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Chemical constituent influence on ionizing radiation treatment of a wood–plastic composite

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HIGHLIGHTS

- Feasibility of electron beam treatment of a wood plastic composite.
- No significant impact on modulus of elasticity.
- Increase in ultimate strength and hardness.

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ABSTRACT

A dose range of 0–200 kGy was used to irradiate polyethylene-based wood plastic composite (WPC) specimens. An evaluation of mechanical properties in bending resulted in an increase in ultimate strength, but little effect to stiffness. The bending test results were compared to previous testing in order to examine reproducibility. Hardness tests were also conducted, revealing an increase at a dose range of 0–250 kGy.

1. Introduction

Methods involving the use of electromagnetic radiation to cure the resin in engineered wood products can be found in research publications and reviews (Cleland et al., 2003; Maloney, 1996; Pereira et al., 2004). Radio frequency waves, for example, have been used to cure medium density fiberboard (Pereira et al., 2004) as well as laminated veneer lumber adhesives, and a microwave-type heating system has been used to cure the thick billet sizes of parallel strand lumber (Maloney, 1996). Irradiating engineered wood products with ionizing radiation is also an area of research still being explored (Larsen, 2006). Electron beam (EB) irradiation is a well-established method of crosslinking polymers (Berejka and Cleland, 2011). Electron beam (EB) irradiation is used to create and enhance polyethylene (PE)-based products; therefore the concept of treating extruded polyethylene (PE)-based wood plastic composites (WPCs) with EB radiation should provide a feasible method of enhancement. This feasibility study is based on a light construction material, commonly used in residential deck construction that is commercially available at any supply store.

This paper investigates the effects of the individual components of a WPC after irradiating the lignocellulosic composites with ionizing radiation from an EB. In a previous study (Palm et al., 2014) polyethylene crosslinking dominated the degradation of the cellulosic wood fibers and increases in strength and stiffness were found. In this study, the flexural strength and stiffness of the PE-based WPC is determined and compared to previous work (Palm et al., 2014), which should indicate any possibility of reproducibility. The hardness of the material is also examined.

2. Constituent properties and radiation effects

The 2.54 cm (1″) thick 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 TREX Accents composite decking, rated for 4.8 kPa at a 0.4 m (16″) on center span. This was

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the same material used in a previous study by the authors of this current study (Palm et al., 2014). While most information is proprietary, some information on the product tested was available from the manufacturer. Based on the manufacturer's literature, the PE-based WPC is an extruded blend of wood fibers (50–60%) in a PE (40–50%) matrix, and less than 1% carbon black by weight. The material safety data sheet (MSDS) states that the PE is mostly linear low-density PE (LLDPE), derived from recycled grocery bags and stretch film. The source of the wood fibers is waste material, obtained from furniture makers or waste pallets.

According to the manufacturer's information combined with statistical sampling data (Kollmann and Côté, 1984), the four main chemical constituent percentages are: PE (40–50%), cellulose (19–26%), hemicelluloses (10–24%), and lignin (9–21%). Cellulose, hemicellulose and lignin are the chemical constituents of wood fibers that make up the 50–60% of the composite. Between 0 and 250 kGy, radiation has have one of three effects on each chemical constituent (Berejka and Cleland, 2011): (1) crosslinking of the polymer chains is the primary effect on PE, (2) main chain scission is the primary effect on cellulose along with hemicelluloses, and (3) radiation has no effect on lignin. Ideally for this experiment, crosslinking of polyethylene would dominate over scissioning of cellulosic material. These constituents are discussed in the following sections.

2.1. Polyethylene

The radiation crosslinking benefits of polyethylene were discovered in the late 1940s and early 1950s as a result of work done by Malcom Dole's group (Rose, 1949) and Charlesby (1952). PE crosslinking occurs after hydrogen abstraction reactions, initiated 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 crosslinked polyethylene exhibits valuable increases in mechanical strength properties when compared to that of polyethylene (Bharat, 2001; Dawes et al., 2007). The increase in some of these properties at elevated temperatures finds value in higher temperature applications such as wire insulation (Berejka and Cleland, 2011). EB processing of PE is used in wire jacketing, hot water tubing, closed cell foams (Berejka and Cleland, 2011), and joint replacement (Atkinson and Cicek, 1984; Manning et al., 2005).

Crosslinking, G(X) values (events per 100 eV) for LLDPE are 0.7–1.09 (Dawes et al., 2007). An overall average of G(X) value of 1.0 was determined for LLDPE, low-density PE (LDPE), and high-density PE (HDPE). Scission in PE occurs in low numbers with G(S) values of less than 0.1 (Dawes et al., 2007).

2.2. Cellulose

Ionizing radiation is believed to break down the glucosidic bonds in cellulose chains, but there is evidence of cleavage of bonds within the pyranose ring as well. Substantial radiation chemical yields of glucose and xylose have been reported by Ershov and Klimentov (1984). Other evidence for the reactions listed is the formation of carbonyl and carboxyl groups at the site of bond cleavages, along with the evolution of hydrogen, carbon dioxide and carbon monoxide gases. Infrared spectra showed changes after irradiation that leads to breakdown of the ordered system of intermolecular hydrogen bonds (Ershov and Klimentov, 1984). This means that hydroxyl groups are involved in less stable hydrogen bonds. The molecular weight drops as radiation cleaves both the amorphous and crystalline regions of cellulose (Driscoll et al., 2009). X-ray diffraction indicates that at a dose of 9400 kGy, cellulose can be characterized as having an amorphous structure (Ershov and Klimentov, 1984).

The yield of degradation or G(S) value of various kinds of cellulose materials is 6.0 ± 1.0 (Charlesby, 1952; Dawes et al., 2007). However, this measurement is independent of dose rate, initial degree of polymerization, or atmospheric conditions of irradiation.

2.3. Hemicelluloses

Cellulose fibrils are embedded in a matrix material within the cell wall that consists of hemicelluloses. Hemicelluloses are polymers built from several different kinds of sugar monomers, mostly glucose and xylose. Hardwoods in particular have a uniquely high content of a partly crystalline, partly acetylated xylan, which accounts for 20–35% of solid hardwood. A second hemicellulose, glucomannan, accounts for 3–5% of solid hardwood (Kollmann and Côté, 1984). In softwoods, however, xylan makes up 10% of the total, and another hemicellulose, galactoglucomannan, makes up 20%.

Some literature is available on the radiolysis of xylan and glucomannan (Ershov, 1998; Rakhimov et al., 1990). Once irradiated, the radicals detected for xylan are the same as those formed upon irradiation of cellulose, and therefore it can be assumed that ionizing radiation results in cleavage of the glycosidic bond. The production of CO_2 also indicates rupture of the pyranose ring. Despite the similarities with the radiation chemistry of cellulose, the calculated degradation yield is 8.0 ± 1.0, slightly larger than cellulose. In other words, xylan will degrade more rapidly than cellulose (Ershov, 1998). The process of degradation was also found to be dominant when glucomannan was exposed to 100–400 kGy doses of gamma radiation (Rakhimov et al., 1990).

2.4. Lignin

Lignin is a complex, three-dimensional, phenyl-propane polymer. Wood fibers have lignin and possibly other extractives that will serve as protection from radiation (Mclaren, 1978; Smith and Mixer, 1959). It is accepted that highly aromatic polymers such as polystyrenes, epoxies, along with aromatic polyamides and polyesters are radiation resistant (Dawes et al., 2007). While some show signs of crosslinking and others scissioning, lignin remains practically unchanged at levels as high as 1800 kGy (Skvortsov, 1990). The aromatic type phenyl groups within lignin are highly resistant to radiation degradation. In their study of Redwood chips, Smith and Mixer (1959) reported that at 65 kGy, the average number of cellulose chain breaks per molecule by gamma radiation almost doubled when lignin and other extractives were removed from cellulose and hemicelluloses. Ershov and Klimentov (1984) found the yields of carbohydrate radicals of three to five times more in independent cellulose when compared to irradiated wood.

The aromatic ring structure has alternating double and single bonds, which means two sigma bonds and one pi bond per carbon atom, and therefore one sigma bond at each atomic bond location. Pi bonds are delocalized around the benzene ring. The beam energy from the accelerated electrons resonates through the cyclic ring structure, having little effect.

3. Methods

3.1. Electron beam

10 WPC deck boards at 3.66 m (12′) length were cut into 0.91 m (3′) sections. The resulting dimension of the 40 specimens is $2.54 \times 13.97 \times 91.44$ cm^3 ($1 \times 5.5 \times 36$″). All WPC specimens were irradiated with a 90 kW, 3 MeV Dynamitron EB accelerator at IBA