Study of the processing and properties of mixtures of recycled plastics for outdoor applications

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Abstract

There is an industrial need for the study of the mechanical and physical properties of recycled polyolefins mixed with cellulose fibres from diapers, wood and packaging containing aluminium. The recycling of thermoplastic based residues is economically very interesting for the easy reprocessing of these materials and flexible shaping using conventional moulding processes as injection moulding or intrusion. Various mixtures of these materials were characterized in terms of their constituents and properties determined using 200 mm square mouldings of 10 mm and 5 mm thickness, in view of their potential application in urban furniture. These mouldings were processed by injection moulding and by intrusion that is a method which uses an extruder for delivering the melt directly into a mould. The mouldings were tested in terms of their physical properties and mechanical performance in impact and flexion. The intrusion process yielded mouldings with properties similar to injection moulding but appeared to be more attractive for requiring lower moulding pressure and thus lighter and cheaper tooling. The morphology of the mouldings, observed by bright field light microscopy showed some contamination and voiding associated to specific processing conditions and moulding size, the thinner mouldings having less voids than the 10 mm thick. The higher voiding in the thicker mouldings was detrimental to the flexural and impact performances of the parts. The coefficient of linear thermal expansion was determined and showed to be sensitive to the percentage of LDPE and fibres in the mixtures. The flexural stiffness of the moulded plates was assessed using the whole mouldings in the 3-point support test and showed the positive influence of the wood fibres and the diaper content in the mixtures. Conversely the Charpy impact performance was affected by the diaper content, but benefitted from the presence of aluminium from the recycled packaging. The prediction of these properties using the law of mixtures for predicting the overall density in the mouldings was not particularly accurate for the mixtures with recycled diapers.

1- INTRODUCTION

The massive production of different plastics materials for many new applications allowed the development of new industries, but the high consumption of plastics goods by the population and industry induced the society to the production of tonnes of municipal solid waste that needed to be recycled, land filled, or incinerated [1]. Plastics waste is not anymore a waste problem. Plastics waste is synonymous of resource of energy and easy accessible second raw materials since they exist everywhere and are materials of low added value. Thus, they can be purchased at very low prices when comparing with virgin raw materials [2]. Nowadays, recycled polyolefins are used as the main material for the fabrication of boards that can be later applied in decking, retaining walls, garbage structures, among other application [3]. The recycling company Govaerts (Alken, Belgium) is now looking for a backup plan in case of supply failure that would enable continuing production. Materials such as diapers, rich in cellulose fibres, natural wood fibres and low density polyethylene (LDPE) with aluminium are easily available and can be used as filling materials, allowing the improving of mechanical, thermal and physical properties. A major issue in achieving true reinforcement by the incorporation of natural fibres is the inherent incompatibility between the hydrophilic fibres and the hydrophobic polymers which results in poor adhesion and therefore poor ability to transfer stress from the matrix to the fibre[4]. Other problem associated to natural fibres processing is voiding. This is due to the high percentage of moisture in the wood cellulose and the temperatures well above 100ºC reached in processing [5]. Materials such as diapers are rich in chopped cellulose fibres and due to the quality requirements they have, namely the expiration date, the consumer only has a limited period of time to use them. When the expiration time arrives many diapers are sent for recycling. Due to their fibre content, they can be used in other applications in formulations with other recycled materials [6]. TetraPak is a packaging solution using paper, LDPE and aluminium, in percentages of 75%, 20%, and 5% respectively [7]. These authors reported on the recycling of aluminium and LDPE, achieving a recovery of LDPE with 18% to 20% aluminium. Nevertheless no studies have been done till the moment on the use of recycled polyolefins and natural fibres, specifically from diapers. This paper reports on a study of application of recycled polyolefins with fibrous materials and aluminium in moulded plates that could be used in urban or garden furniture.

2- MATERIALS AND METHODS

Materials. The materials used in this study were supplied by Govaerts, these being: cellulose fibres from diapers (Diapers), wood dust cellulose fibres Lignocel BK 40-90 (Lignocel), LDPE with 12% and 20% aluminium (*LDPE-A12* and *LDPE-A20*). These recycled materials consist of TetraPak packages, compatibilizer Priex 20093 (Priex) grafted with maleic anhydride, and recycled polyolefins (*Recycled PE*). The *Recycled PE* has MFI of 2,2–3,0 g/10 min (Temp, Load), and is a mixture of 30% polypropylene (PP), 55% high density polyethylene (HDPE) and 15% LDPE. With these materials, 11 formulations were processed as shown in Table 1.

Formulation #	Recycled PE	Priex	Lignocel	Diapers	LDPE-A20	LDPE-A12
1	100%					
2					100%	
3						100%
4	90%					10%
5	90%				10%	
6	87%	3%	10%			
7	80%					20%
8	80%				20%	
9	72%	3%	25%			
10	72%	3%		25%		
11	47%	3%		50%		

Table 1 – Recycled material formulations

Thermal analyses. TGA and DSC analyses were performed at 10ºC.min^{-1} on a Netzsch STA 449 F3 Jupiter for determining the melt temperature and the temperature of degradation.

Test part. This was a 200 mm square plate with thicknesses of 5 mm and 10 mm. The filling was made through a 130 mm long runner and a 5 mm wide side gate.

Processing. The formulations were dried in an oven during 5 h at a 70˚C and atmospheric pressure. The 5 mm and 10 mm thick test parts were moulded at 230ºC by injection moulding with cycle time of 75 s. The 10 mm parts were also processed by intrusion, a moulding process already used in recycling [8], using the same processing temperature and a longer cycle time of 150 s.

Characterization of the mouldings

All test data were calculated as the average of five specimens.

Thermal expansion. The thermal expansion was determined upon testing 14.6×9.8×140 mm specimens heated over 20 min from 20ºC to 80ºC, and kept at that temperature for 30 min. The coefficient of thermal expansion was calculated as:

$$\alpha = \frac{\Delta L}{L_0 \times \Delta T} \tag{1}$$

where ΔL is the variation of the length of the sample (mm), L_0 is the initial length of the sample (mm), and ΔT is the temperature variation (K).

Morphology. For the morphological characterization, it was used a stereoscope Olympus SZ-ET. The samples were first subjected to polishing, starting with sandpaper with roughness of 120 grit and then successive sandpapers of 320, 100, 2400 and 4000 grit to achieve a smooth surface. A Leica DFC280 digital firewire colour camera system was used to get the images.

Density. The calculation of the density was done using the Archimedes impulsion method for solids, as for the ASTM D 782-00 standard. The samples were parallelepipeds with area of ±4 cm² cut with a DeWalt DW876 band saw. The samples were weighed with a SCALTEC SBC31 balance; the immersion liquid was isopropanol with density of 801 kg/m³.

Impact. The 5 or 10 mm thick impact test specimens (80×10 mm) were sawn from the moulded plates. The Charpy impact test was conducted according to the ISO/WD 179-1 standard [9] using a Ceast 6545/000 pendulum.

Flexural stiffness. The flexural stiffness of the full plates was determined by the three point support test proposed by Pouzada and Stevens [10]. For a square plate this stiffness is given by the expression:

$$C = \frac{3R^2 B(v)}{4\pi h^3} S_p - \left(\frac{3R^2 B(v)}{4\pi h^3} S_p\right) \times 0{,}045 \tag{2}$$

where *R* is the radius of the plate (m), *B(ν)* is a function of the Poisson ratio (ν), S_p is the slope of the force versus deflection trace (N/m), *h* is the thickness of the plate (m) and 0,045 is a factor of correction for square plates. The tests were made in a universal testing machine, Instron 4506 with 5 kN load cell, at 21ºC. The test speed was set to 5 mm/min.

3- RESULTS AND DISCUSSION

Thermal analysis

The TGA tests done for the various materials showed no signs of degradation below 250ºC, although the natural fibres in Lignocel presented a mass loss of 6% due to their moisture content. The DSC test confirmed the existence of PP, LDPE and HDPE in the *Recycled PE* material. The characterisation of diapers showed two small peaks in the ranges of 110ºC-130ºC and 150ºC-170ºC, which correspond to LDPE and PP. The mixtures of LDPE with aluminium displayed a single peak of energy in the 100-120ºC range, corresponding to LDPE.

Characterization of the mouldings

Coefficient of thermal expansion. The results showed in Fig. 1 confirmed that the fibres restrained the expansion of the polymer matrix. Although the fibre content is smaller than in diapers, the lignocel formulation has a smaller coefficient of thermal expansion. The result obtained for *Recycled PE* base material is $4{,}6\times10^{-5}$ K^{-1}. The more expanding formulations were *LDPE-A20* and *LDPE-A12*.

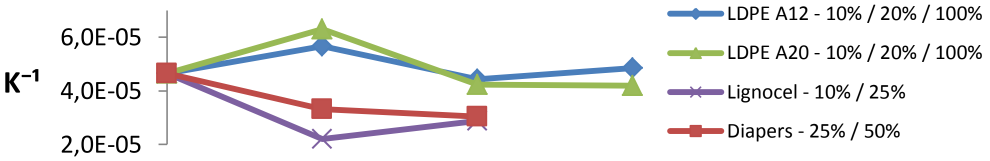

Figure 1 - Coefficient of thermal expansion of the recycled formulations

Morphology. The stereoscope analyses on the formulations 10 (with 25% diapers) and 7 (with 20% *LDPE-A12*) showed large grey pigmentation of the polymer matrix and a not defined shell and core. The analyses also showed many immiscible materials most of them probably from *Recycled PE*. The images in figure 2 show that the 5 mm thick samples show no signs of voiding or fibre agglomeration, which is a sign of good dispersion of the fibres. In injection it was observed two distinct materials that are not miscible, one with blue colour and the other with white colour; these materials are unknown and must come from the recycled. In the intrusion mouldings it was possible to see fibres of different size dimensions and colours that most probably come from the diapers material, also no signs of fibre agglomeration.

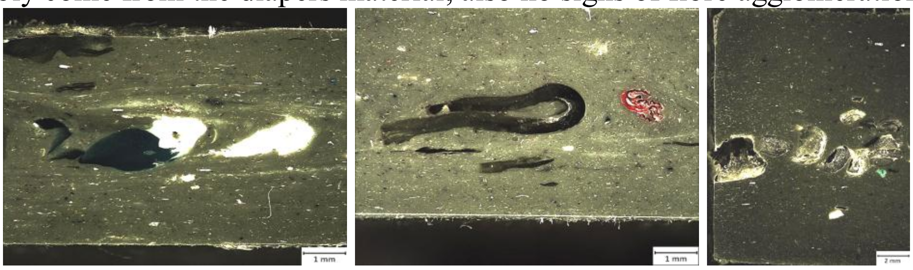

Figure 2 - Mouldings of formulation 10 (Recycled PE+25% diapers): a. - 5 mm injection, b. - 5 mm intrusion, c. - 10 mm intrusion.

The images of formulation 10 (*Recycled PE*+25% diapers) in 10 mm-thick intrusion mouldings, showed voiding in the core. The void size and depth are irregular. The 5 mm-thick samples didn't show signs of voiding in injection or in intrusion. There is orientation of fibres near the surface and vertical orientation in the core. This is the effect of the shear flow during the filling. No agglomeration of fillers was observed and they seemed to be well dispersed in the polymer matrix through the whole flow length.

Density. For all formulations, the 5 mm-thick parts were denser than the 10 mm, as summarised in Figure 3. This results from the voiding in the 10 mm-thick mouldings as observed in the morphological analyses. In the 5 mm-thick parts the addition of fibrous materials or aluminium slightly increased the density, this being more evident in the formulations with fibres than with aluminium.
In pieces of 10 mm, the addition of fibres promoted the density increase while the addition of LDPE with aluminium promoted its decrease. The formulations with higher density were those with aluminium, *LDPE-A12* and *LDPE-A20*. The nominal density of the *Recycled PE*

874 kg/m³. In the studied formulations the density was around 910 kg/m³ in 10 mm-thick mouldings and 960 kg/m³ in the 5 mm-thick.

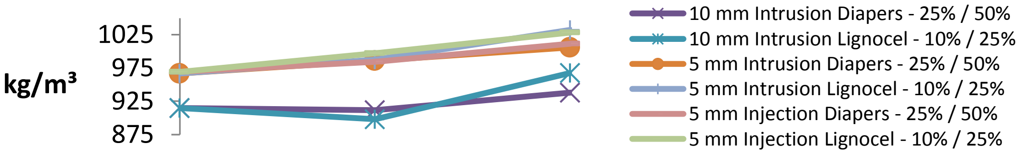

Figure 3- Density data of fibrous formulations

Flexion. As like in the densities results, in flexion the 5 mm-thick plates showed better performance in flexure than the 10 mm-thick parts. The 5 mm-thick parts showed identical flexural stiffness irrespectively of the processing method, intrusion or injection moulding.

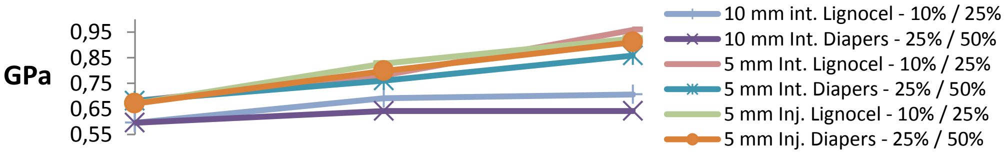

Figure 4 - Flexion data of fibrous formulations

The data in figure 4 showed that the addition of fibres promoted the increase of the flexural stiffness. It was also possible to conclude that cellulose fibres in lignocel promoted higher flexural stiffness these being more efficient that those in the diapers. Higher fibre content promoted higher flexural stiffness. The addition of LDPE with aluminium promoted a small decrease of the flexural stiffness of the recycled. The materials with lower flexural stiffness were, as expected, the materials with higher content of LDPE that is a material very ductile. *LDPE-A12* and *LDPE-A20* were the materials with lower flexural stiffness, around 0,35 GPa. According to the Govaerts company the modulus of *Recycled PE* is 0,49 GPa, the best value obtained for the same formulation was 0,68 GPa for Intruded pieces of 5mm thickness and the lowest was 0,6 GPa for 10 mm thick-intruded pieces.

Impact. The impact resistance results were not so stable as the density and flexion. Although the parts with 5mm thickness presented better impact resistance than the parts of 10 mm thickness, the standard variation was higher for parts with 5mm thickness than for parts with 10mm thickness, as shown on figure 5. The analysis of the results allows concluding that the addition of fibres to *Recycled PE* promoted the decrease of the impact resistance. Higher fibre content promoted lower impact resistance, the exceptions, were the formulation of 25% Lignocel and 50% Diapers with 10mm thickness.

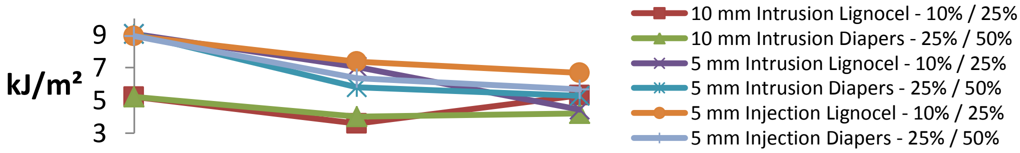

Figure 5 – Flexion data of fibrous formulations

The addition of LDPE with aluminium in the recycled polyolefins, promoted the decrease of the impact resistance for concentrations lower than 10%, although in pieces of 5mm thickness Injected and pieces 10mm thickness Intruded the impact resistance increased. For higher concentrations the impact resistance was enhanced. The materials with higher resistance were the materials with higher content of LDPE with aluminium, *LDPE-A12* and *LDPE-A20*. For *Recycled PE*, the values declared by Govaerts were lower than the obtained values, 9,08 kJ/m² was the best obtained and 7,62 kJ/m² was the declared.

4- CONCLUSIONS

This study envisaged to help the recycling company Govaerts to consider alternative possibilities for their current recycled formulations based on polyolefins. It was found that formulations with packaging waste or diapers are a possible solution. Voiding has an important influence in the final properties of the processed parts. The morphological analyses showed voids in 10 mm-thick parts, which are less dense than voidless 5 mm-thick mouldings. The thermal expansion coefficient of the formulations with aluminium or fibres was significantly reduced, especially in the case of fibres. The formulations with fibres have consequently higher flexural stiffness, and even with low content of fibres, this effect is obvious. As for impact resistance, the addition of fibres reduced the toughness. The cellulose fibres from lignocel lead to better results than from diapers, either in flexion or in impact. The formulations with *LDPE-A12* or *LDPE-A20*, from TetraPak packaging, slightly reduced the flexural stiffness. As for impact, the composition with concentrations of these aluminium containing recycles, up to 10%, reduces the impact resistance. However it improves for concentrations higher than 20%.

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