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# Characterization and biodegradability of polypropylene composites using agricultural residues and waste fish

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ABSTRACT

The objective of this research was to study the potential of waste agricultural residues such as rice-husk fiber (RHF), bagasse fiber (BF), and waste fish (WF) as reinforcing and biodegradable agents for thermoplastic composites. Addition of maleic anhydride grafted polypropylene (MAPP) as coupling agent was performed to promote polymer/fiber interfacial adhesion. Several composites with various polypropylene (PP) as polymer matrix, RHF, BF, WF, and MAPP contents were fabricated by melt compounding in a twin-screw extruder and then by injection molding. The resulting composites were evaluated through mechanical properties in terms of tensile, flexural, elongation at break and Izod notched impact following ASTM procedures. Biodegradability of the composites was measured using soil burial test in order to study the rates of biodegradation of the composites. In general, the addition of RHF and BF promoted an increase in the mechanical properties, except impact strength, compared with the neat PP. According to the results, WF did not have reinforcing effect on the mechanical properties, while it could considerably improve the biodegradation of the composites. It was found that the composites with high content of WF had higher degradation rate. Except impact strength, all mechanical properties were found to enhance with increase in cellulosic fiber loading In addition, mechanical properties and biodegradability of the composites made up using RHF was superior to those of the composites fabricated with BF, due to its morphological (aspect ratio) characteristics.



## 1. Introduction

Sustainability and eco-efficiency in technical applications has become quite important during the last two decades resulting from ecological concern, environmental awareness, new legislative regulations, and Greenpeace groups [1]. Petroleum-based synthetic polymers are widely used in modern society. However, the annual worldwide disposal of approximately 170 million tons of petrochemical plastics in commonly used commodities such as polyolefin in packing, bottle and molding products is a significant environmental problem, especially with the continuously increasing production and consumption of these materials (Fig. 1) [2,3]. Furthermore, plastic wastes are an undesired pollutant in soil, rivers and marine. Because of their resistance to microbial attack, they tend to accumulate in the natural environment. Although the biodegradable polymers can partially solve the problem of non-biodegradable plastic waste pollution, the majority of biodegradable polymers are not widely used because they are too expensive and the range of the material selection suitable for various end-use products is limited [4].

Wood plastic composite (WPC) is part of a new generation of reinforcing products in recent years. It composes of a thermoplastic polymer as matrix and cellulosic material which act as the reinforcing filler. There are many studies concerning the use of natural fiber as reinforcing in polymer composite systems. These reinforcing materials can be naturally degraded by microorganisms and play a significant role in degrading natural organic substances in the ecosystem [5]. Polypropylene (PP) undoubtedly has been one of the best candidates as matrix material for WPCs because of its low price, thermal stability, and widespread use in technical applications, e.g. in the automotive industry. Numerous publications can be found on the effect of addition of cellulosic fibers on the physical and mechanical properties of polymer matrix composites with focus on PP matrices [1,6,7].

Biodegradation occurs with enzymatic action and involves living microorganisms. Almost all microbial degradations are carried out by both fungi and bacteria. There are four biodegradation environments for polymeric products namely soil, aquatic, landfill and compost. Each environment contains different microorganisms and has its special conditions for degradation. In soil, fungi are mostly

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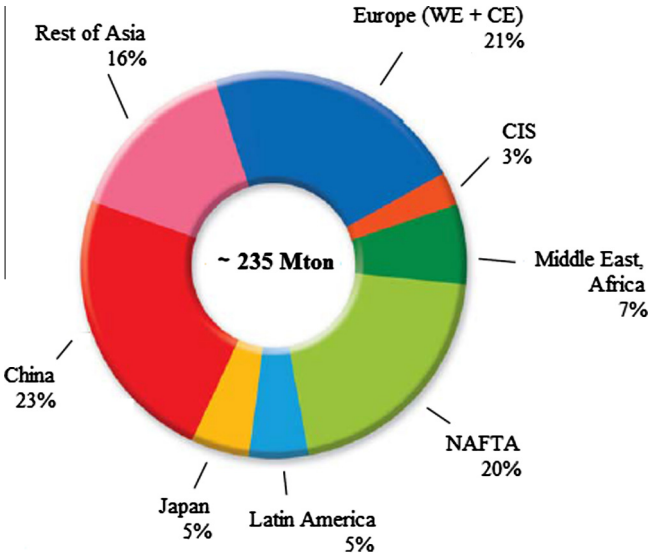


**Fig. 1.** World plastic production in 2011 [3].

responsible for degradation of organic matter including cellulosic fibers and polymers [8]. The biodegradation rate in WPCs depends on a number of factors including fiber content, the biodegradability of each component and the quality of the interface. The fiber addition generally increases the degradation rate of composites and alkaline treatment of fibers produce a slightly higher degradation rate than pure matrix [9]. In addition, the additives used (e.g. plasticizers, fillers, etc.) are important in biodegradation kinetics as well as the type of polymer reflected in molecular weight, structure and crystallinity [10]. In addition, the rate of biodegradation depends in general on the substrate composition and the existing microorganisms.

The main objective of this work was to study the potential of waste materials such as rice-husk fiber (RHF), bagasse fiber (BF), and waste fish (WF) as reinforcing and biodegradable fillers for WPCs in order to evaluate and compare their mechanical properties and biodegradability. In addition, the effects of fiber loading and WF contents on the above-mentioned properties were studied.

## 2. Methods

### 2.1. Materials

Two types of agricultural residues were investigated in this study: RHF and BF. The important chemical components and fiber morphology of lignocellulosic materials are given in Table 1. In order to reduce extractives effects, the lignocellulosic materials were treated with water at 50 °C for 48 h. RHF and BF were produced by refiner mechanical pulp (RMP) process. Consequently, the samples were oven-dried and stored in sealed plastic bags for subsequent use. The moisture content of oven-dried fiber was lower than 3%.

**Table 1**
Chemical and morphological properties of used materials.

| Properties | RHF | BF |
|---|---|---|
| Cellulose (%) | 48.9 | 55.3 |
| Lignin (%) | 19.1 | 21.0 |
| Extractives (%) | 2.5 | 2.9 |
| Ash (%) | 12.3 | 1.9 |
| Fiber length (mm) | 0.80 | 0.96 |
| Aspect ratio | 89 | 42 |

**Table 2**
Material formulations used to prepare composites.

| Specimen no. | PP (wt.%) | MAPP (wt.%) | RHF (wt.%) | BF (wt.%) | WF (wt.%) |
|---|---|---|---|---|---|
| RHF1 | 58 | 2 | 40 | 0 | 0 |
| RHF2 | 58 | 2 | 30 | 0 | 10 |
| RHF3 | 58 | 2 | 25 | 0 | 15 |
| BF1 | 58 | 2 | 0 | 40 | 0 |
| BF2 | 58 | 2 | 0 | 30 | 10 |
| BF3 | 58 | 2 | 0 | 25 | 15 |

Injection molding grade PP, with trade name V30S, was supplied by Arak Petrochemical Co. (Iran). The PP was in the form of pellets with a melt flow index of 18 g/10 min and density of 0.92 g/cm$^3$.

Maleic anhydride functionalized polypropylene (MAPP) with a molecular weight of 52,000, acid number of 9 mg KOH/g, and melting point of 158 °C was provided by Eastman Chemical Products, Inc.

Kilka (*Clupeonella engrauliformis*), obtained from Iranian waters of the Caspian Sea, was used as WF. It is the most abundant fishes in the Caspian Sea [11]. The chemical analysis of WF showed 70% protein, 7.8% fat and 6.6% moisture content.

### 2.2. Preparation of samples

Formulation of the mixes and abbreviation used for the respective mixes prepared are given in Table 2. Composites were produced in a two-stage process. In the first stage, fibers, PP, WF, and MAPP pellets were premixed by hand at various formulations, and the mixtures were then fed into a laboratory co-rotating twin-screw extruder. The temperature profile in the extruder was 165/170/175/180/185 °C and the screw speed was set at 70 rpm. In the second stage, the extrudate in the form of strands were allowed to cool to room temperature and then granulated using a CW Brabender Granulator. The resulting granules were dried at 105 °C for 24 h before being injection-molded.

### 2.3. Mechanical testing

The specimens were tested following ASTM standard D 638 for tensile properties, ASTM D 790 for flexural properties and D 256 for notched Izod impact strength (ASTM 1999). Tensile and bending tests were conducted using a Universal Testing Machine (Santam model STM-150) at a speed of 5 and 10 mm/min, respectively. Isod impact test was performed with a pendulum apparatus (Zwick model 1446) using conventional V notched specimens. Four replicates were tested for each property under each formulation.

### 2.4. Soil burial test

The soil burial test was carried out on a laboratory scale to examine the biodegradability using the method which reported by Behjat et al. [8] briefly as follows. First, rectangular sheets (10 cm × 10 cm × 1 mm) of specimens were buried in the soil by random pattern. The pot containing the soil and samples were incubated at almost constant temperature of 27 °C for four months. The moisture content was maintained at 45–60% of the soil's maximum water holding capacity. This humidity is optimal for microbial activity. In addition, the pots were covered with plastic film to avoid water evaporation from the soil surface. Biodegradation was estimated by monitoring changes in weight as a function of burial time. The samples were removed from the soil every 20 days. The debris on the specimens was removed by washing with water. The samples were then dried in an oven at 100 ± 5 °C for 24 h. After