

Composite Materials of Polypropylene and Waste Jute Fibers

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The replacement of glass fibers polymer composites with those containing polymers and cellulose fibers, friendly for environment and human health has gained a special importance in recent years. The paper presents some mechanical and thermal characteristics of composites of neat polypropylene and jute fibers from technological wastes, treated and untreated. The use of waste fibers in polypropylene led to a significant increase of the elastic modulus and tensile strength which highlights the reinforcing effect of the waste fibers, the effect being more important in case of jute fibers treated with aminosilane.

Keywords: polypropylene, composites, jute, mechanical properties

The replacement of glass fibers polymer composites with composites of polymers and cellulose fibers, friendly for the environment and human health has gained a special importance in recent years [1 - 20], the abundance of raw materials being a major advantage of the employment of cellulose fillers in industrial applications. This fact is additionally stimulated by the increasingly pressure on industry to use clean technologies. Moreover, in the last 20 years, natural fibers have attracted the attention of researchers and technologists due to environmental benefits, their low price, low density and very good specific mechanical properties (by weight) [1 - 4]. Apart of these, biodegradability, small abrasivity and low emission of carbon dioxide are added.

The main constituents of vegetable fibers are cellulose (60...80%), hemicellulose (12...23%) and lignin (1...20%). Vegetable fiber characteristics vary according to the amount of these three main components, the plant part from which they were extracted, planting places, plant age and other reasons. The main criterion in choosing the type of plant fibers to reinforce polymers is the level of mechanical characteristics, directly related to the degree of polymerization of cellulose. Other criteria are humidity, usually between 8 and 13%, or content of wax which covers them and which influences the fiber wetting by the polymer in the composite material [1 - 2].

Recent works in the field of biodegradable materials concern the employment of cellulose fibers issued from plants [2, 4, 6, 21 - 27]. Thus, thermoplastic polymer composites with long vegetable fibers showing anisotropic properties were tested in the automobile industry, the main funder support of the research in the field of natural fiber composites. Increasing the length of natural fibers, for example the flax ones, from 6 to 12 mm, resulted in a 20% improving of tensile strength and 100% rising of impact strength of polypropylene/20% flax composite [7]. Vegetable fibers have been also tested as reinforcements in biodegradable matrices, e.g. cellulose fibers in starch, leading to improved mechanical properties as well as increase of transition temperature and decrease of water sensitivity [28].

Cellulose fibers resulting from textile, wood or agricultural wastes, could also become reinforcements for polymers, the preparation of composites from thermoplastic polymers and waste vegetable fibers allowing the recovery, in important technical applications, of the large amount of technological wastes accumulating in the

production processes of textiles, prefabricated wood or agricultural products. Thus, the flax fibers wastes from machining, carding and spinning were tested as reinforcements in low density polyethylene and recovered polyethylene, resulting in an improvement of flexural elastic modulus, a decrease of elongation at break with the maintenance of mechanical resistance between the same limits [29].

In order to be used as reinforcements in polymers, textile wood or agricultural wastes must be brought in a form easy to handle and well dispersed in polymers. The difficulty to disperse textile wastes in polymers is due to several reasons including their aspect, strong hydrophilic character (because of the high cellulose content which makes them incompatible with synthetic polymers - hydrophobic materials), the difference in bulk density of the main components of the composite material (polymer pellets and waste fibers), low wetting capacity of fibers by the polymer melt and poor adhesion at the interface in the composite material. It is therefore necessary to a priori use of special treatments in order to increase the fibers compatibility to polymer matrix, to be really effective in improving mechanical properties of polymers.

Beside mechanical characterization, thermogravimetric analysis (TGA) is a useful tool for highlighting the thermal stability of polymers after cellulosic filler adding.

In this paper, some results concerning the laboratory and pilot research on the preparation, as well as mechanical and thermal characterization of polypropylene composites with natural jute fibers issued from technological wastes are presented.

Experimental part

Materials

A polypropylene homopolymer Moplen HP 500 N (PP), characterized by melt flow index 12 g/10 min (230°C/2.16 kg), tensile strength, 35 MPa, Charpy impact strength (unnotched specimens), 110 kJ/m², was used as a polymer matrix. A maleinized polypropylene Polybond 3200 (MA-PP) 2.2% maleic anhydride, MFI = 115 g/10 min, d = 0.91g/cm³ was used as compatibilizing agent. As fillers were used jute fibers (I), technological wastes from textile factories, from spoolers, have undergone the following treatments:

-yarn washing to remove traces of dust, grease or chemicals by using an alcoholic solution followed by repeated water rinses;

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Table 1
EXTRUSION PARAMETERS FOR PP/JUTE FIBERS COMPOSITE PREPARATION

Extrusion parameters at Leistritz	Values
Cylinder temperature (zones), °C	190; 200;215; 195; 200; 200; 200; 200; 200
Nozzle temperature, °C	200
Screw rotation speed, min ⁻¹	50
Temperature (in material), °C	190

- drying in a circulating air oven at a temperature of 60 - 70 °C for about 3 h and then in a vacuum oven;
- cutting at a size of fibers of 3 ... 6 mm.

Chemical treatment was applied to some of these fibers for surface hydrophobization, by suspending the fibers in an alcoholic solution containing a silane coupling agent (3-aminopropyl-trietoxy silane), stirring at room temperature and then at high temperature (100...120° C) for at least 2 h, followed by the removal of traces of water and alcohol by drying in a circulating air oven and then in a vacuum oven.

Polymer composites preparation

Two methods were applied to obtain the composite material, namely:

- melt mixing of polypropylene, MA-PP and jute fibers in variable concentration (10, 20, and 30% percent by mass) in a mixing chamber at high temperature followed by two-roll mill forming and plate pressing (process 1);

- mixing the polymer with MA-PP in an intensive mixer followed by feeding this mixture and treated fibers (20% compared to the polymer) in a double screw extruder (process 2).

In case of the first process, a Brabender Plastograph, chamber 50 cm³, was used for melt mixing in the following conditions: temperature 180°C, 75 rpm rotor speed and mixing time - 12 min. The mixtures were pressed at a temperature of 190°C for 10 min, from the resulted plates of composite material, specimens for mechanical characterization being prepared.

In the second process, for mixing polymer matrix with jute fibers, a double-screw extruder - Leistritz 30...34 was used, the working conditions being presented in table 1.

The samples used for determining the mechanical characteristics were prepared by injection on an Engel - 40/25 injection machine at a nozzle temperature of 200°C and a pressure of 1400 kgf/cm².

Characterization of polymer composites

Mechanical characterization

Composites obtained according to the first process (1) were characterized by determining the tensile mechanical properties, according to SR EN ISO 527:2000 on 2 mm thick specimens type IB, obtained by pressing - embossing, with a speed of 50 mm/min in case of the tensile strength determination and 2 mm/min to determine the elastic modulus. A Zwick 1454 testing machine coupled to a PC was used. Shore D hardness was determined according to SR ISO 868:1995 using a Zwick penetrometer on 3 mm thick specimens.

Composite materials obtained by the second process (2) (injection molded specimens 4 mm thick) were characterized by measuring the above listed characteristics. In addition, the flexure strength and elastic modulus according to SR EN ISO 178:2003, speed 50 mm/min on specimens of IB type and Izod impact strength on notched specimens according to SR EN ISO 180:2001, were determined.

Thermal characterization

Thermal characterization of composites was performed by using a Du Pont TGA 2000 analyzer, from 20 to 700°C at a heating rate of 20°C/min in air.

Results and discussion

The fibers were visualized with an optical microscope, which allowed revealing the diameter of the fibers and their aspect ratio. Figure 1 shows the image obtained with the optical microscope (transmission - dark field) of a sample of jute fibers treated according to the methodology specified above, resulting a diameter of 10.....40 μm and an aspect ratio between 20 and 50.

The results of the mechanical characterization of polymer composites obtained in the case of the first process are shown in figure 2.

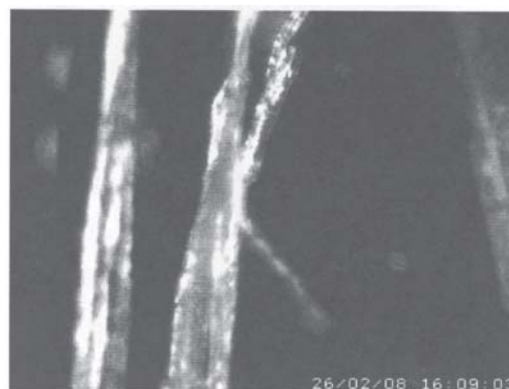


Fig. 1 Image obtained at optical microscope for jute fibers (x120)

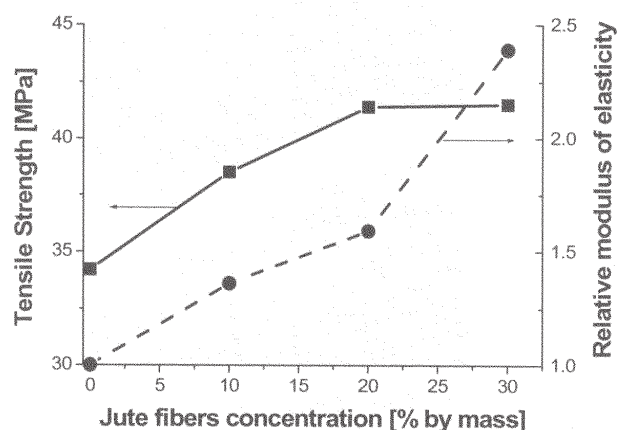


Fig. 2 Mechanical properties of composites PP/jute fibers wastes obtained by process 1 depending on the concentration of fibers

To highlight more clearly the reinforcing effect, the tensile elastic modulus was given as a relative modulus, defined by means of the ratio between the composite tensile elastic modulus and that of the polymer matrix.

Figure 2 shows that both tensile strength and elastic modulus increase with increasing amount of jute fibers wastes (at a concentration of 30% jute fibers a value of

Table 2
RESULTS OF MECHANICAL CHARACTERIZATION OF PP COMPOSITES
WITH TREATED AND UNTREATED WASTES FIBERS

Composite	Tensile strength MPa	Elongation at break %	Relative modulus of elasticity MPa	Hardness Shore D ° Sh
PP	34.2	20	1,00	66
PP/20%I	41,4	10	1,59	70
PP/20%I _t	41,5	14	1.85	70

Table 3
RESULTS OF MECHANICAL CHARACTERIZATION OF PP/WASTES FIBERS COMPOSITES
OBTAINED BY PROCESS 1 AND 2

Mechanical characteristics	PP Process 2	PP/20% jute Process 2	PP Process 1	PP/20% jute Process 1
Tensile strength at break, MPa	33,1	46,2	34,2	41,4
Elongation at break, %	30	16	20	10
Tensile modulus of elasticity, GPa	1,29	2.01	1,29	2,06
IZOD Impact strength, kJ/m ²	1,4	1,7	-	-
Hardness Shore D, ° Sh	64	69	66	70
Flexure strength, MPa	45,0	54,2	-	-
Flexure modulus of elasticity, GPa	1,80	3,49	-	-

almost 2.5 times higher than that of polypropylene matrix being reached).

The mechanical properties of composites containing 20% jute treated fibers (I) and, respectively, untreated (I), are presented in table 2 and compared with the polypropylene matrix. The elongation at break decreases strongly along with the introduction of waste cellulose fibers while the tensile strength, elastic modulus and Shore D hardness increase significantly. The values of tensile strength are similar for the two types of composites, with treated and untreated fibers, but the elastic modulus and the elongation at break are higher for silane treated fibers composites, pointing out an increased reinforcing effect of PP matrix and, consequently, the efficiency of the applied treatment.

Results of mechanical characterization of PP/20% waste jute composite – second process are presented in table 3, and compared with the values obtained for polypropylene and similar composite prepared by melt mixing - pressing.

Several aspects can be observed:

- for tensile strength and, especially, for tensile elastic modulus close values are obtained (almost identical in the case of modulus) for samples made by extrusion-injection as compared with samples obtained by specific laboratory technology;

- elongation at break decreases almost by half for both processes (1 and 2) after the addition of the waste fibers, while the tensile strength increases by 40% for samples

obtained by process 2 as compared to only 20% for those obtained by process 1;

- the increase of hardness when waste fibers are added is similar for both types of samples, i.e. 4...5 °Sh;

- the impact and flexure strength of the composite material are about 20% higher than for polypropylene;

- flexure elastic modulus of the composite material containing waste jute fibers is about 2 times higher than that of polypropylene obtained in the same conditions, by extrusion-injection.

One can remark that the composite material obtained on the pilot equipment (second process) under controlled conditions, resembling to those from industrial facilities, have similar levels of mechanical characteristics compared with the composite material made according to the same formulation but in the laboratory facility (or similar increases over control). All this results show an appreciable improvement of mechanical properties, sometimes spectacular, as for flexure modulus, due to waste fibers adding in polypropylene.

TGA diagrams of composite materials (process 1) of polypropylene and jute waste fiber (20% and 30%) are shown in figure 3, while table 4 displays the main thermal characteristics resulting from TGA diagrams: T1 - the onset temperature of degradation, T2 - temperature at which degradation rate is maximum and T3 – the end temperature of degradation, p% - the mass percentage of degraded material at the end of decomposition.

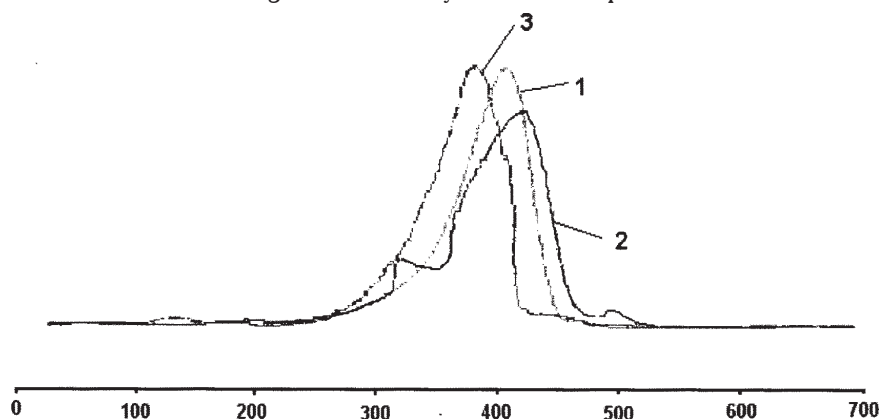


Fig. 3 TGA diagrams of composit materials:
1 – PP; 2 – PP/20%waste jute fibers; 3 - PP/30%
waste jute fibers

Table 4
THERMAL CHARACTERISTICS OF PP/WASTES FIBERS COMPOSITES

Composite	T1 °C	T2 °C	T3 °C	p %
PP	253.50	403.85	440	98.24
PP/20% jute	252.40	412.26	443	94.76
PP/30% jute	235.58	375.24	410	94.16
PP/20% treated jute	226.57	366.24	395	93.47

One can see that waste jute fibers placed in concentration of 20% in polypropylene do not cause significant changes in thermal stability of polypropylene, the degradation taking place in the same temperature range 250 - 440°C. However, an additional peak appears (315°C) due to the degradation processes of jute fibers, occurring with a maximum intensity at 310 - 325°C [2]. Increasing the concentration of waste jute fibers to 30% causes a visible decrease of thermal stability, onset temperature degradation decreasing significantly (235°C instead of 253°C).

The shift of the degradation range of composite with 30% fibers from 250 - 440°C to 230 - 410°C makes invisible the peak due to cellulose degradation in this composite.

Composite with 20% treated jute fibers exhibits a weaker thermal stability than the similar composite with untreated fibers, the thermal stability being even weaker than that for the composite with 30% fibers concentration. This decrease of degradation temperature may be due to the influence of silane treatment process that took place at 120°C for 2 h, leading, in addition to cellulose - silane linking, to chemical changes of cellulose which negatively impacted on the thermal stability of polypropylene.

Conclusions

Polymer composites from polypropylene and jute fibers wastes in various concentrations, treated and untreated, were obtained by two processes. The introduction of waste fibers in polypropylene leads to a significant increase of the tensile elastic modulus and strength (similar values for both methods applied). This highlights the reinforcing effect of waste fibers, the effect being more important in case of jute fibers treated with aminosilane. Furthermore, the thermal stability of composites with up to 20% jute fiber waste was similar to that of the polypropylene matrix, but at a higher concentration of fibers (30%) a weaker thermal stability as compared with natural polypropylene was observed.

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