Recycled Injection-Molded and Fibre-Reinforced Polyethylene / Polypropylene

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Abstract: In this presentation, we are concerned with the fabrication and characterization of samples of compression-molded and injection-molded recycled polyethylene, with, in some cases, addition of cellulose fibre. We wish to know what is the performance of such plastics, as compared to the virgin materials and what is the effect of added fibre in both molding modes.

INTRODUCTION

Recycling of plastics is growing in Canada and Quebec, as well as the other materials recycling. Data of Basset (Ref. 1) show the proportion of plastics in the waste stream from household waste: this proportion of plastics amounts to a few per cent and is not large, especially on a weight basis. Paper and other cellulosics are by far the main components. Additionally, since blue-box programs have been introduced in large urban centers, there has been a levelling off of the production. In Quebec, the production of recycled plastics is still growing. Table 1 gives values for recycled polymer material as to its origin: postconsumer (blue box), industrial and consigned (refundable containers) (Ref. 2,3). This table does not take into account the material recycled inside plants. Thus the ratio between recycled polymers of industrial and consumer (postconsumer and consigned) origin is approximately 2 to 1. At present, the volume of postconsumer recycled plastics has probably topped 10 000 tonnes. Since the consumption of such plastics is about 145 000 tonnes in Quebec, that would place the recycling rate at about 7 %, which is quite similar to values quoted for western Europe (Ref. 4,5).

Table 2 gives the nature of recycled polymers for 1994. PE stands out as expected, but the volumes for PET, PVC and PP are fairly respectable, too.

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Precedent work (Ref. 6-8) has shown that recycled HDPE had very satisfactory properties, even when mixed with reinforced fibre. Overall, there is not much data in the literature on recycled polymers and even less on polymer blends.

Table 1. Recycled plastics in Quebec (10^3 tonnes) [a]

	1992	1993	1994
Postconsumer	4.0	4.0	7.2
Industrial, commercial	23.0	28.0	25.6
Consigned	6.0	6.1	8.1
Total	33.0	38.1	40.9

[a] Adapted from Ref. 2.

Table 2. Recycled polymers in Quebec (10^3 tonnes) in 1994 [a]

	Industrial and commercial	Postconsumer	Total
LDPE	13 960	600	14 560
HDPE	7 320	1 880	9 200
PET	380	5 010	5 390
PVC	6 170	70	6 240
PP	8 840	2 890	11 730
PS	720	180	900
ABS	350	0	350
Other	520	210	730
Total	38 260	10 840	49 100

[a] Adapted from Ref. 2.

As will be seen later in this paper, the PE used here does contain ca 5-6 % PP. There is phase separation in model studies of PE/PP blends (Ref. 9) although mechanical properties cited are quite reasonable. PE/PP blends exist on the market, with, however, PP being the continuous phase (Ref. 10). In fact, some of the recycled PE we have tested can contain up to ca 40 % PP.

EXPERIMENTAL PART

Material

Recycled PE was from sorted household waste (from Transplastec, St-Bruno, Quebec) with ca 5 % PP, with a density of 0.941 g/cm^3, with no discernible odor. Thus it could be classed as HDPE, since medium density PE is between 0. 926 and 0.940 g/cm^3. Details of recycling procedure are as follows (Ref. 11): in the recycling plant, about 5 tonnes of recycled materials is treated per batch. Visual sorting eliminates plastic film and containers contaminated with grease or oils. Tissues and non-plastic material (~36 %) are also eliminated, followed by crushing/cutting-up of the material to about 1-cm particles and by pneumatic separation of paper and films. The mix is then washed, air-dried, sorted per density and hot-dried. Finally, particles are extruded and granulated. Melt flow index as to ASTM D-1238 varies between 1-3. (Ref. 11).

Compression molding

Prior to compression molding, samples containing fibres were homogenized in a Rheomix (Rheocord system 40, Haakebuchler), at 170 °C. After 20 min, at 40 rpm, a constant torque was reached. All samples, with or without fibres, were molded at 107 MPa, 170 °C for 6 min.

Injection molding

Recycled polymer was dried and, if needed, mixed with dried CTMP wood fibre in a Brabender, cut up and dried again. It was injection-molded in a Nissei 62-tonne press at ca 177 °C, with 20-s injection time in a mold for ASTM D-638 dumbbell type samples.

Testing conditions

Compression-molded samples had a section of 4.4 mm^2 while injection-molded ones were of the ASTM D-638 type. All (six samples per experimental condition) were tested for tensile properties with an INSTRON at 5 mm/min loading rate (Ref. 11,12).

Differential scanning calorimetry (DSC) was done with a Perkin - Elmer DSC-2 at 10 °C/min with 10-mg samples in aluminium pans.

RESULTS AND DISCUSSION

From DSC data of the material, as received from the recycling plant, knowing the (endothermic) heat of fusion of PE and PP (Ref. 13), the relative proportion of each polymer can be ascertained. No other peaks were present in thermogram, which would be evidence of the presence of other crystalline materials. Additional FTIR work confirmed absence of any detectable amounts of PS or PVC.

From precedent calibration of DSC with pure isotactic PP, the percentage of PP in this blend was estimated at 4.7 %., assuming the PP phase has the same degree of crystallinity as in the pure state. From the data of Avalos et al. (Ref. 10), crystallization of PP does not seem to be hindered by the presence of LDPE, although their study was done in mostly high-PP composition ranges. There is a difference, as seen later in the text, in crystallinity between compression- and injection-molded samples.

If the heat of fusion of PE is taken as 245.3 J/g (Ref. 13), then the percentage of crystallinity of the remaining 95.3 % of PE material can be calculated as 49.2 %, assuming there are no other constituents in the recycled blends. The real value is a little higher since there are some pigments (TiO_2, inks) left in the blend.

Mechanical properties

Young's modulus is 690 MPa for compression-molded recycled PE-PP vs 865 MPa for the same but injection-molded. This compares favorably with values of ca 600 MPa for extruded recycled HDPE (Ref. 7). Strength-at-yield (Table 3) is about 20-21 MPa for both processes for our product while Yam (Ref. 7) cites values of 32 MPa for extruded recycled HDPE and Vaccaro et al. (Ref. 9) cite values from 34.7 MPa for PP to 10.4 MPa for LDPE, with a continuous variation in the blend (compression-molded). Although PE/PP is considered an incompatible blend, in Vaccaro's work, the yield strength is nearly proportional to volume fraction of PP added to LDPE, without compatibilizer added. As seen in Table 3, strengths-at-yield are not much affected by either processes or by the presence of fibre.

Table 3. Mechanical properties of recycled plastic, compression- and injection-molded, with or without added fibre (average of six samples) [a]

	Young's modulus, MPa	Strength-at-yield, MPa	Elongation-at-yield, %
Compression molding			
PE	690 (47)	20.7 (0.3)	9.7 (0.5)
PE + 10% fibre	1012 (2)	22.0 (0.2)	6.0 (0.1)
Injection molding			
PE	865 (28)	21.0 (0.8)	20.7 (0.2)
PE + 10% fibre	1202(35)	22.0 (0.2)	14.8 (0.5)

[a] Ref. 11,12.

The relative increase in Young's modulus is 25 % for the recycled PE going from compression molding to injection molding. However, with 10 % added fibre in both, the relative increase is less, 19 %. The relative increase in compression-molded samples upon addition of 10 % fibre is 47 % , while in the case of injection molding, the increase in properties upon addition of 10 % fiber is a bit less, 39 %. These relative variations are illustrated in Fig. 1. Although absolute numbers are al-ways higher for injection molding with respect to compression molding, the smaller relative increases upon reinforcing fibre addition may be due to fibre attrition.

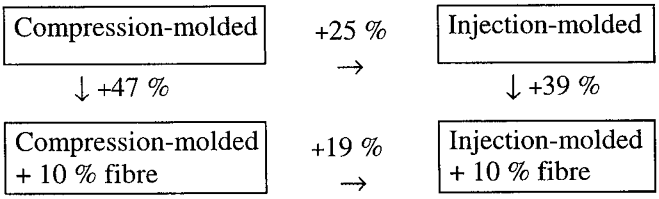

Fig. 1. Changes in Young's modulus of recycled PE as to the process used

Crystallinity

The DSC fusion peak of PE in the recycled polymer is somewhat larger with the injection-molded samples than in the compression-molded samples. PE heat of fusion in the compression-molded samples was 120.7 J/g while it was 158.5 in the injection-molded ones, an increase of 31%. Very little change in crystallinity, i.e. in the heat of fusion, was observed in injection-molded samples upon addition of 10% cellulose fibre.

Water absorption

Water absoption was estimated as 0.9 % over three months, for the 10 % cellulose fiber-containing composites. There was a concomitant loss of modulus in the composites of about 16 %. Water-repellent treatment with acetic anhydride lowered water absorption and mechanical properties loss by two thirds (Ref. 11,12).

Fibre orientation

Along with polymer orientation in injection molding, fibers, if present, are also bound to be oriented. According to a separate study, in similar conditions, with injection-molded polystyrene and 10% cellulose fibre (Ref. 14), cellulose fibres were dyed and the ASTM samples were microtomed. Fibres are indeed oriented, especially near the surface. In one of the planes microtomed, which was perpendicular to the injection axis, one can see a circular pattern, which is the image of the fountain effect in injection molding.

Fiber attrition

Since plastics have a rather low modulus and high weight, one can consider strengthening them with inorganic (glass) or organic (cellulose) fibre. In injection molding, material is compounded and injected through a small aperture, with strong concomitant shear, which results in fibre length breakdown. This was shown (Ref. 15) for a polystyrene - cellulose fibre mix. Fibre was extracted from the thermoplastic matrix through dissolution of PS, and fibre length distribution characterized. The mean fibre length was 0.75 mm before compounding, 0.5 mm after compounding (and the same for compression molding) and 0.25 mm after injection molding. In this case here with recycled PE, very similar conditions apply and were used, but dissolving away the PE/PP matrix may not be as easy and was not tried. Very similar results were obtained by Yam et al. (Ref. 7), for compounded and compression-molded samples.

From calculations involving micromechanical analysis (Ref. 15), critical fibre length for such composites was found to be around 0.8 mm. Thus a good part of the fibre are lower in length than that and so some of the reinforcing effect is lost.

Some differences between recycled plastic composites and wood

Mechanical properties

An important difference between a recycled (or virgin) plastic material and wood is the thermal expansion coefficient. For dry wood it is $(2\text{-}5) \cdot 10^{-6}$ cm/cm °C (in the longitudinal direction; across the grain, the values would be about 10 times higher) while for recycled plastics it is ca $96\text{-}155 \cdot 10^{-6}$ cm/cm °C (Ref. 1). Thus plastics expand or contract with temperature changes about thirty times more than wood: a plastic board three meters long will expand about 0.5 cm over a 20 °C rise in temperature. A cellulose fibre-filled or reinforced sample will expand much less. Another important difference is the modulus: the flex modulus of plastics, unoriented, is about only a third of that of wood: so a plastic beam will flex more under load, especially more so because its density is near unity, which is double that of some woods. That is why such composites are not used for structural ends. When making decking, fences, stairs or outdoor tables, these being important end uses, out of recycled plastics, these differences must be accounted for in material layout and architecture.

Other properties and applications

Plastic 'lumber' does have several advantages over wood and concrete, especially in outdoor, nonstructural markets such as decking, oudoor furniture such as picnic tables, benches, parking bumpers, exterior flower containers, waste containers, marine applications, etc, requiring materials impervious to moisture, resistant to impact, paintable (usually not required), recyclable, which cannot rot or degrade in UV light and that are resistant to insects and fungi, and, finally, need minimal maintenance.

CONCLUSIONS

Recycled PE/PP blends do seem to have properties similar to those of virgin HDPE. The presence of PP and contaminants at low concentrations does not hamper performance and there is no sign of phase separation. There is a large increase in properties upon going from compression molding to injection molding, due to increased orientation and crystallinity. The polymer blend is strengthened by addition of cellulose fibre, although excessive compounding will reduce the positive impact of fiber on the mechanical properties.

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RERERENCES

(1) D. Bassett, *Recycl. Prod. News*, **5** (1), 3 (1997).

(2) L. Fradette, B. Germain, *"Les Plastiques"*, Recyq-Québec Report, Québec Gov., St-Foy, Canada, 1997.

(3) Anon: Transplatek Industries, *"Faisabilité technique du recyclage des matières provenant de la collecte sélective"*, Report to the Ministere de l'Environment du Québec, Québec, Canada, 1987.

(4) J.H.van Lochem, C. Henriksen, H.H. Lund, *J. Reinf. Plast. Compos.*, **15**, 864 (1996).

(5) G. Menges, *Pure Appl. Chem.*, **68**, 1809 (1996).

(6) K.L. Yam, S. Kalyankai, S. Selke, C. Lai, *ANTEC '88* Proceedings, 1809 (1988).

(7) K.L. Yam, B.K. Gogoi, C.C. Lai, S.E. Selke, *Polym. Eng. Sci.*, **30**, 693 (1990).

(8) S. Selke, K.L. Yam, B. Gogoi, C.C. Lai, *Polym. Mater. Eng. Sci.*, **58**, 750 (1988).

(9) E. Vaccaro, A.T. Dibenedetto, S.J. Huang, *J. Appl. Polym. Sci.*, **63**, 275 (1997).

(10) F. Avalos, M.A. Lopez-Manchado, M. Arroyo, *Polymer*, **37**, 5681 (1996).

(11) H. Chtourou, "*Valorisation des plastiques recyclés par addition de fibres de bois*", M.S. Thesis, Laval University, Québec City, Canada, 1990.

(12) Chtourou, B. Riedl , A. Ait-Kadi, *J. Reinf. Plast. Compos.*, **11**, 372-394 (1992).

(13) J. Brandrup, E.H. Immergut, *Polymer Handbook*, Wiley-Interscience, New York 1975.

(14) B. Riedl, and M. Bouaziz, unpublished work.

(15) Z. Yu, B. Riedl, A. Ait-Kadi, unpublished work.

(16) Anon, Cascades Inc, promotional bulletin, Montréal, Canada 1997