The use of Polyethylene and Rice Straw for the Production of Wood Plastic by using New Coupling Agent

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ABSTRACT

This research studies the use of rice straw (RS) and polyethylene(PE) in the production of wood plastic composite (WPCs), using PE wax as a new coupling agent. PE wax, a by-product in the Sidpec Company, is incorporated in the matrix

to act as a coupling agent between the matrix and the straw. FTIR spectrum for this wax indicate a low-density polyethylene and containing C=O group, similar to indicated of the C=O group which is similar to the maleic anhydride grafted.

The raw materials were compounded in a two roll-mill, and the final composite were obtained by controlled hot-press moulding with varying rice straw content (20,35,and 50 wt.%), and addition of coupling agent (PE wax).

(PE-RS) composites exhibited excellent dimensional stability and mechanical properties. Incorporation of PE wax coupling agent in composite formulation improved the dimensional stability, durability, and the mechanical properties. WPCs based on (PE-RS) composites have potential application in construction industry. Hence, WPC can work on reducing the Egyptian natural wood as it will have the same appearance but with better properties such as resistance to fungal and thermite attacks, less water absorption and ease of manipulation.

The mechanical properties of the composite in tensile, and impact strength for three different rice straw mass fractions have been examined. Preliminary studies showed that the addition of this wax has improved the properties of the composite.

1. INTRODUCTION

Natural fiber-reinforced plastic composites have recently gained importance in various applications especially in the field of building materials and automotive components. The fibers offer advantages of large availability, annual renewability, low cost, light weight, competitive specific mechanical properties, reduced energy consumption, and environment friendliness.

Wood-plastic composites (WPCs) are now being marketed for various applications such as building products, automotive and packaging materials. As building products WPCs are used for applications like decking, fencing, siding, window frames and roof tiles. However, the use of WPC as building products has resulted in concern about the durability of these products when exposed to outdoor environments [1-4]. Outdoor durability includes thermal stability, moisture resistance, fungal resistance, and ultraviolet (UV) stability [3, 5]. UV exposure is reported to cause the composites to undergo photo-degradation leading to undesirable effects, including loss in mechanical properties and surface quality[6-8]. The moisture is absorbed by WPCs due to the hydrophilic characteristic of the wood component, this results in swelling, dimensional instability and degradation of mechanical properties [6-8]. The exposure to both UV radiation and moisture cause more detrimental effects to WPCs than the exposure to UV radiation alone [9].

The use of compatibilizers in natural fiber reinforced polymer composites is to improve poor interface adhesion between the hydrophilic fiber and the hydrophobic polyolefin matrix. Among numerous compatibilizers are maleic anhydride grafted polyethylene and polypropylene (MAPE and MAPP). They are considered to be the most effective interphase modifiers for polyolefin / natural fiber composite. Their success depends on the polar interaction and covalent link between anhydride carbonyl and hydroxyl groups of the fiber surfaces [10–13]. As well as their good compatibility with matrix [14–18]. The improvements with the coupling agent are believed to be due to the formation of ester bonds between the anhydride carbonyl group of MAPE and hydroxyl groups of the wood fibers [19, 20].

Literature study showed that the main natural fibers used to reinforce thermoplastics mainly include wood, cotton, flax, hemp, jute, sisal, and oil palm fibers [21]. The rice straw fiber is important reinforcing filler for thermoplastic composite because of its cellulosic characteristics. The whole rice straw fiber used is composed of leaf sheath, straw leaf blade, straw stem and knot [22]. In Egypt rice cultivation in the river Nile Delta produces large amounts of rice straw as residue. The composition analysis of rice straw was the following: cellulose (41-57%), hemicelluloses (33%), lignin (8-19), residual ash (8-38%), [23]. Due to its low nuitrituion value for cattles and its bulk size, farmers tend to dispose of the rice straw by burning which results in the emission of large volume of smokes. This causes a significant contribution to the air pollution known as the "Black Cloud" [23].

In this study, the wood plastic composites are prepared from a mixture of (HDPE)/rice straw particles (RS). The addition of PE wax as a compatibilizer or coupling agent between the hydrophilic rice straw particles and the hydrophobic PE matrix was also studied. The IR spectrum of PE wax (fig 1), produced as a by – product from SIDPEC company, showed the presence of the C=O in the structure of the PE wax, which indicated it might play the role of the coupling agent by forming an ester group similar to the MAPE with the hydroxyl group of the rice straw. The effect of addition of UV stabilizer was also checked for importing mechanical stability towards long time exposure to UV light. The composites were manufactured via a two roll-mill, and the final composite formed by mechanical hot-press moulding, and tested for tensile, and impact.

2. Materials and Methods

2.1. Materials

High density polyethylene (HDPE) was obtained from *Sidi Kerir Petrochemical Company (SIDPEC)*, with density 952 kg/m³, melt index 26 g/10min and melting temperature 126°C as indicated by the manufacture. PE wax was obtained also from *SIDPEC* with density 900 kg/m³ . The structure of the PE wax has been characterized by infrared spectra which is illustrative in (Fig. 1), shows a peak at 720 cm⁻¹ which is characteristic of the =C-H group, a peak at 1465 cm⁻¹ which is characteristic of the alkyl CH, a peak at 2920 cm⁻¹, which is characteristic of the alkane C-H, and a peak at 1638 cm⁻¹, which is characteristic of the C=O group [$Y \notin$].. The ASTM D664 has shown it has an acid number of 1.2(mg KOH/g). Rice straw was obtained from Egyptian farms. The straw was dried and milled to pass through a 20-mesh screen. Chimasorb UV stabilizer obtained from *Ciba Speciallity Chemicals*, was used as received with molecular weight >2500, and melting range (100-135°C) as indicated by the manufacturer.



Fig. (1) FTIR for Polyethylene wax

2.2. Preparation the rice straw (RS) fiber / HDPE composites

The prepared rice straw fiber was further dried in an oven at 75°C for 10 hr to remove the moisture. The compounding process used for preparing rice straw (RS) fiber / HDPE composites included two steps. First step:-all the HDPE was melted at 170 °C at a rotation speed of 60 rpm, in the two roll-mill (Shaw Robinson / 2799), then the PE wax and UV stabilizer were added. Later, the RS was added to the mixture and the process continued till homogeneity was obtained. The second step:- The resulting blend was placed in a three-piece stainless steel molding set and compression-molded in a (SAUMYA) hot press, at 30 tons/cm² and 170°C for 10 min. At the end, it was allowed to cool to room temperature by cooling water passed through the press under the same pressure.

Composite Code	
	Composite formulations
vHDPE	Virgin high density polyethylene
A1	80% HDPE + 20% RS
A2	77% HDPE + 20% RS + 3% CA
A3	76.5% HDPE + 20% RS + 3% CA + 0.5% UV
B1	65% HDPE + 35% RS
B2	62% HDPE + 35% RS + 3% CA
B3	61.5% HDPE + 35% RS + 3% CA + 0.5% UV
C1	50% HDPE + 50% RS

Table (1). Composite formulations of the RS-HDPE composites.

C2	47% HDPE + 50% RS + 3% CA
C3	46.5% HDPE + 50% RS + 3% CA + 0.5% UV

RS :- Rice Straw Fiber vHDPE :- Virgin High Density Polyethylene

CA :- Coupling Agent (PE wax) UV :- Ultra Violet Stabilizer (chimasorb)

The RS/HDPE composites formulations in (Table 1). The RS/HDPE panels were firstly conditioned at $23\pm2^{\circ}$ C and RH of $50\pm5\%$ for at least 40 hr according to ASTM D618-99 before the tests.

2.3. Water absorption and thickness swelling

Water absorption and thickness swelling tests were conducted accordance with ASTM D-570, the specimens were immersed in water for 2 hr and 24 hr, respectively, at $23\pm1^{\circ}$ C. The weight gain and thickness increase were measured 20 minutes after the samples were removed from the water. For the 24 hr water immersion tests, all specimen were oven-dried at 105°C for 24 hr to obtain the oven-dry mass for the calculation of the panel water absorption using the following equation:

Waterabsorption =
$$\frac{(m_t - m_o)}{m_o} \times 100\%$$

where, m_o and m_t are the oven-dry mass and the mass after time in water, respectively. In the water immersion tests, thickness of each composite sample was also measured for determination of the thickness swelling (TS) using the following equation:

$$TS(\%) = \frac{(h_t - h_o)}{h_o} \times 100\%$$

in which h_{o} and h_{t} are the panel thickness before and after the water immersion, respectively.

Water absorption and thickness swelling tests were conducted for three replicate.

2.4. Mechanical Properties

2.4.1. Tensile Properties

The tensile tests were conducted for the different formulations according to ASTM D 638, with a Universal Testing Machine (Instron Co.). The tests were performed at a crosshead speed of 5 mm/min and with 5 kN load cell. Samples in the form of dumpell with dimensions of $165 \text{mm} \times 19 \text{mm} \times 3.0 \text{mm}$ (thickness).

2.4.2. Izod impact test

The impact test was conducted according to ASTM D 256 on an impact tester MT220 (Kerbschl AGPRUFER) at room temperature. The impact test specimens with dimensions of 63.5 mm x 12.5 mm x 3.0mm (thickness).

3. Results and Discussion

3.1. Water absorption and thickness swelling

3.1.1. Water absorption

The result of water absorption for 2 hr and 24 hr water immersion tests are presented in (Figure.^{\(\)}).The pure HDPE sample exhibited lower water absorption compared with the all RS/HDPE composite formulations. All the WPC samples had very low percentage of water absorption after 2 hr and 24 hr water immersion tests. With the increase in RS content, there are more water-residence sites thus more water was absorbed. Furthermore, the addition of PE wax significantly reduced the water absorption of the composites in the same composite formulation. We suggest that an interaction between the RS and HDPE in the presence of PE wax (coupling agent), according to less water absorbed than those of non-coupled samples. The influence of UV stabilizer addition has no important effect in the water absorption tests.

According to Stark [25], less coating by the polymer occurs with WPC having higher wood fine content, wood easily expose on the surface of the WPC. Therefore, higher amount of water absorption is observed.

According to Stokke and Gardner [26], water absorption in composites is mainly due to the presence of lumens, fine pores and hydrogen bonding sites in the wood flour, the gaps and flaws at the interfaces, and the micro-cracks in the matrix formed during the compounding process. The presence of hydroxyl and other polar groups in various constituents of the wood flour resulted in poor compatibility between the hydrophilic wood flour and the hydrophobic polyolefin and thus weakened the interfacial bonding.



Fig.(^{*}) Water absorption of HDPE/RS composites after 2 hr and 24 hr 3.1.2. Thickness swelling

Thickness swelling of the RS-HDPE composites increased with the water absorption and thus showed a similar trend, the impact of RS to polyolefin ratio, coupling agent and UV stabilizer are shown in (Fig. 3). The pure HDPE sample exhibited lower thickness swelling compared with the all RS/HDPE composite formulations. With the increase in RS content, the thickness swelling increased. Furthermore, the addition of PE wax significantly reduced the thickness swelling. It was observed that the influence of UV stabilizer addition has no important effect in thickness swelling tests.

According to Matuana LM et al., [27], with the addition of coupling agent the interface bonding between wood flour and HDPE was improved because the anhydride moieties in MAPP entered into an esterification reaction with the surface hydroxyl groups of wood flour, this lowered the water absorption sites and reduced the water absorption by MAPP coupled composites. The thickness swelling had the linear relationship with the water absorption and changes attributed due to the similar mechanism as that of water absorption.





3.2. Mechanical properties

3.2.1. Tensile properties

The results of the tensile tests for all composite formulations for control samples, after water immersion and after weathering are given in (Figure. 4 and 5). The tensile strength are presented in (Figure. 4), the HDPE sample exhibited higher tensile strength compared with the all RS/HDPE composite formulations. The tensile strength decreased with increasing the RS content in all composite formulations. The addition of PE wax significantly increased the tensile strength, this is due to the bonding between the RS and HDPE. The addition of UV stabilizer has no significant effect.

The improvement with the PE wax addition are believed to be due to the formation of ester bonds between the(C=O) group of the oxidized PE (PE wax) and hydroxyl groups of the RS fibers. This hypothesis is confirmed by previous studies showed the (C=O) group of functionalized polyolefin coupling agents entered into an esterification reaction with the surface hydroxyl groups of RS fiber [19,20].



Fig.(4) Tensile strength of HDPE/RS composites

(Figure. 5), show the young's modulus (MOE), the pure HDPE sample exhibited lower MOE compared with the all RS/HDPE composite formulations. The increase of RS content has no significant change on MOE. The addition of PE wax increase the MOE of the composites. The addition of UV stabilizer has no significant effect on MOE. For all composite formulations the MOE are identical, however a sharp decrease was noticed for the samples of 50%RS without PE wax addition.



Fig.(5) Tensile modules of HDPE/RS composites.

According to Lu JZet al., [28] and Balsuriyaet al., [29], the observed increase in the tensile strength is attributed to the improved interfacial bonding between the wood flour and the HDPE matrix as well as the modification of individual components. According to M.A. Kalnins et al., [30], UV light and water may also act synergistically to degrade the WPCs in the following way. Exposing the WPC to UV light degraded hydrophobic lignin, leaving hydrophilic cellulose at the surface which increased the surface wettability, causing the surface to become more sensitive to moisture.

3.2.2. Izod Impact test

The results of the impact tests for all composite formulations are given in (Figure. 6). The pure HDPE give higher impact strength compared with the all RS/HDPE composites. The impact strength decreased with increasing RS content. The addition of PE wax increased the impact strength of the composites.



Fig.(6) Impact strength of HDPE/RS composites .

The improvement with the PE wax addition are believed to be due to the formation of ester bonds between the(C=O) group of the oxidized PE (PE wax) and hydroxyl groups of the RS fibers. The addition of UV stabilizer has no significant effect in the impact strength.

According to Lu JZ, et.al,[28], and Balsuriya PW, et.al,[29], the observed increase in the impact strength is attributed to the improved interfacial bonding between the wood flour and the HDPE matrix as well as the modification of individual components.

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