Characterisation of Filled and Recycled PA6

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Summary: Filled PA6 are important representatives of engineering plastics used in automotive components. Nowadays, the demand of plastic recycled grades is increasing in this branch of industry but polymer recycling can undergo thermomechanical degradation processes with the results of a poor secondary material, regarding its properties. In this paper an investigation of thermal, mechanical (tensile, flexural and impact tests) and rheological properties of a sample of recycled and filled PA6, is reported as a function of the number of reprocessing operations (3 times) and of the fraction of recycled material (15, 30 and 50%) added to the virgin material. Recycled PA6, used in this study, comes from fibre grade production waste. Material was filled with 20% glass beads and 10% glass fibre, according to the specifications of the application, mainly to obtain a lower shrinkage in the end product. This work also shows that the mineral fraction, not being degraded during the injection process, allows better recyclability to the filled material. The properties of the recycled material remain below the virgin, and the best combination of both appears to be the mixture with 30w.% recycled fraction, which shows a lost of properties similar to 3 reprocessing operations.

Introduction

Nowadays, the environmental questions are becoming more and more important in the industrial world. Particularly for plastics processing companies, concern is mainly focused in the reduction and recycling of wastes generated during transformation processes such as off-spec resin, trims of moulded parts or poor quality parts. Part of the solution to this problem consists in the use of recycled materials to produce new products. Blending recycled polymers with the same virgin polymer is a common industrial practice to reuse plastic scrap.

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The offer of recycled plastics in the current market is becoming wider. Not only many different materials are available, but also many different qualities of each given type of plastic, which differ mainly from the previous sorting and washing process. The degree of purity will be determinant for the studied application. There is still a need to characterise the recycled material and evaluate its behaviour after recycling. Mechanical recycling of thermoplastic materials causes decomposition of the polymeric chains, leading to the loss of mechanical properties[1-3)].

Polyamides as the most important representatives of engineering termoplastics, are well known for a wide range of properties[4-5)]. Fibre grade production wastes are the main source for polyamide recyclates. Previous investigations have shown that unmodified polyamide materials are excellently suitable for utilisation by using mechanical recycling processes[6)], while the recyclability of specially modified types like glass fibre reinforced compounds is rather limited[6-7)]. However, the use of a recycled PA6 with the studied filler fraction (20% glass beads and 10% glass fibre), as far as we know, has not been reported.

This study investigates the recycling possibilities of a commercial PA6 (polyamide or nylon 6) recycled from fibre grade production waste, in order to substitute partially or totally the current material, a commercial virgin filled PA6, used in the production of automotive components.

In figure 1 the different stages of the investigation method is represented. The 1st stage consisted in the selection of several recycled materials in the market. These materials were characterised and compared with the virgin material. The recycled material with most similar properties to the virgin material was choosen for the investigation. In the 2nd stage the variation of the properties with different fractions of recycled material was studied. Finally in the 3rd stage the recycled material was reprocessed several time (until 3 times).

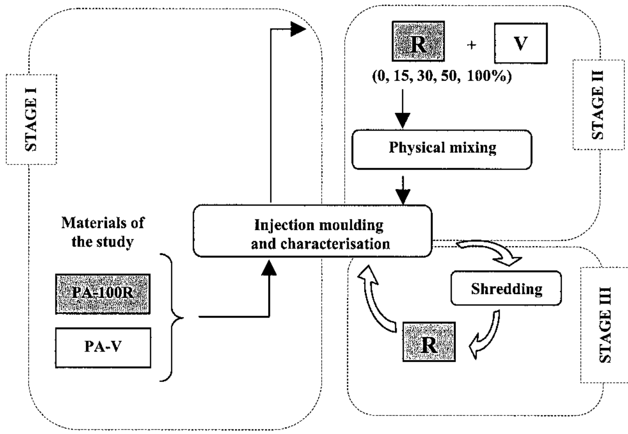

Fig.1 : Investigation method

Materials

The virgin material of this investigation is a commercial grade of polyamide 6 (PA6) filled with 20% weight glass beads and 10% weight glass fibres. This material is coded as PA-V.

In a previous study a market investigation was carried out to find the most similar recycled material (PA-100R) to the virgin material for this aplication. This material comes from different fibre grade production waste and mineral fillers (glass beads and glass fibres) were added by extrusion to obtain the same composition of the virgin material. This material was delivered by a local company specialised in compounding of recycled plastics.

The mixtures of virgin and recycled material are represented with the following code: PA-(weight % of recycled material)R.

Additionally, the variation of the properties with the number of processing operations was studied with scraps from injection moulding production plant. The aim was to evaluate the possibility to reuse the scraps in the injection moulding production plant. In this study industrial specimen were shredded and injection moulded. The resulting material, PA-I2, was characterised and the same process of shredding and injection moulding was repeated to obtain the material PA-I3.

The different formulation of PA6 were physically mixed before processing. Table 1 explains the codification of each formulation studied in this investigation.

Table 1. Codification and description of the studied materials.

Code	**Description**
PA-V	Virgin PA6
PA-15R	Mixture 15% PA-100R + 85% PA-V
PA-30R	Mixture 30% PA-100R + 70% PA-V
PA-50R	Mixture 50% PA-100R + 50% PA-V
PA-100R	PA6 100% recycled
PA-I2	Virgin PA6 reprocessed and granulated twice
PA-I3	Virgin PA6 reprocessed and granulated 3 times

Experimental Part

Injection Moulding Processing

PA6 materials were dried at 80°C for 4 hours previous to the injection moulding, that was carried out in a Mateu & Solé Meteor 440/90 injection moulding machine. The mould was heated at 80°C and provided dumbbell and prismatic specimens for tensile and impact/flexural test respectively.

Characterisation

Mechanical tests samples were submitted to humid environment to reach 2.1% (±0.2) water absorption.

Tensile tests were performed according to ASTM D-638M-91a and flexural tests according to ASTM D-790M-93, both at room temperature in a Galdabini 1890 Sun 2500 universal testing machine with a load cell of 25 kN and a videoextensometer. Tensile tests were carried out at a crosshead speed of 5 mm/min and flexural tests 2,8 mm/min.

Charpy impact tests were carried out according to ASTM D-256 at room temperature in a CEAST pendulum with 2 J capacity providing an impact speed of 2.9 m/s on specimens notched after processing.

The viscosity measurements were carried out according to ISO 307 standard.

Melt flow index tests were carried out in a CEAST machine with an applied total charge of 49,05 N and a test temperature of 230°C. Samples were dried in an oven at 80°C for 4 hours.

Results and Discussion

Due to confidential reasons, values are shown in relative units related to values of PA-V equal to 100.

Tensile Properties

In figures 2, 3 and 4 are represented the tensile properties with significant importance in this study: Young modulus (E), maximum tensile stress (σ_{max}) and elongation at break (ε_b).

Influence of the Proportion of Recycled Material

Young modulus and maximum tensile stress decrease already with a small proportion of recycled material and this trend is stopped by 50 w.% recycled material, while elongation at break keep decreasing until reaching a decrease of 36% for the PA-100R. The material PA-50R has a slightly lower maximum tensile stress than PA-100R; this is probably due to a certain incompatibility between PA-V and PA-100R. These results usually appear when studying blends of a virgin and recycled sample of the same polymeric material.

Influence of the Number of Reprocessing Operations

There is a decrease of 28% in the maximum tensile stress and 11% in the Young modulus after 3 reprocessing operations. Elongation at break shows an unexpected behaviour as the material PA-I3 has a higher value than PA-I2.

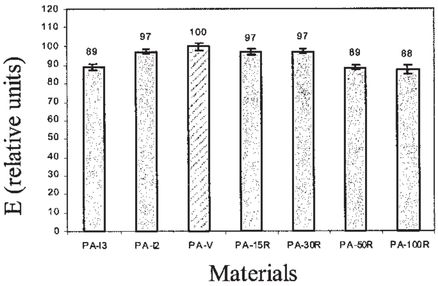

Fig. 2: Young modulus (E)

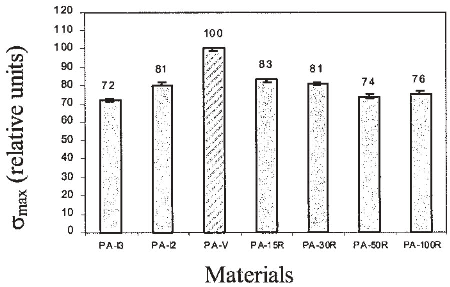

Fig. 3: Maximum tensile stress (σ_{max})

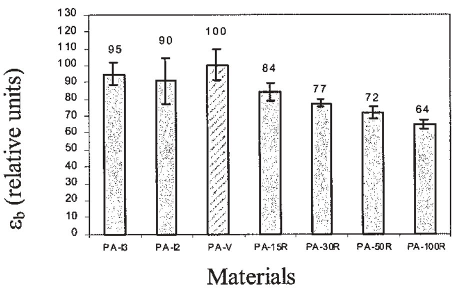

Fig. 4: Elongation at break (ε_b)

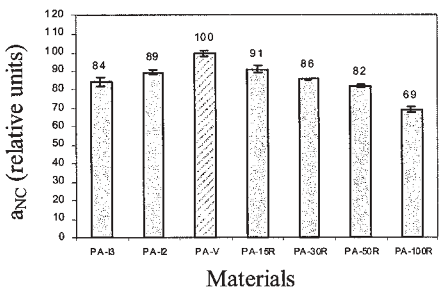

Fig. 5: Impact strength (a_{NC})

Impact Properties

In figure 5, results of impact tests are represented. With a similar trend observed in tensile and flexural properties, impact strength decreases with an increasing fraction of recycled material and an increasing number of recycling operation processes. Impact strength gives the most important difference between virgin and recycled material, where the 100 w.% recycled material reaches a decrease of 31% related to the virgin material. After 3 recycling operation processes the material PA-I3 shows a decrease of 16%.

Flexural Properties

Results of flexural tests are shown in figures 6 and 7. The variation of elastic modulus (E_f) and maximum flexural stress (σ_f) have a similar behaviour than tensile properties.

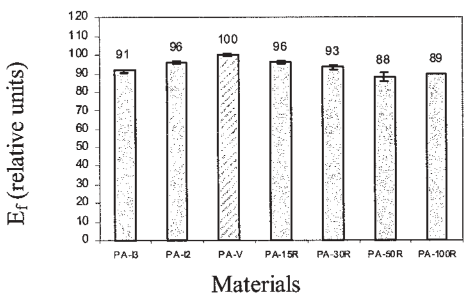

Fig. 6: Elastic modulus (E_f)

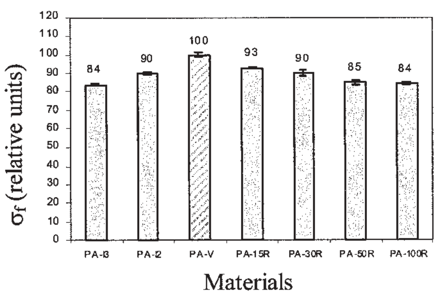

Fig. 7: Flexural stress (σ_f)

Viscosimetry and Melt Flow Index (MFI)

Figure 8 shows that recycled material (PA-100R) has a 37% lower intrinsic viscosity [η] than virgin material (PA-V). This is probably due to the fact that the recycled material is a different PA6 grade and has endured previous termomechanical degradation processes. The material PA-50R was obtained from physically mixing PA-V and PA-100R, without any thermal operating process. As it was expected, PA-50R has a [η] average value of both materials.

There was a decrease of about 19% in the intrinsic viscosity of the material after one injection moulding process. This variation is related with a decrease in molecular weight of the polymer, due to thermomechanical degradation by chain scission. This is of significant importance to the mechanical performance of the moulded parts because mechanical properties are directly related to molecular weight of the polymer.

In figure 9, results of the MFI test are similar to those of intrinsic viscosity. The fluidity increases with an increasing number of recycling operation processes and an increasing proportion of recycled material.

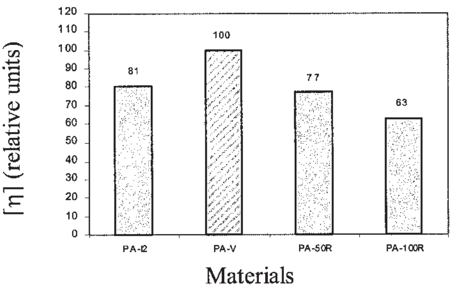

Fig. 8: intrinsic viscosity [η]

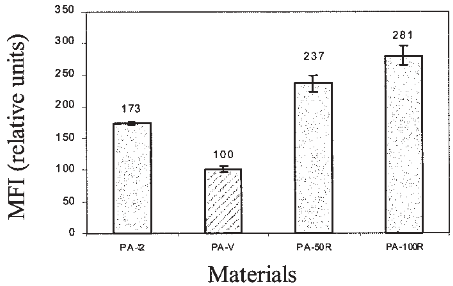

Fig. 9: MFI

Conclusion

- A lost of mechanical properties is shown with an increasing proportion of recycled material. Most sensitive properties are maximum tensile stress, elongation at break and impact strength.
- A lost of mechanical properties is shown with an increasing number of recycling operation processes. Most sensitive properties are maximum tensile stress, flexural stress and impact strength.
- The recycled material shows a lower intrinsic viscosity than the virgin material, due to the previous use of the recycled polyamide coming from fibre production waste.
- The best combination of recycled and virgin material appears to be the mixture with 30 w.% recycled fraction, which shows a lost of properties similar to 3 reprocessing operations.
- This work also shows that the glass filler fraction, not being degraded during the injection process, could allow better recyclability to the filled material. More studies in the future will focus on that point.

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