Recycling of Commingled Plastics Waste Containing Polypropylene, Polyethylene, and Paper

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ABSTRACT: Polymer waste recycling is a major technical problem, because large amounts of synthetic polymers are produced every day and polymeric wastes are gathered from municipal solid wastes. There are a few polyolefins, such as polyethylene (PE) and polypropylene (PP) with huge amounts of paper in the waste materials. In order to recycle the commingled plastics waste that contains paper, hydrolytic treatment is needed prior to conventional processing. In this project, the optimum conditions of hydrolytic treatment of paper and the mechanical properties and morphological state of different compositions of PP high-density PE (HDPE) blends with paper were studied. Ethylene-propylene-diene copolymer (EPDM) was added to improve the mechanical properties of blends. The results show that the hydrolytic treatment of paper improves the mechanical properties, such as the tensile strength and modulus of the PP/HDPE/paper composites relative to the untreated samples, and up to 30% paper can be added to commingled PP and HDPE blends. The EPDM was used as an impact modifier. The plastics waste containing paper can be used in applications such as artificial wood. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 80: 2573–2577, 2001

Key words: plastic waste; recycling; blend; polypropylene; polyethylene; paper

INTRODUCTION

The present study is part of a project on recycling of commingled plastics containing polypropylene (PP), polyethylene (PE), poly(vinyl chloride), and paper. Generally, reprocessing of plastics waste that are contaminated with more than 5% paper by conventional plastics processing machinery is difficult and becomes almost impossible at paper levels exceeding 15%.[1] But on the basis of these studies, it can be concluded that the process of commingling plastics waste containing 30–40% paper is feasible.[2] The method used is based on a hydrolytic treatment of the plastics waste containing paper prior to conventional processing. During this treatment the contaminating paper component undergoes embrittlement and easily disintegrates into smaller fragments when subjected to the shear forces acting in the plastics processing machines.[3,4] However, the plastic materials are not influenced by hydrolytic treatments. Thus, there is no need for separation of paper and plastics fractions before processing. The homogeneity of the cellulose fiber filled plastic composites is improved and therefore enhances the mechanical properties.[5]

Generally, the recycling process consists of six stages: waste collection, separation, grinding, cleaning and washing, drying, and processing. These stages are dependent on the economic conditions and technical availabilities of different countries.[6] In this article we focus on the virgin

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Table I Mechanical Properties of 90/10 (w/w) HDPE/Paper Hydrolyzed in Reflux Condition for 1 h in Acidic Solution

Treatment Condition	Modulus (MPa)	Tensile Strength (MPa)	Elongation at Break (%)	Impact Strength (J/m)
Untreated	820	20	12	84.3
Treated with HCl 10% (v/v)	647	21	14	110.5
H_2SO_4 10% (v/v)[a]				
HCOOH 10% (v/v)[b]				
HCl 5% (v/v)[c]				
HCl 10% (v/v)[d]				

[a] This acid could not hydrolyze paper entirely in 1 h.
[b] This acid could not hydrolyze paper entirely in 1 h.
[c] The complete hydrolysis time is about 3 times that of 10% acid.
[d] The complete hydrolysis time is about 5 times that of 10% acid.

materials and we will continue this project with waste materials.

EXPERIMENTAL

Materials

High-density PE (HDPE, GF 4760, MFI = 21 g/10 min, d = 0.954 g/mL), PP (X30S, MFI = 9 g/10 min, d = 0.9 g/mL, Arak Petrochemical) and ethylene-propylene-diene copolymer (EPDM) (Vistalon 7500) were used in this study. HCl and H_2SO_4 were commercial products and HCOOH was obtained from the Merck Co.

Testing

The samples were prepared for testing by mixing them in a Haake Buchler Rheomix 750 internal mixer and then a compression molding technique was used for preparation of testing bars. Tensile testing was performed on an Instron 6025 tensile testing machine according to ASTM D638. The Izod impact property was determined using an instrumented impact tester (Zwick 5102) according to ASTM D256, and the results were obtained from at least a five-sample test. For investigating the morphological state of the various blends a Cambridge S360 Scanning electron microscope (SEM) was used.

Hydrolytic Treatment and Blend Preparation

For paper hydrolysis, two inorganic and an organic acid (HCl, H_2SO_4, and HCOOH) with different concentrations were prepared (Table I). In this method HDPE granules with 10% (w/w) paper were mixed and boiled for about 1 h in reflux conditions. Then the hydrolyzed product was separated from the acid solution and washed with tap water until neutral. After drying at 70°C for 12 h, the HDPE and hydrolyzed paper were mixed in a Haake mixer at 160°C for 7 min.

The mechanical properties of the samples were determined. The composition of the samples and the mechanical properties of the blends are listed in Table II. Then paper (10–40%, w/w) was added to the blends with optimum mechanical properties. These samples were subjected to hydrolysis conditions and the structure and mechanical properties were determined. EPDM (2–10%, w/w) was added to improve the impact strength of these blends, and again the mechanical properties were determined.

RESULTS AND DISCUSSION

Table I shows the effect of various acids and their concentrations on the hydrolysis time and me-

Table II Composition and Mechanical Properties of Blends

PP/PE (wt %)	Tensile Strength (MPa)	Impact Strength (J/m)	Modulus (MPa)	Elongation at Break (%)
0/100	15	389	650	120
10/90	16	94	700	45
20/80	18	20	800	11
40/60	23	21	850	7.5
60/40	24	12	1000	4.5
70/30	26	15	1010	7
80/20	27	35	1050	11
100/0	30	22	1200	13

Table III Mechanical Properties of Blends with Paper

Blends of PP/HDPE/ Paper (wt %)	Modulus (MPa)	Tensile Strength (MPa)	Elongation at Break (%)	Impact Strength (J/m)
10/90/0	700	15.5	45	94.0
9.5/85.5/5	950	18	15	70
8/72/20	940	17.5	5	60
7/63/30	1100	17.6	3	59
6/54/40	1200	17.7	3	59
80/20/0	1050	27	12	35
76/19/5	1450	26	10.5	22
64/16/20	1470	27	5	20
56/14/30	1800	28	3	19
48/12/40	1805	29	3	19

chanical properties of treated and untreated samples. Two methods were used: hydrolysis in suspension and hydrolysis after impregnation with an acid solution. In the first method the materials were suspended in an acid solution and the mixture was boiled for about 1 h (acid conc $\geq$ 10%, v/v). Generally, higher temperatures allowed the use of a lower concentration of acids. In the second method the materials were impregnated in an acid solution for 1 week (acid concn = 5–10%, v/v) and then they were boiled for 20 min. In our continuing studies we adopted the second method.

Table I shows untreated samples having lower impact strength and elongation at break than the treated samples. Hydrolysis with HCl 10% (v/v) had better results than other acids; thus, HCl was

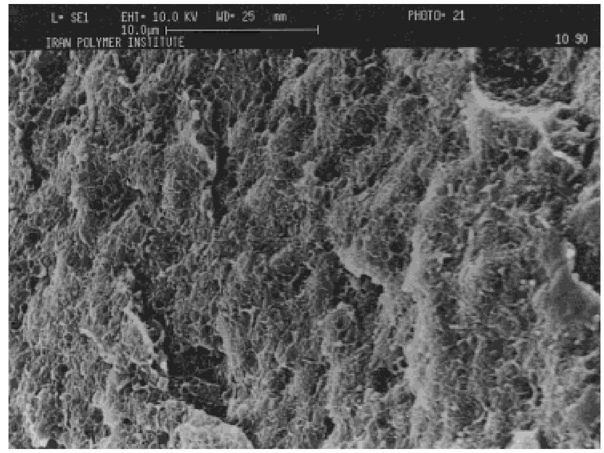

Figure 2 A micrograph of a 10/90 PP/HDPE blend.

chosen for the hydrolytic treatment. After hydrolysis cellulose undergoes embrittlement and easily breaks into small fragments when fed with plastics into a compounding machine. Thus, at a melting temperature of about 170°C, a reasonable homogeneity of the materials was obtained with enhanced mechanical properties.

Table II shows the composition of various blends of PP/HDPE prepared in this work with their mechanical properties. The presence of a maximum in the modulus and the value of the tensile strength with an 80/20 PP/HDPE (wt %) blend, together with the fact that the 10/90 PP/HDPE blend exhibits a considerably higher elongation at break than others, suggests that at low HDPE content the PE acts as a stiffener for the PP. This is probably because of the high crystallinity of HDPE. At high HDPE content the PP would plasticize the HDPE, making it more duc-

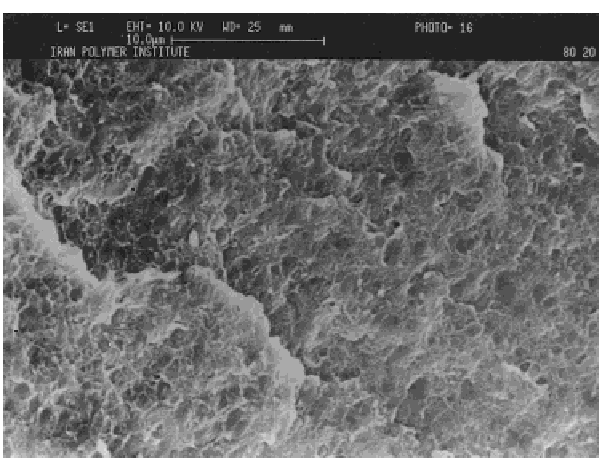

Figure 1 A micrograph of an 80/20 PP/HDPE blend.

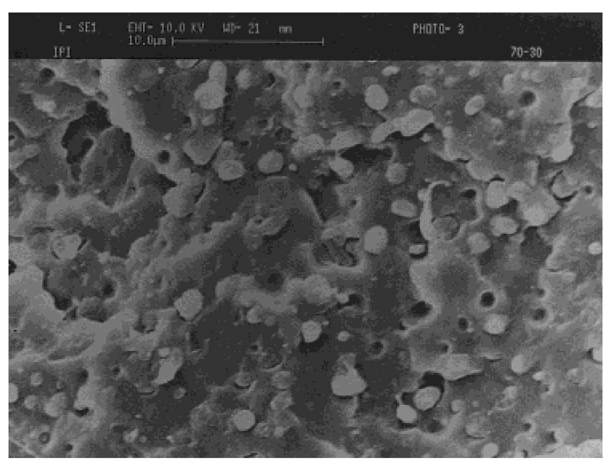

Figure 3 A micrograph of a 70/30 PP/HDPE blend.

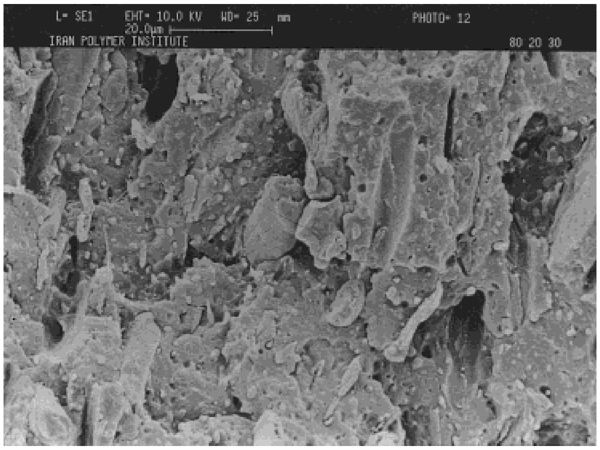

Figure 4 A micrograph of an 80/20 PP/HDPE blend with 30% (w/w) paper.

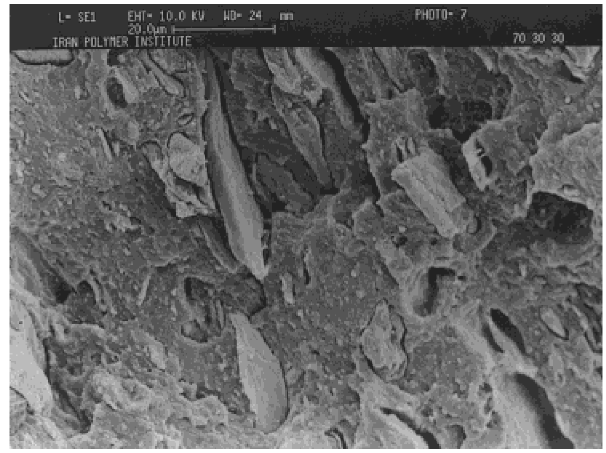

Figure 6 A micrograph of a 70/30 PP/HDPE blend with 30% (w/w) paper.

tile.[7] When adding up to 40% paper to the PP/HDPE 80/20 and 10/90 blends, the presence of cellulose fiber increases the tensile strength and modulus of the PP/HDPE/paper composites. But the impact strength and elongation at break decrease, because in this case the paper acts as a filler and reduces the consistency of the matrix, therefore, decreasing the mechanical properties (Table III).[1]

The SEM micrographs of the PP/HDPE blends (10/90, 80/20, 70/30) show that the interfacial adhesion and therefore the compatibility of the phases and mixing in the 10/90 and 80/20 blends are better than the other compositions such as the 70/30 blend (Figs. 1–3), and these phenomena are confirmed by the mechanical properties (Table II). The micrographs of 10/90, 80/20, and 70/30 PP/HDPE blends with 30% (w/w) paper (Figs. 4–6) show that the wetting of the matrix with paper causes the adhesion and distribution and therefore the mechanical properties in the 10/90 and especially in the 80/20 blends to be better than the other blends (e.g., 70/30) and so helps to better mix the paper with the blends. In practice 30% (w/w) paper is more suitable than 40% (w/w) paper.

EPDM (2–10%, w/w) was added to improve the mechanical properties of 80/20 and 10/90 PP/HDPE blends with 30% (w/w) paper. Figures 7

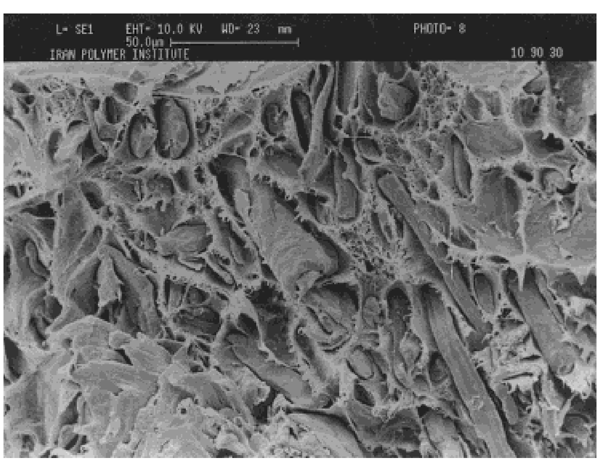

Figure 5 A micrograph of a 10/90 PP/HDPE blend with 30% (w/w) paper.

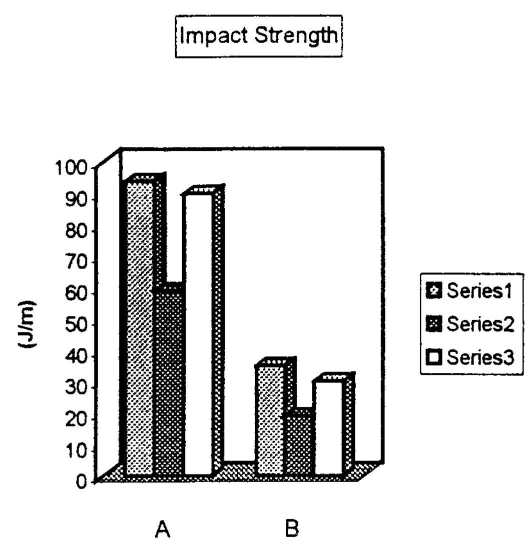

Figure 7 A comparison of the impact strength of PP/HDPE (10/90 and 80/20, series 1), PP/HDPE/paper (7/63/30 and 56/14/30, series 2), and PP/HDPE/EPDM/paper (6/54/10/30 and 48/12/10/30, series 3) blends in A and B groups.

and 8 show that the impact strength and elongation at break of the 10/90 and 80/20 PP/HDPE blends with 30% (w/w) paper and 10% (w/w) EPDM are improved, but the tensile strength is decreased (Fig. 9). In this case EPDM acts as a compatibilizer and impact modifier for the PP/HDPE blends and helps the compatibilization of blends of PP and HDPE. Because at the end the usage of these blends as artificial wood is considered, the improvement of the impact strength is therefore more important than the decreasing of the tensile strength. Thus, the blends 80/20 and 10/90 PP/HDPE with 30% (w/w) paper and 10% (w/w) EPDM can be used as artificial wood.

CONCLUSION

Hydrolysis treatment of paper improves the homogeneity and mechanical properties of the PP/HDPE/paper composites. The blends with 10/90 and 80/20 PP/HDPE ratios have the best mechanical properties, so we can modify the compositions of the municipal solid waste to these ratios and then add about 30% paper into it and improve the mechanical properties (especially the impact strength) with EPDM. The final product can be used as artificial wood.

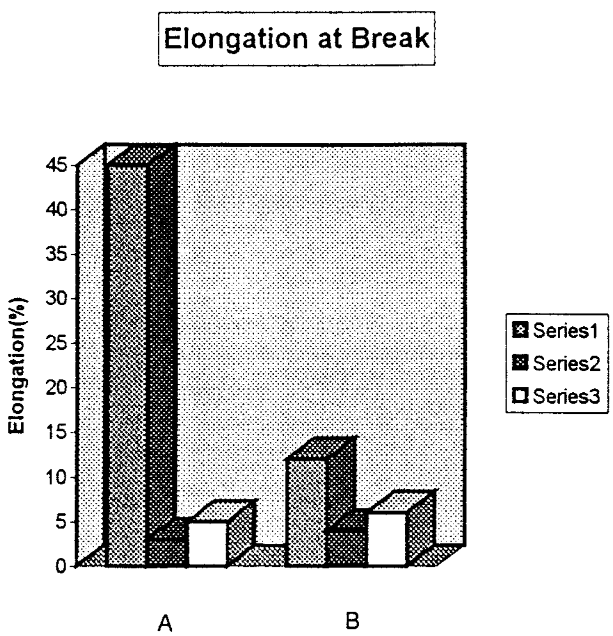

Figure 8 A comparison of the elongation at break of the samples described in Figure 7.

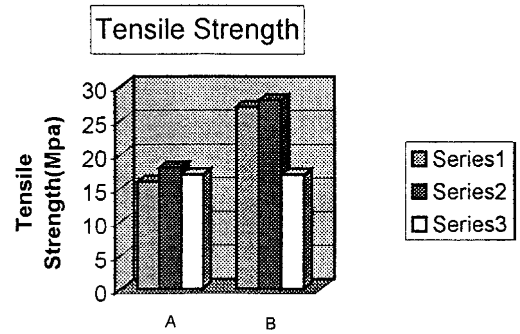

Figure 9 A comparison of the tensile strength of the samples described in Figure 7.

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