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# Sheet Forming Studies using Low-cost Composites

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**ABSTRACT:** This paper focuses on formability study of recycled polymeric matrix composites. Two thermoplastics are selected to create the composite, i.e. polyethylene terephthalate (PET) and high density polyethylene (HDPE). The recycling process is thermo-mechanical, and no addition of co-polymers, anti-oxidants or additives is used. Two sets of composites were studied. For the first set a PET/HDPE composite with ratio 60/40 was prepared, while the second one was a PET/HDPE/fiber glass composite. The PET/HDPE resulting matrix has the same aspect ratio as the first one and the final configuration matrix/fiber has a 70/30 relationship. The forming limit diagram (FLD) was obtained by following the methodology proposed by Bhattacharyya and his colleagues. The obtained results are very encouraging and open a new market for the recycling process.

**KEY WORDS:** polymeric matrix composites, recycling, PET/HDPE composite, forming limit diagram.

## INTRODUCTION

**P**OLYMERIC MATRIX COMPOSITES (PMC) are used in engineering applications as they present low density and high strength. However, they are not used in large-scale applications due to their high cost. The large variety of thermoplastic matrices allows us to experiment with different

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types of resin combinations creating the so-called melt-blended matrices, which can be seen as alternatives. Melt-blended matrices can help reduce cost and, at the same time, increase specific properties such as strength. One low cost source of such type of resins is recycled thermoplastics. Post-consumer polyethylene terephthalate (PET) in a large city such as Belo Horizonte – Brazil, for instance, has its local price equivalent to fifteen US cents/kg while polystyrene (PS), high and low density polyethylene (HDPE, LDPE), and polypropylene (PP) have an even lower cost. Their cost ranges from 5 to 10 US cents/kg [1]. Their low cost is due to the large amount of post-consumer plastic waste generated daily in large cities worldwide. To give an example, a city in an emergent country with a population of three million inhabitants produces each day around 400 tons of plastic waste [2]. As a consequence, municipalities in Latin America, especially in Brazil, are gradually getting more concerned about a 25% increase in plastic waste generation per year while the landfill area is only increasing at a 7.5% annual rate. Therefore, as Scott [3] points out, by the year 2015 there will be no disposal options for plastic waste.

In 1997, Jones and Baumann [4] reinforced the importance of implementing a recycling strategy due to the large increase of plastic waste generated and the cost to manage it. They also stated that although there are governmental regulations in countries such as Germany, recycled materials are not used not only because of their physical properties but also due to their surface appearances. In other words, many designers are afraid of using them as they can be rejected by the market. But this attitude can change. Recently, Martins and De Paoli [5] stated that around 15% of all rigid plastics and films consumed in Brazil are recycled and returned to industry. Moreover, according to Selke [6], the total amount of plastics in the US municipal solid waste stream is around 10%; of this amount 44% is from containers and packaging. A large quantity of these containers is from soft drink bottles. The fact that some states in the US are engaged into an environmental strategy, e.g. Michigan, where the recycling rate is close to 100% proves the potential for recycling plastic waste as well as changes in the market attitude.

One possible solution for the municipality plastic waste problem is the recycling procedure. According to Scott [3], plastic waste management can be done by three different approaches. The first one is the mechanical recycling approach, which is a technique similar to procedures used to recover materials suitable for second use. The second one is known as energy recovery, which can be performed by two distinct ways: (a) incineration where the hydrocarbon polymers replace fossil fuels and (b) pyrolysis or hydrogenation to low molecular weight hydrocarbons for use either as portable fuels or as polymer feedstock. A third approach is defined by Scott

[3] as the biological recycling. This approach takes advantage of the polymer biodegradation, which is highly dependent on the polymer type and environmental conditions. Moreover, this type of recycling most often involves not only high costs and complex procedures but also potential damages to the environment. Due to all these problems, a feasible solution for the increase of plastic waste generation is the thermo-mechanical recycling. This technique contains a combination of mechanical recycling where the thermoplastics are granulated and later on transformed by techniques such as extrusion or thermoforming.

Furthermore, as said by Brandrup et al. [7], to be economical, a mechanical recycling procedure must be designed in such a manner that the amount of energy needed to produce the virgin material plus the energy to dispose of the material must be equal to the energy to recover the post-consumer materials plus the energy during the reprocessing itself. This balance, however, does not take into consideration environmental benefits. When environmental gains are considered, it could allow for higher energy consumption during the mechanical recycling. The increase in cost would be compensated by indirect cost reductions in landfills, for example. Unfortunately, the large majority of the population does not consider the indirect cost reduction as a social benefit.

The objective here is to study the formability of a recycled polymeric matrix composite (RPMC), where no addition of any kind of stabilizer, compatibilizer, or additive is applied to the blended matrix. Moreover, the fiber reinforcement is done by discontinuous fiber glass random oriented on the recycled matrix. Finally, it is important to mention that the mechanical properties will be lower compared to those of virgin materials, but the overall cost effectiveness is better.

## **MATERIALS AND SAMPLES MANUFACTURING**

Before describing the sample manufacturing procedure developed in this paper, some comments must be made. First, the recycling process used in this paper is essentially thermo-mechanical. Second, the entire analysis performed here takes into consideration the macro-mechanical approach as defined by Tamma and Avila [8]. This analysis is mainly concerned with the composite macroscopic mechanical properties and it will be associated to the mechanical properties changes due to the recycling process [9].

The materials were selected after taking into consideration not only their quantity in the urban plastic waste but also their potential use in civil and mechanical engineering areas. According to Brazilian Business Commitment for Recycling (CEMPRE) [10], the total amount of plastic waste in urban solid waste in Brazilian cities is around 18%. From that amount, the

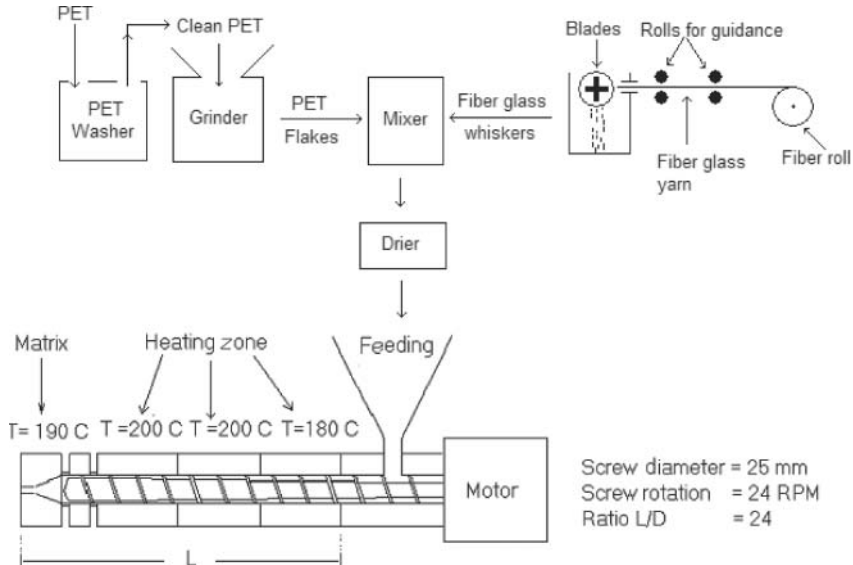
following percentages were obtained: HDPE and LDPE 36%, PET 20%, PVC 13%, PP 11%, and 20% others. Furthermore, the postconsumer PET in Brazil has a singular characteristic: around 2/3 of all PET used in the manufacturing of soft drink bottles is green, and as such researchers focused on green PET bottles. As most of the time bottles and caps are discarded together, it was decided to create a particulate composite material made of a PET matrix (bottles) and HDPE particles (caps), with and without fiber glass whiskers reinforcement. Notice that pure PET at room temperature has a brittle behavior, to make it ductile the HDPE particles are added to the PET matrix.

The reason for using fiber glass instead of natural fibers, e.g. jute, flax, sisal, hemp, relied mainly on two factors: (a) their dimensional homogeneity, constant cross section, which leads to a standard methodology for computing stiffness and strength, and (b) its capacity of standing to high levels of humidity without developing fungi or mould. Notice that natural fibers such as flax, hemp, and sisal not only have large variations on cross section shape and dimensions but also present a nonlinear stress-strain curve [11]. Moreover, the hydrophilic nature of organic fibers could guide to the presence of fungi that could cause fiber degradation which leads to poor mechanical properties [12].

Pawlak et al. [13] point out that to be able to obtain the mechanical characterization of polymeric scraps, it is necessary to homogenize the material. Nevertheless, as the materials used are from postconsumer bottles, a pretreatment must be done before the homogenization procedure.

The pretreatment consists of three different steps, namely: washing, granulation, and drying. The granulated material average length and width are 8 and 2 mm, respectively. The drying program was developed considering the grain dimensions and their thermal-mechanical properties. Moreover, the HDPE is assumed as spherical or ellipsoidal inclusions in the PET matrix. By taking into account this geometric arrangement, it is possible to estimate the composite's thermal properties, i.e. thermal conductivity and thermal expansion coefficient. After determining the material properties, it is feasible to define the drying procedure. It consists of a gradual increase in temperature, which goes from room temperature to 100°C at a rate of 5°C/min. When the steady state has been reached, the temperature is kept constant for a 24-h period. This procedure guarantees levels of humidity less than to one percent per volume, as mentioned by Faria and Ávila [14].

The homogenization procedure was performed by extrusion. For this specific case, the extrusion operational parameters are presented into the diagram shown in Figure 1. Due to whiskers dimensions, diameter of 10  $\mu\text{m}$  and length of 10 mm, and the extruder mechanical characteristics, diameter

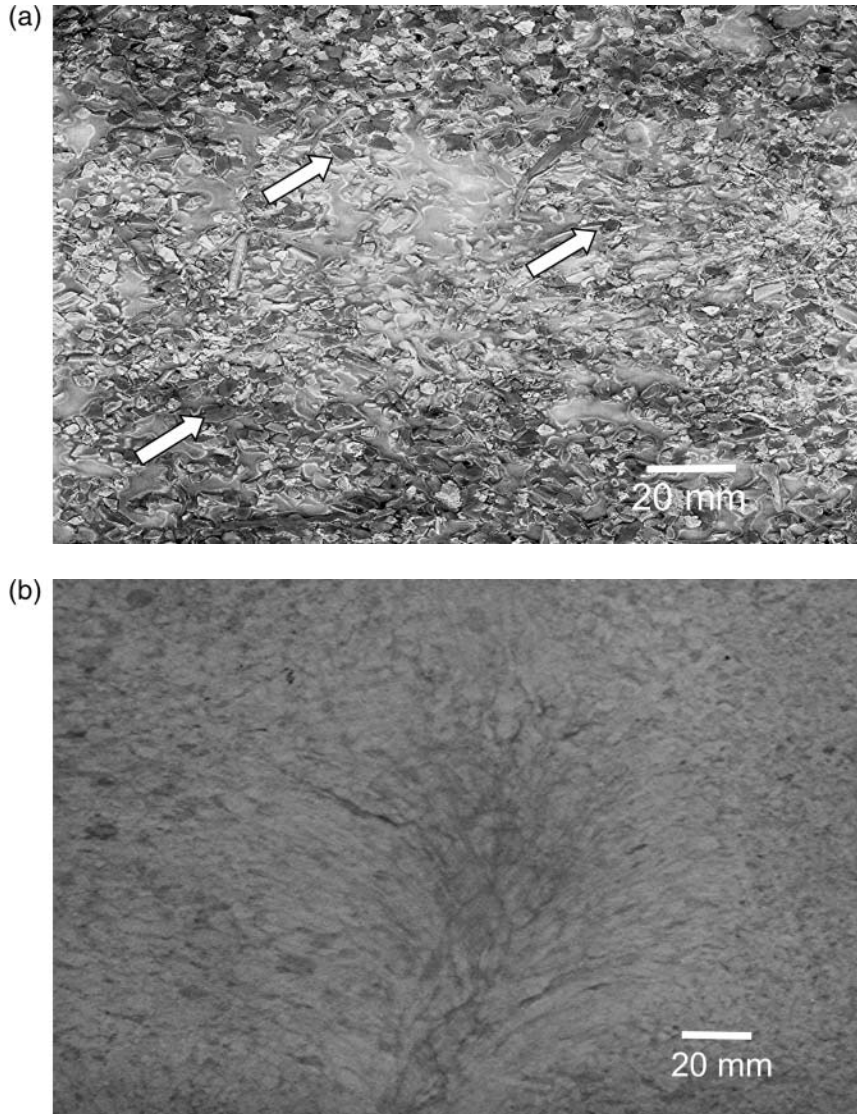


**Figure 1.** Diagram for pretreatment and extrusion procedures including fiber glass reinforcement.

of 25 mm and variable worm-lead from 25 to 15 mm, it is possible to state that most of the whiskers would be surrounded by the mixed matrix during the extrusion. Moreover, considering the shear forces usually developed during the extrusion process, it is feasible to believe that the fiber reinforcement can be assumed as short fibers, with length less or equal to 10 mm, random oriented in a homogeneous matrix. As a consequence, the composite can be assumed as quasi-isotropic without loss of generality.

The next step is the manufacturing of rectangular plates, 247 mm by 214 mm and 2.0 mm thick, by hot compression. During hot compression, a uniform force of 6.0 tons at  $200 \pm 4^\circ\text{C}$  is applied for a period of 2.5 h. The values are obtained by the heat transfer equations given by Mantell and Springer [15] and preliminary tests. Notice that low temperature could cause lack of adhesion between the phases. Hence the plate becomes brittle. In Figure 2(a) the white arrows point out some of the regions where lack of adhesion occurred. On the other hand, an excessive applied pressure could induce a nonuniform flow, as it can be seen in Figure 2(b).

Notice that for large scale operations, the hot compression systems must be changed to a continuous hot rolling process right after the extrusion step. Although the productivity during the continuous production is higher, the costs are also considerably higher. The hot compression seems to be a good technical alternative for small scale operations, e.g. local recycler co-op,



**Figure 2.** Plates made by hot compression: (a) lack of adhesion; (b) excessive applied force.

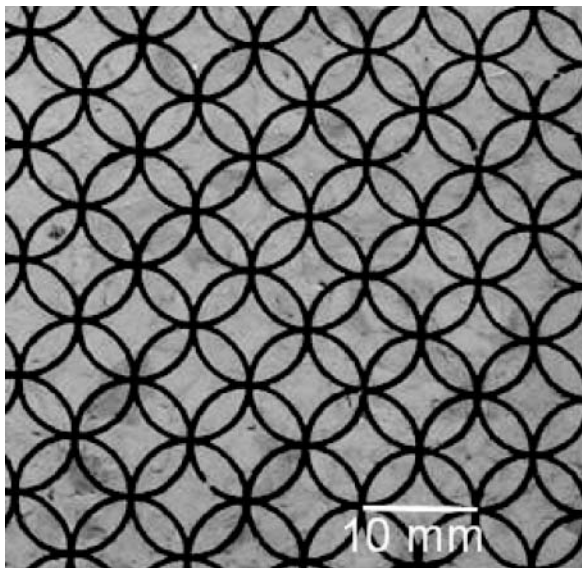
even though a considerable part of the product's final cost will be represented by the energy cost. Nevertheless, if the environmental and social gains are taken into consideration the final economical balance will be on the surplus side.

## EXPERIMENTS

Even though the ASTM D 638 [16] is the standard tensile test for polymers, it was decided to use the ASTM D 3039/3039M [17], due to the blanks geometric configuration, i.e. rectangular flat plates. The tensile tests were performed on an INSTRON 4482 universal testing machine at room temperature and with a constant speed of 2.0 mm/min.

The sheet forming limit tests follow the procedure proposed by Martin et al. [18]. It consists of setting a series of rectangular planar blanks with constant thickness and length, and variable width. On the top surface of each blank, a silk screening process was used to apply an array of circular grids with a diameter of 10 mm, as can be seen in Figure 3. All blanks have thickness and length equal to 2.0 and 200 mm, respectively. The blanks width, testing temperature and testing speed are listed in Table 1. The hemispherical punch has a diameter of 155 mm and their dimensions are compatible to those presented in Gosh et al. [19].

Once the blanks are prepared, the original circle grid diameters ( $d_0$ ) are measured. Then, the blanks are heated and tested by deep drawing up to failure. A homogeneous deformation will change circles to ellipses, and the major and minor surface strains can be calculated from the measured lengths of the major and minor axes,  $d_1$  and  $d_2$  using a measuring



**Figure 3.** Circular mesh on the blank's surface.



**Table 1. Experimental variables for formability tests.**

Sheet width (mm)	Forming temperature (°C)	Forming speed (mm/min)
25	95	2
50	95	2
75	95	2
100	105	1
125	105	1
150	105	1
175	105	1
200	105	1

microscope, for example. Wagoner and Chenot [20] defined the principal natural strains as the following:

$$\varepsilon_1 = \ln\left(\frac{d_1}{d_0}\right); \quad \varepsilon_2 = \ln\left(\frac{d_2}{d_0}\right); \quad \varepsilon_3 = -\varepsilon_1 - \varepsilon_2 \quad (1)$$

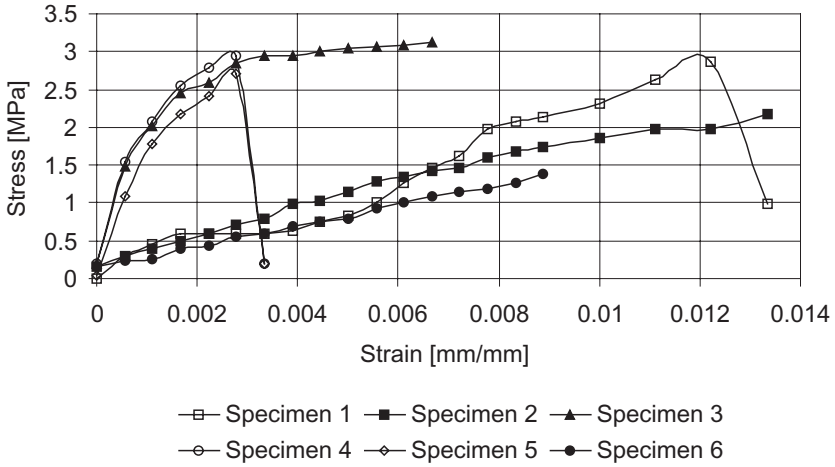
To construct the forming limit diagram (FLD), deformed circles (ellipses), both close to and within necked or fractured areas, are measured. Gosh et al. [19] identify three types of ellipses: type I, fractured (the fracture passes through the ellipse); type II, necked or fractured affected (the ellipse lies either within a necked area or within the fracture-affected zone); and type III, acceptable (the ellipse falls outside the area affected by necking or fracture). Once all principal strains are calculated, a  $\varepsilon_1$ - $\varepsilon_2$  plot is created. The FLD locus must be above the type III ellipses and below types I and II ellipses.

## RESULTS AND DISCUSSION

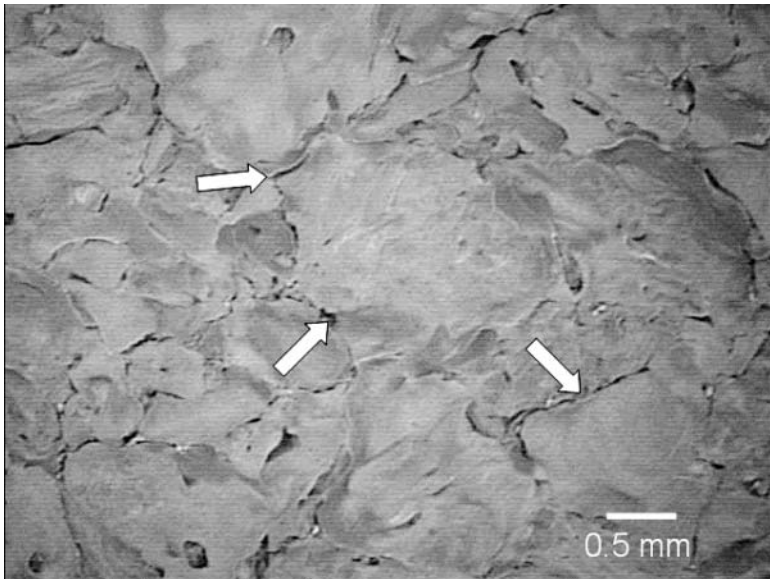
Two distinct materials were studied. The first one was a PET/HDPE composite material with a weight ratio close to 60/40. The second one was a PET/HDPE fiber-reinforced composite. In the second case, the reinforcement was made of glass fibers whiskers in a ratio fiber/matrix, in weight, close to 30/70. The PET/HDPE relative amount for the reinforced composite was the same for both specimens. The goal is to evaluate not only their stiffness and strength but also their formability.

### Tensile Tests

ASTM D 3039/3039M specimens were prepared for each group of composites. Figure 4 shows the stress-strain behavior for each PET/HDPE composite. The small strains at failure for the #4 and #5 specimens seem to



**Figure 4.** Stress–strain curves for the 60/40 composite without fiber reinforcement.



**Figure 5.** Macrograph showing lack of adhesion and intergranular micro cracks.

be due to premature local necking caused by a local lack of adhesion that could lead to the development of intergranular micro cracks. The macrograph shown in Figure 5 locates these micro cracks. When the overall composite behavior is compared against the virgin components,

HDPE or PET, it seems to have a poor performance. However, two factors must be considered: (a) they are recycled materials and (b) there is no use of any compatibilizer during their manufacturing. Moreover, according to Scott [21], a degradation of mechanical properties during the extrusion process could be attributed to the oxidation process. However, the stiffness and strength obtained are still suitable for low loading engineering applications, e.g. small houseware devices, wheels for supermarket carts, etc.

The results for the PET/HDPE/FIBER composites are shown in Figure 6. As expected the fiber addition leads to slightly high values of strength and stiffness. The same behavior, a small increment on strength and stiffness, was also observed by Singleton et al. [22] when studying recycled composites. In both cases, the modest increase on strength could be attributed to the small interaction between fibers and the recycled matrix.

It is evident that a non trivial phenomenon occurred during the #4 specimen test. By analyzing the specimen fracture region, PET inclusions were noticed. The nonblended PET inclusions served as cracks nucleation sites that reduced the composite strength. The arrows in Figure 7 show the cracks generated by the PET inclusions.

### The Forming Limit Diagram (FLD) Construction

The FLD involves four different stress fields, i.e. pure shear, simple tension, plain strain tension, and biaxial tension. Martin et al. [23] suggested that to be able to cover all these fields the blanks' width must change.

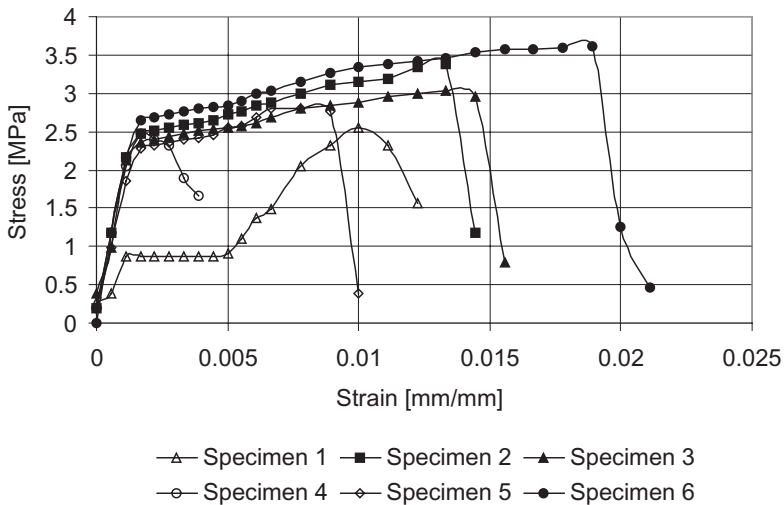


Figure 6. Stress-strain curve for the PET/HDPE/FIBER composite.

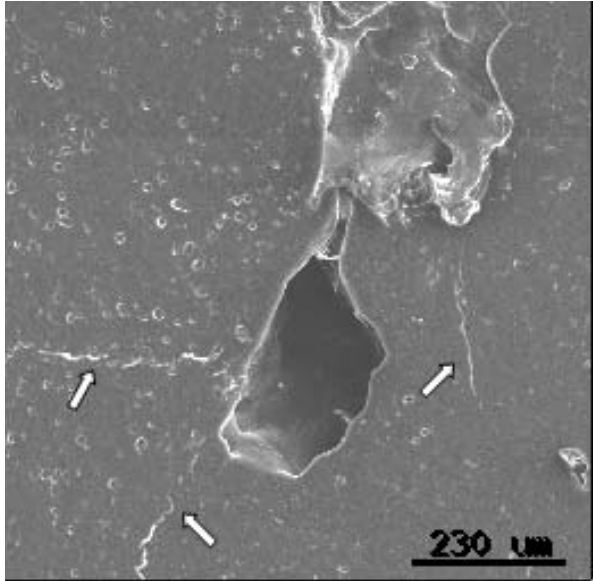


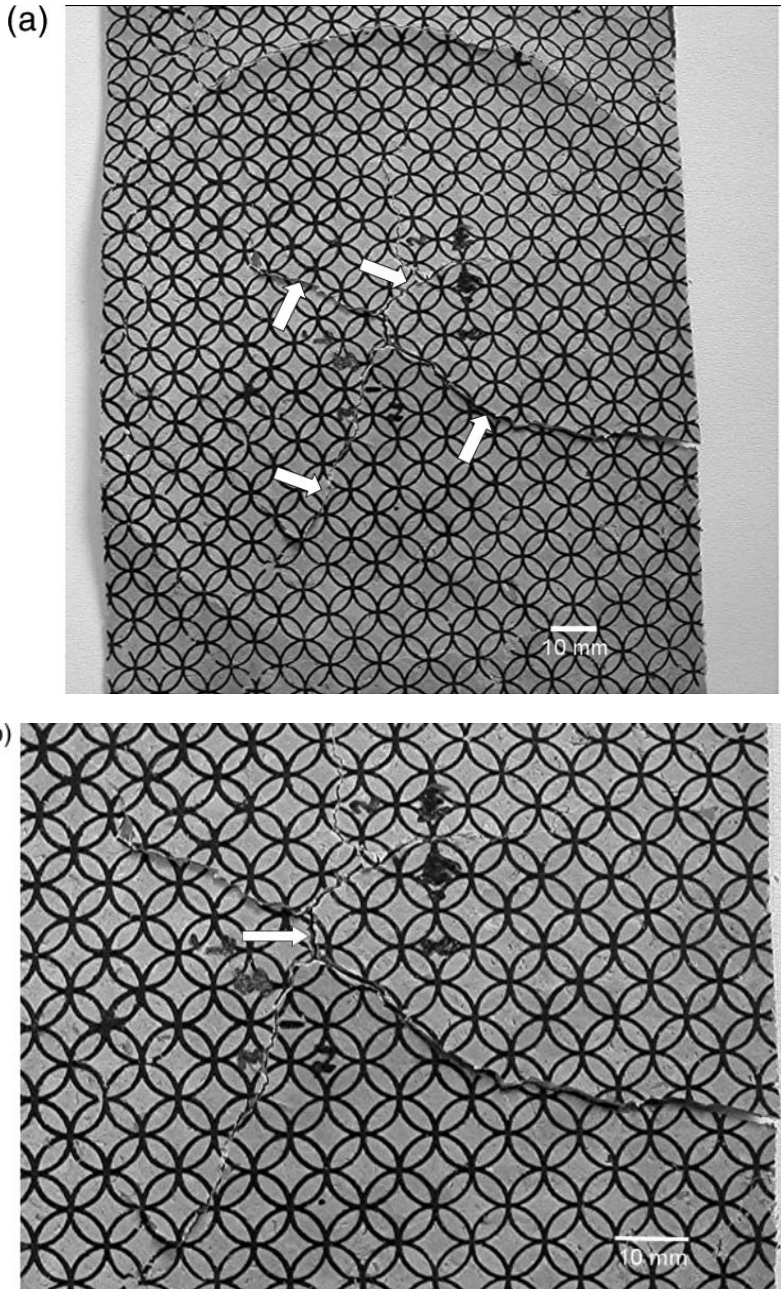
Figure 7. Cracks generated by nonblended PET inclusions.



Figure 8. Shape fracture in a 50-mm wide blank.

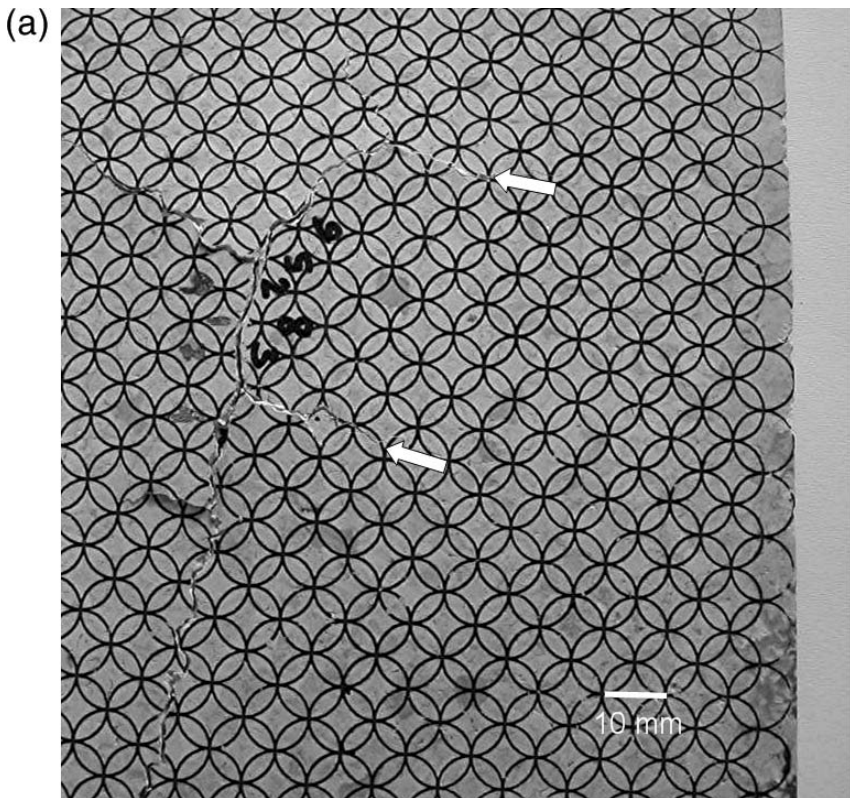
The variation used in this paper is listed in Table 1. However, the main question relies on the fact that this composite is made of recycled materials and we do not know its behavior under stamping operations.

The first blank considered is the one with 50 mm width. As it can be seen in Figure 8, the crack is linear from one side to the other side of the blank. It seems to be caused by shear deformation. As the blanks become wider, the crack's shape changes. The cross shape crack is shown in Figure 9(a), it is

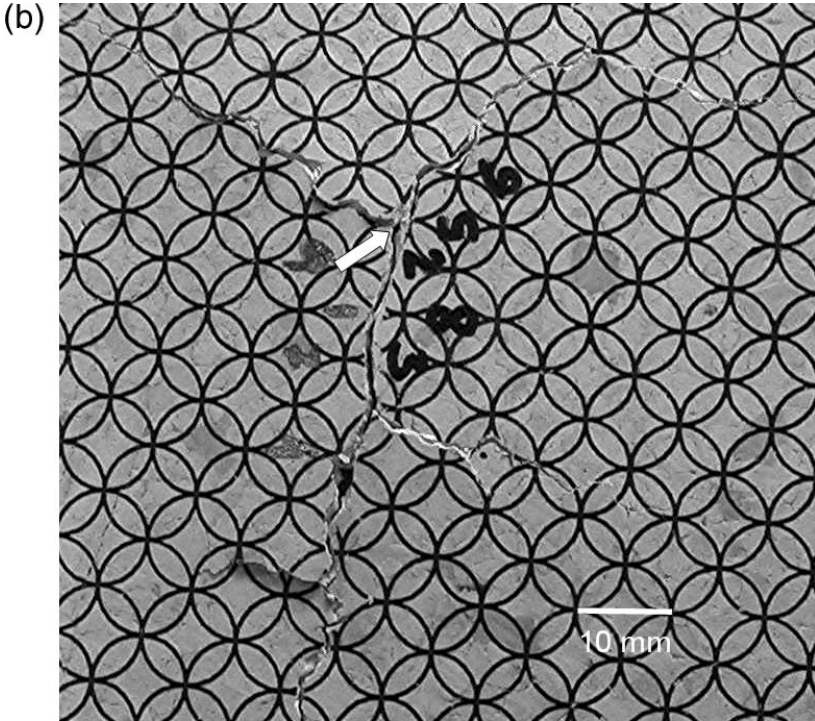


**Figure 9.** (a) Shape fracture in a 125-mm wide blank; (b) shape fracture detail in a 125-mm wide blank.

important to notice the propagation path, i.e., it is from the center to the edges. This effect is in part due to the stress distribution created during the deep drawing process and the free-edge effect. A fracture closer view is shown in Figure 9(b), where the fracture initiation locus is indicated by a white arrow. For wider blanks, e.g. 150 and 200 mm, the free-edge effect is no longer experienced. As can be observed in Figure 10(a), the fracture does not reach the blank's edge (the white arrows indicate the cracks ends), in other words, their locus is limited to the blank's central region. This could be an indication that the stress field changes from simple tension to plain strain tension. It seems that the highest strain was located at the center of the blank, which could lead to crack formation, as indicated in Figure 10(b). By changing the friction coefficient, it is possible to obtain the biaxial tension stress field. However, the crack shape is similar to the ones presented



**Figure 10.** (a) Shape fracture detail in a 200-mm wide blank; (b) shape fracture in a 200-mm wide blank.

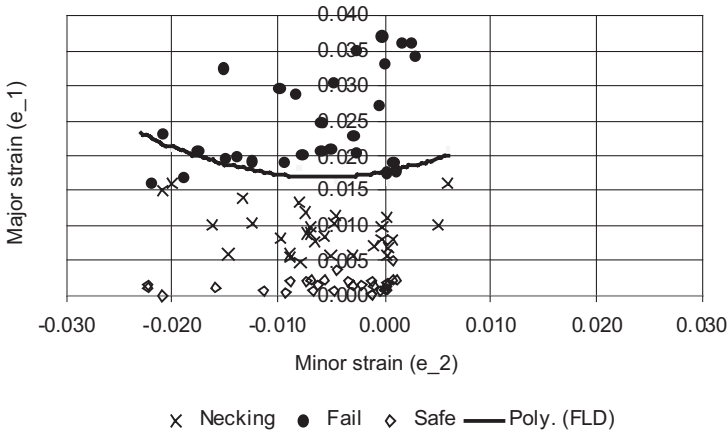


**Figure 10.** *Continued.*

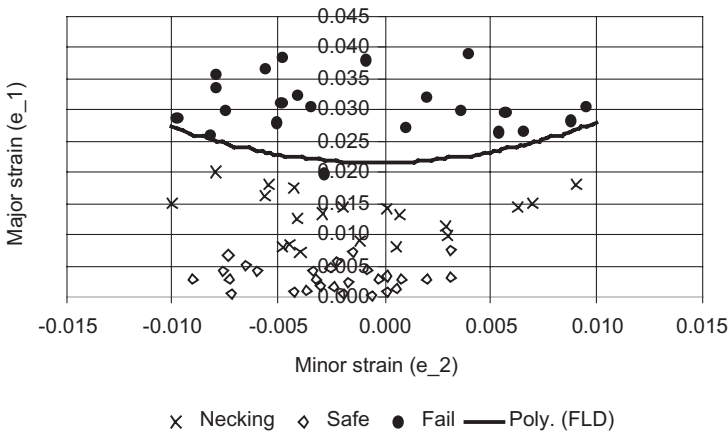
in Figure 10(a) and (b). The strains obtained for this case are larger and they are represented by the points on the boundaries of the FLD chart. By considering all facts listed before, it is possible to conclude that even though the composite was made of recycled materials, its behavior under stamping operations is similar to those made of virgin materials. Moreover, all four stress fields could be detected by using the methodology proposed by Martin et al. [23].

The final step is the FLD construction for both composites studied. The consolidated forming limit diagrams for HDPE/PET and HDPE/PET/Fiber glass composites are shown in Figures 11 and 12. Although the safe strains seem to be very low for some practical applications, the reasons could be due to the following factors: (a) use of recycled thermoplastics without addition of co-polymers, and (b) matrix high viscosity. The use of additives could lead to a much flexible matrix as it increases its elongation, which could guide to higher safe strains.

As mentioned by Martin et al. [23], the matrix viscosity is also a key parameter for forming operations. As temperature and viscosity for the



**Figure 11.** FLD for HDPE/PET composite.



**Figure 12.** FLD for HDPE/PET/Fiber glass composite.

present case are inversely proportional, an increase on temperature will cause a decrease on viscosity. As a consequence, the matrix flow under tension will be considerably higher, this would bring higher strains. However, as we are dealing with a recycled matrix the degradation issue must be addressed. Moreover, for practical applications, a large increase on forming temperature could cause premature deformations on blanks due to the matrix low viscosity. The forming temperatures used in this study were obtained considering a balance of all these factors. For a different type of HDPE/PET ratio, the same considerations must be done.



By analyzing the two FLD curves, it is possible to conclude that the fiber addition to the composite brought up more stiffness to the composite. Besides the increase in stiffness, an increase in elongation was also noticed for the reinforced composite. It seems that the added fibers increased the composite formability – an important property to the manufacturing process such as deep drawing or stamping.

Finally, when the overall composite behavior is considered, it appears that although safe strains seems to be low, they are distributed along the four stress field range, i.e. on the FLD the strain points are from pure shear, simple tension, plane strain, and biaxial tension as discussed by Avila and Jabbur [24]. The experiment performed by Martin et al. [18] in unidirectional  $[0]_4$  fiber glass/polypropylene composites has a clear plane strain nature, in other words, the points are concentrated around the major strain axis, which brings as a consequence small values for minor strains. They not only show the presence of low safe strains for the cross ply  $[0,90]_{2s}$  and angle ply  $[0,+60,-60]_{2s}$  fiber glass/polypropylene, but they also mention that most of the safe strains are located on the FLD left side, which makes impossible the existence of a biaxial tensile stress field. In short, it seems that there is no universal composite that could be applied in a general way.

### CLOSING REMARKS

The formability of a recycled polymeric matrix composite (RPMC) was studied, and by analyzing the data obtained it is possible to conclude that the thermoforming procedure of this material is feasible. Moreover, the addition of fibers in a random distribution makes the RPMC sheets more susceptible to stamping operations. Thermoforming operations using RPMC can open a new market for recycled materials, especially as a cheap option to injection molding.

The recycled composites studied are able to support stress fields from shear tension, to pure tension, plane strain, and biaxial tensions. This could be evidence of more flexibility that can be applied at industry. However, some improvements must be made with regard to the safe strains.

### ACKNOWLEDGMENTS

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