the dose level at which the composite begins to lose strength, and the possibility of incorporating coupling agents and nano-reinforcements into the crosslinked matrix for further strength enhancement. Due to the fact that crosslinked PE is also proven to have better mechanical properties at elevated temperatures (Weisner 1991); it would be useful to have a better understanding of the mechanical properties at those temperatures.

Also of interest, and possibly crucial to any industry acceptance would be a cost-benefit analysis of a high-energy electron beam into an extrusion line. Increasing the dose level may result in a stronger, stiffer material, but this must be weighed against the initial cost of incorporating the accelerator into the extrusion line. One can see, however, the advantage of a stronger, stiffer WPC. A thinner WPC would then satisfy the current 16-inch-on-center span recommendation for the decking product. However, if the same thickness is used, a wider span between joist supports could be implemented. In either case, fewer materials are used in manufacturing, resulting in enhanced overall industrial efficiencies along with environmental benefits.

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