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Development of Rubber and Agricultural Fiber based Biocomposite for Industrial Application

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ABSTRACT

Flax, which is known for its linens and oils that are used for industrial products, can also be utilized as a cost effective and environmentally acceptable approach to the creation of a partially biodegradable biocomposite. Biocomposite material is investigated by combining recycled tire rubber, flax and linear low density polyethylene (LLDPE). The manufacturing process which be used to fabricate the biocomposite product included Extrusion and Compression Molding. Optimizing and studying the composition percentages of the compounds were studied in this paper. Moreover, the properties of the product were observed by using tensile test, tearing test, water absorption test, hardness test and Differential Scanning Calorimetry.

Keywords: Flax, LLDPE, Recycled tire rubber, Biocomposite, Extrusion, Compression molding

INTRODUCTION

Recently, there have been a number of research studies on the replacement of synthetic fiber with agricultural fibers as the reinforcement in plastic industry [Joffe et al. 2003; Panigrahy et al. 2006]. The advantages with natural fiber used as potential reinforcing replacement of synthetic fiber in composite, are its low density, low processing temperature, non-abrasive nature, renewable and relatively cheaper in cost [Mohanty 2003; Velde 2002]. Flax fiber is the most common fiber reinforcing replacements under study in Canada. This is because oilseed flax crop is abundantly grown in Western Canada and a strong bast fiber can be extracted for commercial and industrial use, e.g. composite application.

Rubber is the general term of elastomer, which requires vulcanization to form and has the characteristic of a thermoset. Waste tire rubber is not a favorable material for recycling due to its thermoset characteristics. Presently, the recycled rubber has not been used as a replacement for new or synthetic rubber in significant quantities, because the current devulcanization technique cannot achieve the desired properties to replace the virgin rubber in cost-effective way [CalRecovery 2004]. Therefore the waste tires are left for the landfills and junkyards, and sometimes they are used as a fuel by incineration [Adhikari et al., 2000]. There is only a small percentage of waste tire rubber being mostly recycled and used in landscaping applications and some other products in Canada. Therefore there is a great need to find some value-added applications for waste tire rubber.

The initial goals of this research were to integrate the agricultural residue and recycled rubber in manufacturing of new materials, and to generate some inventive materials equivalent to existed ones by replacing of synthetic fiber with flax fiber/shive.

Developing new and innovative products utilizing flax crop is important. First, the flax crop production is added the value. Second, the environmental is cleaned by reducing CO_2 release of burning the crop in the field as well as using recycled tire rubber as one of the main component of this material. Moreover the environmentally unhealthy synthetic fibers such as fiberglass can be reduced in the plastic industry.

The key item of this research was the recycled tire rubber biocomposite materials. The thermoplastic is utilized as the binder, and recycled tire rubber acts as matrix. The principal reinforcement will be natural flax fiber derived from oilseed flax straw. The flax shive acts as the filler in the biocomposite.

The overall goal of this research project is to investigate a viable biocomposite material from the flax fiber/shive, recycled tire rubber and thermoplastics, and to optimize the extrusion and compression molding process. The specific objectives of this project are:

1. To develop a new biocomposite material made from flax fiber/shive, recycled rubber and thermoplastics via extrusion and compression molding processes.

2. To determine and optimize composition percentages of the compounds and processing variables for the

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biocomposite material and determine their physical, mechanical and thermal properties.

3. To investigate the optimum flax fiber/shive loading in the biocomposite.

MATERIALS

The basic ingredient for the rubber biocomposite was the ground recycled tire rubber, untreated flax fiber and thermoplastic. A small amount of lubricant (wax) was added to the biocomposite to provide a decrease of resistance with the equipment during the process.

OILSEED FLAX FIBER - Flax fiber is an important bast fiber from the dicotyledonous Linum usitatissimum plant. Oilseed flax plant has a single stem, nearly one meter in height and the flax stalks vary between 2.5 to 4 mm in diameter [Britannica 2008]. Flax fiber strand is in the range of 30 to 75 cm [Britannica 2008]. Table 1 shows the chemical composition of flax fiber and shive. Flax fiber can withstand processing temperatures as high as 250ºC [Sreekala et al. 2000]. The strength characteristics of fiber depend on the properties of the individual constituents, the fibril structure and the lamellae matrix. Flax fiber possesses high specific strength and stiffness and can be used as reinforcement in polymeric resin matrix to make useful composite materials [Joffe et al. 2003; Panigrahy et al. 2006]. Mechanical properties of the flax fiber are shown on Table 2.

Table 1. Comparison between chemical composite of flax fiber and flax shive [Panigrahi et al. 2006]

	Composition (%)				
	Cellulose	Hemicelluloses	Lignin	Extractives	Ash
Flax fiber	78	6	5	2	2
Flax shive	53	13	24	15	>2

Table 2. Mechanical properties of flax fiber [Bledzki et al. 1999]

	Density (g/cm^3)	Elongation (%)	Tensile Strength (MPa)	Young's Modulus (GPa)
Flax fiber	1.5	2.7 – 3.2	345 - 1035	27.6

The decorticated flax fiber (clean fiber), contained about 80% fiber and 20% shive, Saskatchewan-grown oilseed flax, was produced and supplied by Bio-fibre Industries Ltd. at Canora, SK, Canada. The natural flax fiber was used as received without any treatment. The flax fiber was ground by knife mill (passed through 2mm screen). The shives served as structural stabilizing filler and the fibers acted as reinforcement that aided in reducing the flexibility of the rubber material and therefore maintaining its structural integrity [Rana et al. 2006]. The particle density of the flax fiber was 1576 ± 85 kg/m^3, determined by using the gas pycnometer (MVP-2, Quanta Chrome Corp., Boynton Beach, FL, USA). The degradation temperatures and glass transition temperatures of fiber and shive were found from the Differential Scanning Calorimetry (DSC) method.

RECYCLED GROUND TIRE RUBBER - Recycled ground tire rubber (GTR) was supplied from Shercom Industries Inc., Saskatoon, SK. Canada. The scrap rubber tires were ground into fine particles. The particle size of the ground tire rubber was roughly over 90% retained in the sieve opening size from 0.210 to 0.589 mm. The geometric mean particle diameter was 0.415 ± 0.15 mm which determined according to ASTM D5644 standard [ASTM 2003f]. The particle density was 1226 ± 12 kg/m^3, determined by using the gas pycnometer.

Due to the vulcanization on the rubber, the polymer molecules are very strongly cross-linked. This strong covalent bonds and linked cause the rubber an insoluble and infusible thermosetting polymer [Wikipedia 2007a]. However, a tire is mainly made of rubber compound and cords. The rubber compound was made by mixing polymer, reinforcement materials, softener and various chemicals. The main materials for tire are carbon black, polymers, antioxidants and waxes [RMA 2007; Toyo Tire 2007]. The main polymers used for tires are listed on Table 3. All of the following polymers are synthetic rubber except Natural Rubber. The average tensile strength of the new tires is 13.73 MPa on the tread part and 8.53 MPa on sidewall [Ratrout et al. 2006].

Table 3. Main polymers used for tires [Toyo Tire 2007]

Polymer	Uses
Natural Rubber (NR)	General Use, TB Tread
Styrene Butadiene Rubber (SBR)	PC Tread
Butadiene Rubber (BR)	Sidewall
Isoprene Rubber (IR)	Partially used to replace NR
Halogenated Butyl Rubber	Inner Liner

Some of the advantages of using GTR in the biocomposite material are: natural thermal insulation ability, high flame resistance and extreme resistance to degradability. However, some fine metal belts and fibers still could be found in the ground tire rubber.

THERMOPLASTICS - Linear Low Density Polyethylene (LLDPE): ExxonMobil (Escorene®) LLDPE LLP8460.29, rotational molding resins in 35 US mesh powder form, was used as basic binder. LLDPE is economical, strong in stiffness, impact resistant and good moisture barriers. LLDPE is naturally translucent and flexible; and has UV-8 type protection [Safetanks.com. 2007]. Some of the important properties are enlisted on Table 4 and Table 5. Moreover, the glass transition temperature is also concerned for the end-use for outdoor cold winter. Nominal LLDPE has glass transition temperature at –73˚C [InterNet 2007].

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Table 4. Important physical and mechanical properties of LLDPE [Matweb 2006]

	Density (g/cm^3)	Melt Flow Index (g/10min)	Tensile Strength (MPa)	Flexural Modulus (GPa)
ExxonMobil LLDPE LLP 8460.29	0.938	3.30	17.8	0.772

Table 5. Important thermal properties of LLDPE [Matweb 2006]

	Melting Temp (^oC)	Crystallization Temp (^oC)	Deflection Temp at 0.46 MPa (^oC)	Deflection Temp at 1.8 MPa (^oC)
ExxonMobil LLDPE LLP 8460.29	127	112.7	62	39

PARAFFIN WAX - "CONROS" Parowax (Conros Corp, North York, ON. Canada), a pure refined paraffin wax, was used as the lubricant in this study. Paraffin wax is a mixture of saturated aliphatic hydrocarbons refined from petroleum [Lutz et al. 2001]. Paraffin wax is a white, odorless, tasteless, waxy solid, insoluble in water, with a typical melting point between about 47°C and 64°C [Wikipedia 2007b]. Density of paraffin wax is 0.93 g/cm^3 [ASTAR 2007]. This wax was used extensively because of low cost, good color, relative nontoxicity, and reasonable stability to heat, light and oxidation [Lutz et al. 2001]. Adding paraffin wax may serve as the most suitable way to improve the process ability of the biocomposite.

EXPERIMENTAL DESIGN

In this experiment, flax fiber/shive content ranged from 0% to 20% w/w; GTR content ranged from 40% to 50% w/w and LLDPE ranged from 25% to 40% w/w. Eight different compounds were prepared using ground flax fiber, GTR, LLPDE and wax. The composite ratios of the mixtures prepared are given as in Table 6. Two compounds were without the extrusion process, and one was without using wax in the compounding.

Table 6. Composite ratios of the compounds

Sample Code	Extrusion	Composition (% in w/w)			
		Flax Fiber/ shive	Ground Tire Rubber (GTR)	LLDPE	Wax
A	Yes	10	40	40	10
B	Yes	15	45	30	10
C	Yes	20	40	30	10
D	Yes	15	50	25	10
E	Yes	20	45	25	10
F	Yes	0	60	30	10
G	No	0	66.67	33.33	0
H	No	0	60	30	10

COMPOSITE PREPARATION AND PROCESSING TECHNIQUES

Flax fiber was ground to short element by using the knife mill (Model 4, Thomas Wiley Laboratory Mill, Thomas Scientific, NJ, USA) with 2 mm screen in our laboratory. Ground tire rubber was run through a large magnet to pick up the rest of the fine metal belts in the rubber. Before extruding, the compound materials were fully blended using the mixer, and stored in sealed plastic bags.

EXTRUSION - Extrusion process basically consists in continuously shaping a fluid polymer through the orifice of a die in the extruder and subsequently solidifying into a product [Henson 1997]. The blends of the polymer compositions can be carried out by a single-screw extruder [Grigoryeva et al. 2005]. The cross-linked rubber can be selectively broken under high shear stress, high temperature, and high internal pressure in a screw type extruder [Fukumori et al. 2003]. The single-screw extruder machine (Akron Extruder Model# 12520, Batavia, OH, USA) with a length/diameter ratio of 20, a barrel diameter of 31.75 mm, 3 barrel heating zones was used to compounding the composite in this study.

The mixture of flax fiber, ground rubber tire, LLDPE, and wax was fed into the single screw extruder with heating barrel to be heated and thoroughly mixed. This step was performed to insure that flax fiber is homogenously distributed in the polymer during the molding process. Some cross-linkages of GTR and LLDPE can be happened during the extrusion compounding process.

The screw speed was run from 30 to 60 rpm. Barrel zone temperatures were set to 171°C (340°F), 188°C (370°F) and 199°C (390°F) from first to third respectively, while the die temperature was at 202°C (395°F). Those temperature setups were optimal after preliminary trials for extrusion of recycled rubber and LLDPE compound. After extrusion, the compounded materials were pelletized and prepared for compression molding.

COMPRESSION MOLDING – The compression molding is a molding method in which the material is preheated and placed into a open heated mold cavity, then the mold is closed, the top plate with pressure is applied to force the melted material to contact all the area inside the closed mold, and the heat and pressure are maintained until the molding material has cured. Hot compression molding method was used in the studies on the composites of tire rubber particles mixed with short sisal fibers [Martins and Joekes 2003; Martins and Mattoso 2004].

An in-lab square steel mold was used to mold a 20 cm x 20 cm square rubber composite sample. The heated press machine (Model 44-226, Miller Machine Tools, J.B. Miller Machinery & Supply Co. Ltd. Toronto, ON, Canada) was used to do the molding.

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