Study on the influence of recycled material on the tensile strength of HDPE products

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Abstract. The content of this paper concerns to the new tendency in industry, the concept of circular economy, based mainly on the decrease in plastic waste pollution by recovery and at source and on the significant optimization of production costs. The mechanical tests have been made on handles injected from several mixtures of grinded HDPE, with different melt flow rates and in different proportions, in order to establish the conformity of the final product to the imposed technical specifications regarding the breaking strength of this. For the industrial application, an important result is the validation of the recipes proposed for the preparation of raw material from recycled HDPE for which the final injected product preserves the physical-mechanical properties within the imposed limits. The study shows that in certain cases, for the preparation of the raw material, it could be used up to 100% recycled material, without affecting the technical specifications of mechanical resistance imposed on the final product.

1. Introduction

The rise in using plastics in any industrial domain has become a trend in the last decades not only because these raw materials are low in cost, weight and are associated with low processing expense but, compared to metals, the plastics additionally offer, [1-6]:
- a wide range of mechanical properties,
- a wide range of manufacturing procedures and technologies
- superior flexibility in part design, even at very complex 3D shape configuration
- easy product assembly, at reduced time and cost, by using snap fit joints and less fasteners,
- corrosion resistance to almost any known chemical,
- no need for secondary finishing operation,
- transparency, in combination with all the above advantages,
- negligible technological waste compared to the manufacture of the parts from metal.

In the field of packaging as well, there is a tendency to increase consumption and the industrial environment responds to this demand with various offers. Due to the population’s increase in purchasing power and the development of the e-commerce concept, the consumption of plastic material for the packaging manufacture is expected to keep the growing tendency for the next decade, [4, 7, 8]. The main challenge that aroused from this tendency is in conjunction with the quantity and the nature of the raw material in-use, to the increase of environmental pollution with non-degradable plastic waste or with low biodegradability.

To this end, the European Parliament has issued Directive 2019/904, mandatory to be implemented in all EU countries, [9], promotes circular approaches that give priority to sustainable and non-toxic re-usable products and re-use systems rather than to single-use products, aiming first...
and foremost to reduce the quantity of waste generated by these. Among the regulations mentioned in this document, with reference to plastic products, especially for disposable ones, the most significant are:

- if there are alternatives to use biodegradable materials instead of regular plastics, their application is more than necessary,
- it is mandatory to adopt measures to reduce the consumption of plastic material for the manufacture of disposable packaging for the food industry,
- the marking and labeling of the products in large and mass series are strictly regulated.

Regarding plastics recycling, the documents of EU, more specifically for plastic packages, proposed the following objectives, [7-10]:

- a selective collecting strategy for plastic tailings, up to 75% by 2025 and 90% by 2029,
- the application of stricter rules on eco-friendly designs for single-use bottles and also for their lids,
- the mandatory use of 25% recycled material for the manufacture of PET containers starting with 2025, a percentage that will increase up to 30 starting with 2030 for all plastic packages.

2. General considerations about using recycled plastics

The primary methods of waste disposal are landfill or incineration but, in the context of the above mentioned regulations, the effective and intelligent recycling became now a major challenge for the plastics processing sectors [7]. New approaches are nowadays successfully used in plastics recycling such as the eco-friendly downgrading and downgauging concepts in combination with intelligent packaging design and digital recycling label but also the recycling in closed-loop of mixed plastics waste (ASTM D5053), [11, 12], and this the last is the stream idea of the present paper.

The most common categories of plastics in household and industrial waste are PP, PET and PE, [1, 8, 10]. Unfortunately, in many countries, the operations of selective waste collecting do not have the expected results. Monitoring this activity reveals that the recycling companies indiscriminately mix the collected waste, and this result further leads to technological difficulties in the subsequent processing phases. In the sorting stage of the recycling process, it is relatively easy to confuse PP and PE and certain incompatibilities in mixing these materials have already been reported through various studies, [1, 10, 13, 14]. According to them, the main source of incompatibility is due to the significant difference between the melting temperature values of the mixed materials. Thus, at a certain values of temperature, several phases will coexist in the volume of the melted mix and this anisotropy will characterize also the material injected into the cavity (nest) and will affect the mechanical properties of the final product.

Next, the low interfacial adhesion between different phases in the melt is also responsible for a decrease in mechanical properties especially related to its intimate morphology, including impact strength, strain at break and ductile to brittle transition, [1, 2, 5]. In this case, the physical-mechanical properties of the products resulting from these mixtures are seriously affected and, if the quality requirements refer to mechanical strength too, the non-conformity rate of the products resulted from the manufacturing process increases dramatically, along with the negative effects related to economic efficiency, [3, 5, 8].

Two plastic materials are perfectly compatible when mixed and the mixture could be processed without any technological difficulties (stable process, easy to control, minimum scrap rates) if their melting temperatures have close values, on condition they have high interfacial adhesion between melted phases.

In this context, the process manager makes decisions based on information regarding the processing of the polymeric material used and its recycling opportunities, the possibility of mixing it with other material, as well as the synoptic picture of the possible defects and the reasons that cause them, [1, 2, 4, 5, 15-17].
Table 1. Mixing compatibility for thermoplastic materials at injection, [18].

| MINOR MATERIAL | ABS   | ASA   | PA    | PBT   | PBT+PC | PC    | PC+ABS | PC+PBT | PE    | PET   | PMMA  | POM   | PP    | PPO   | PPO+PS | PS    | PVC   | SAN   | TPU   |
|----------------|-------|-------|-------|-------|--------|-------|--------|--------|-------|-------|-------|-------|-------|--------|--------|-------|-------|-------|-------|
| ABS            | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| ASA            | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| PA             | 0     | 0     | ✓     | 0     | 0      | 0     | 0      | 0      | 0     | 0     | 0     | 0     | 0     | 0      | 0      | 0     | 0     | 0     | ✓     |
| PBT            | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| PBT+PC         | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| PC             | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| PC+ABS         | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| PC+PBT         | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| PE             | x     | x     | x     | x     | x      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| PET            | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| PMMA           | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| POM            | 0     | 0     | 0     | 0     | 0      | 0     | 0      | 0      | 0     | 0     | 0     | 0     | 0     | 0      | 0      | 0     | 0     | 0     | 0     |
| PP             | x     | x     | x     | x     | x      | x     | x      | ✓      | x     | x     | ✓     | x     | x     | ✓      | x      | x     | x     | x     | x     |
| PPO            | 0     | 0     | 0     | 0     | 0      | 0     | 0      | 0      | 0     | 0     | 0     | 0     | 0     | ✓      | ✓      | ✓     | x     | 0     | 0     |
| PVC            | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| SAN            | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |
| TPU            | ✓     | ✓     | ✓     | ✓     | ✓      | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     | ✓     | ✓      | ✓      | ✓     | ✓     | ✓     | ✓     |

✓ = good compatibility; x = not compatible; o = limited compatibility for low volume of minor material, < 2%

Failure to follow the manufacturers’ recommendations related to mixing compatibility, (table 1), leads to unwanted defects on the injected products. Such recommendations should be taken into account when:

- injecting a mix of polymers (e.g.: PC + ABS) prepared in the workshop ("in-situ"),
- raw material is mixed with recycled material,
- a granulated additive is chosen. The matrix additive or the carrier must be compatible with the material to which it is added.

Observation: even if PC and PA would have superposed ranges of processing temperature, according to the table 1, these materials, virgin or recycled, are not suitable for preparing blends due to the incompatibility at the interface of the phases of melted materials. However, an additive with PS, PPO or even PE as carrier could be added (no more than 2%) to the PC as “main material”, [18].

When polyethylene or different blends are used as raw material, pretreatment of this with prooxidant additives before injection can increase the biodegradability of the processed material, [15, 19]. As a design approach, this is a convenient and appropriate solution, ecofriendly and convergent to the concept of circular economy and a way to reduce the pollution of the environment with plastic material, [7, 20].
3. The aim of the paper

If the consumption of plastic material is increasing worldwide, the efficiency of the associated recycling system calls for systemic thinking and improved waste chain management with more efficient processes and technologies.

In the domain of plastic processing, the technological waste means:
- parts resulted from injection at the change of the raw material,
- injected products from the injection machine setup,
- samples for quality measurements,
- nonconforming products that do not meet the quality requirements, injection runners.

Generally, at the waste selection, it is recommended to sort them by type (nature) of material and color, [7, 8, 10, 20]. After selective collection, the high-density polyethylene (HDPE) waste, material referred to in the study implemented and presented below, is grinded to a 3-6 mm granulation, stored in a tight container and could be reused in the injection process under specific conditions, adapted to each product, depending on its quality requirements. The product injected from recycled material is a "handle", used for transport and handling, (figure 1), that is attached to a 5 liter PET bottle (figure 2). Mechanical strength, easy fastening on the bottle and ergonomically design are requirements always mentioned in the product quality sheet.

![Figure 1](image1.png)  
**Figure 1.** Handle design and working position.  

![Figure 2](image2.png)  
**Figure 2.** Handle applications.

The content of this paper is in the trend with the recommendations for reducing waste, even from the source, by recovery and reusing the material from technological waste, inherent in the injection manufacturing process, and reuse it to manufacture products of the same type or into alternative products according to the quality requirements.

The main objective of this experiment is to optimize the closed-loop recycling step, which have been taken into consideration in the management of the manufacturing process as an important factor, determinative, in the final financial balance, especially when recycled material is used exclusively.

The combinations of recycled material presented in the experimental study, (table 3), are used into the current production of an industrial unit for manufacturing products from HDPE, and the major concern was to find proper combinations that gives, after injection, parts that comply with the requirements for the mechanical resistance outlined for first use raw material products.

4. Materials and methods

Handles were injected using two materials of the same type, HDPE, but with different melt flow rates (MFR), from two different manufacturers. Even if the nature and chemical formula of the materials is the same, the different flow rate denotes longer or shorter macromolecular chains. A material with short macromolecular chains, subjected to a progressive increase in temperature, will switch faster to a molten state, so it will have a lower melting temperature. Large differences between the MFR values
of two materials in a mix denotes significant differences between values of melting temperature of these and immiscibility of the phases in the melt [2, 11].

Wastes from these materials have to be collected selectively, based on material type, and regardless of color, [8, 10, 12, 20, 21]. For the materials in the study, table 2, the MFR values show that the values of this parameter are within the limits allowed according to the datasheet, table 2, for each batch.

Table 2. Physical properties of the virgin HDPE, material A and material B.

| Material          | Nominal MFR for virgin material, [g/10 min] ± 10% | Density [Kg/m³] | Tensile stress at yield [Mpa] ± 5% |
|-------------------|-----------------------------------------------|-----------------|-----------------------------------|
| Material A: HDPE  |                                               |                 |                                   |
| BorePure MB 5569  | 0.8                                           | 956             | 26                                |
| Material B: HDPE  |                                               |                 |                                   |
| Eltex Superstress HD 6070 EA-B | 7.6       | 960             | 31                                |

As can be seen in table 2, the two materials have very close value for the density but the MFR values are significantly different and the tensile stress at yield is 20% higher for Material B compared to Material A.

The results of the MFR values are presented in table 3. Its fall within the approved limits according to the specifications on the raw material datasheet, for the recycled material A or B. For grinded blends of different materials in equal proportions (1:1, 50% + 50%), the flow rate is below but very close to the arithmetic mean of the MFR reference values for virgin materials. The MFR determinations were performed with a Gottfert MI-3 device on sets of 16 samples for each type of recycled material or mixture experimented.

Table 3. Melt Flow Index (MFR) for recovered materials A and B.

| Recovered and regrinded material | MELT FLOW INDEX at 190°C and 2.16 kg load |
|----------------------------------|------------------------------------------|
|                                 | mean | min | max | results |
| blends of Material A type       | 0.761 | 0.754 | 0.767 | OK      |
| blends of Material B type       | 6.977 | 6.929 | 7.062 | OK      |
| regrinded A+B (50/50%)          | 3.203 | 3.105 | 3.353 |         |

The experimental study was performed on five sets of 16 handles were injected using different combinations of recycled mixtures, A and B, following recipes as seen in Table 4.

Table 4. The recipe for blends of recycled materials injected in the five sets of handles in the study.

| Set of tests | Ratio of recycled A, [%] | Ratio of recycled B, [%] |
|-------------|--------------------------|--------------------------|
| A100        | 100                      | ---                      |
| A90B10      | 90                       | 10                       |
| A50B50      | 50                       | 50                       |
| A10B90      | 10                       | 90                       |
| B100        | ---                      | 100                      |
Then, these parts were submitted to tensile tests, forces and elongations at break were recorded using a testing machine specially designed in order to reproduce as accurately as possible the normal conditions of use [22]. The device consists of two tensile components (figure 3). The holding ring, attached to the testing machine baseplate, is designed to reproduce the grip area of the PET bottle (figure 4). The handle is attached to the mobile part of the testing machine and simulates how the human operator grabs the handle.

For the evaluation of the mechanical properties of the materials in the study it was used a Tinius Olsen H25KT testing machine (figure 5). In the regular environmental conditions in the laboratory (temperature: 21.60 °C and humidity: 47.50%, [3, 5, 22], the handles were subjected to tensile test at the speed of 50 mm / min until they break, figure 6. The reference for the resistance at break, nominal value, is specified to be at 500N (mentioned in the quality datasheet) and the minimum allowed breaking force have to be no less than 400N. The maximum value is less important for this parameter but it can be roughly considered to be at 700N.

The handles are injected in flat configuration not in working position (figure 1) in which they fulfill their functional role. This justified by design considerations of the mold and overall cost restrictions, and it is based also on considerations related to the next technological phases: orientation and application on the automated bottling lines, packaging, handling and transport optimization.

After the handle is applied on the PET bottle, it deforms when the container is grabbed. The handle is folded at 90 degrees from the holding ring already attached to the neck of the PET bottle. So, the material at the base of the handle is bent so that the part from the bending center is compressed and the material is subjected to a tensile force on the opposite side. When lifting the container, the tensile
force, that overlaps these strains, relaxes the material tension on the side of the bending center. At the same time, it increases the tension on the opposite side. This is the area where the rupture will start, after some whitening appear on the surface of the material.

5. Results and discussions
Measurements were made for each set of 16 handles resulting from an injection cycle, from a mold with 16 cavities. The values found for the breaking forces and elongations are presented in table 5.

| Injected mat. | A100 Breaking force, [N] | A90B10 Breaking force, [N] | A50B50 Breaking force, [N] | A10B90 Breaking force, [N] | B100 Breaking force, [N] |
|---------------|--------------------------|---------------------------|--------------------------|--------------------------|--------------------------|
| Cavity no.    | Elong. [mm]              | Elong. [mm]               | Elong. [mm]               | Elong. [mm]               | Elong. [mm]               |
| 1             | 578 37.1 612 37.8 556 35.1 507 44.7 565 50.2 |
| 2             | 625 39.1 618 40.9 579 31.1 483 22.9 666 74.3 |
| 3             | 581 24.9 528 34.2 514 34 578 39.8 584 43.2 |
| 4             | 539 35.2 451 33.9 483 32.9 519 30.5 533 43.8 |
| 5             | 557 38.1 505 35.4 538 34.8 507 38.8 543 53.6 |
| 6             | 478 31.7 523 33.5 483 30.2 453 35.3 569 57.6 |
| 7             | 616 34 616 39 598 38.7 556 48.4 592 56.8 |
| 8             | 598 40.4 631 40.6 535 31.7 588 31.4 648 67.2 |
| 9             | 595 29.7 600 38.8 520 31.2 582 39.8 644 49.5 |
| 10            | 575 35.1 612 39.3 579 33.5 568 43.3 601 42.3 |
| 11            | 545 34.7 545 29.7 493 28.9 494 30.9 592 53.1 |
| 12            | 495 30.7 517 31.7 451 30.9 518 45 563 49.3 |
| 13            | 518 33.1 483 31 513 30.5 493 39.5 518 49.4 |
| 14            | 513 31.8 491 32.1 544 37.9 543 29.3 556 44.6 |
| 15            | 597 41.1 618 40.2 481 26.1 455 31.7 579 26.2 |
| 16            | 600 38.3 619 41.2 538 27 541 39 643 55.2 |

The forces v. elongation curves have been generated for each measurement, as shown in figure 7.

![Figure 7: Forces v. elongation curve.](image-url)

Process capability report for the values of forces at break was established by using Minitab 18 software, [23], as shown in the figures below.
Figure 8. Process capability report for breaking forces of A100 set.

Figure 9. Process capability report for breaking forces of A90B10 set.

Figure 10. Process capability report for breaking forces of A50B50 set.
As seen in figure 8, test set A100 had the best capability (Cpk value close to 1.33) for handles injected from 100% recycled material A, only.

**Figure 11.** Process capability report for breaking forces of A10B90 set.

**Figure 12.** Process capability report for breaking forces of B100 set.

**Figure 13.** Average values of force at break for the five sets in the study.
For the set of tests A90B10 and B100, the capabilities are 1.15 and 1.05 (see figures 9 and 12) and for the set of tests A50B50 and A10B90 the capabilities are around 0.9. All the experiments carried out show that the experimental values obtained are relatively scattered but the most important aspect is that all the results are within the limits of the value of breaking force outlined in the product quality datasheet. Given the applications in which these handles are used, there are no functional risks that could affect the user’s safety while using these products and this situation is accepted as such.

The average values of tensile force at break, for the five sets in the study, are presented in figure 13. The data for the elongations was not processed because this parameter is not important in fulfilling the functional role of the product studied. An elongation of the handle material is accepted if the material does not break.

A more detailed analysis of the tensile properties for larger range of grinded blends could significantly increase the degree of recycling of plastic waste at source. The authors of this article continue to carry out studies based on determinations of Tensile Stress at Yield, Tensile Strength at break, Elongations, Testing speed influence, e.a, for plastic parts from current production, injected from virgin materials or with recycled added.

6. Conclusions
An important conclusion drawn from the ones presented above is that all the values for tensile force at break, for the five experimental sets of injected products in the study, are higher than the 400N, limit imposed in the quality sheet for the product so called in the paper "handle".

Also important is to note that, as raw material in the study, it was used only recycled material prepared according to the recipes in table 4 (A100, A90B10, A50B50, A10B90 and B100)

The set B100 (100% recycled material B) had the best results, followed by A100 set (100% recycled material A), which may lead to the following conclusions:

- if the mechanical strength is important for the functionality of the product and it is mentioned as a requirement in the product quality sheet, up to 100% recycled HDPE material can be used without affecting the tensile force of the injected product. But in such cases, special attention should be paid to the sorting of recovered plastics, on batches of material, without mixing the grades between them (even if the same type of material is handled, see the case of HDPE, in the study) and by color if the case of light color products to be manufactured. Dark colored products (black, dark blue, etc.) can go with recycled material up to 100% while light colors can use maximum 10-20% recycled material mixed with raw material. In this case the selective collection of the waste by color is mandatory;

- if the tensile resistance is not important for the product functionality, the mixture of recycled material from different batches would be allowed, the sorting will be less restrictive and also the dosing of the recipe for preparing the raw material. This strategy can be successfully implemented when the recycling by downgrading is applied;

- less satisfactory results in terms of breaking force values were obtained for the parts injected from raw materials prepared according to recipes A50B50 and A10B90, figure 10 and figure 11, mixtures of recycled material in a recipe with 50 and 90% of material B, with higher MFR.

This recycling model can be extended to other products made of polymeric materials and can lead to a degree of recycling of the total amount of waste generated by the similar manufacturing sites, up to 95%.

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