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Study of the Technical Feasibility of the Use of Polypropylene Residue in Composites for Automotive Industry

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Abstract

Polypropylene (PP) is widely used in short-term use artifacts, rapidly discarded and should partially replace neat PP. In addition, it is one of the polymers most used in the automobile industry. This study shows the technical feasibility of partially substituting neat PP for a post-consumer counterpart (PP_r), as well as adding ground glass (GP), used as filler in the polymer matrix. Mechanical and thermal properties of the recycled blends (PP/PP_r) and composites (PP/PP_r/GP) were evaluated. The results demonstrated that the blend with the highest PP_r content obtained a statistically significant decline in elastic modulus, but adding 5 wt% of GP to this blend increased this property, achieving a similar value in relation to neat PP. The composite developed may be a promising tailor-made product with properties resembling those of the virgin plastic. Thus, the automotive industry seems to be a good option for the use of PP_r and GP composites and blends, without increasing product requirements.

Keywords: recycling, polypropylene, automotive, industry, PP, glass powder, composite

1. Introduction

The circular economy promotes sustainability by combining the hierarchy of reduction, reuse and recycling, considering economic and environmental issues [1]. The use of post-consumer materials to manufacture new ones for a new production cycle to minimize the waste of natural resources is the goal of the circular economy [2–6]. The same can be said of plastic, an important class of materials that meets many of society's needs.

Plastics are present in our daily life under different forms and applications, such as office supplies, toys, footwear, civil construction, electrical and electronic components, aerospace, food, the medical and textile industries, packaging, paint

and varnish, as well as the automotive industry, among others. This array of applications is due to their desirable properties in a range of sectors [7, 8].

However, not all plastics return to the production cycle after use. In 2015, only 9% of plastics produced worldwide were recycled, 12% incinerated and the rest buried in landfills [9]. Recycling causes less environmental impact, as reported by Bernardo et al. [10], who assessed recycling in terms of global warming and total energy use, concluding that plastic materials generally display environmental and economic advantages over conventional materials throughout their life cycle, from raw material extraction to synthesis, transformation, transport, use, recovery and destination. Duval and Maclean [11] also found a decline in greenhouse gas emissions and energy required during recycling [12]. Mechanical recycling involves the addition of virgin or recycled material to maintain properties [13].

Plastics can be recycled in different ways, including mechanically, chemically and energetically. Chemical recycling involves physical processes, such as remolding [14–16], and the final product is a monomer or oligomer that can be used in the synthesis of other products. In energy recycling, the energy released from the burning of waste material is reused [14].

Closed-loop recycling occurs when the recycled material replaces the virgin material in the same production cycle as the original product [17, 18]. Open-loop recycling is when the recycled product is used in a different production cycle, that is, the product to be recycled is used to manufacture a product different from the original [17, 19, 20].

A crucial point to stimulate recycling is the search for a different market for the recycled material and more environmentally sustainable processes. Traditionally, recycled products compete with virgin material, which may hinder their market entry. Scientific studies that focus on recycling should also seek to obtain more economically feasible and technically useful recycled products.

The most widely used and manufactured plastics are high-density polyethylene (HDPE), polypropylene (PP), low-density polyethylene (LDPE), poly(ethylene terephthalate) (PET), poly(vinyl chloride) (PVC) and polystyrene (PS) [9]. In the automotive industry, PP is used to manufacture the following items: car trunk lids; battery trays and boxes; heater boxes; tool boxes; seat belt buckling boxes; rear view mirror boxes; electric junction boxes; hubcaps; carpets; battery guards (protection against short circuit); steering wheel covers; shock absorber covers; vacuum hoses; air hoses; consoles; bumpers; glove boxes, among several other uses [21–24].

To comply with the main technical demands of automobile manufacturers, PP compounds must exhibit a suitable balance between stiffness and tenacity, with good thermal resistance, as well as fewer imported raw materials, thereby achieving more competitive prices. In addition to these properties, PP shows good processability [24–26].

An important supplier of materials to the automotive industry is the industrial sector responsible for manufacturing laminated and tempered glass used in motor vehicle windows (laminated glass for windshields and tempered for the other windows). However, in the tempering and laminating processes an industrial residue consisting of glass powder is generated and disposed of in landfills, with no specific use for this material [27]. In addition to the origin of glass powder in laminating and tempering processes [28], the windows that are removed from automobiles are also discarded when they cannot be reused. In such cases, these parts can be collected and recovered, then submitted to separation processes (polymer protection film) and grinding. The glass powder produced can be incorporated into polymer materials, resulting in composites with different properties [29].

Incorporating mineral loads into PP has been the object of studies on the production of materials with different properties [30, 31]. Improving the properties of the final product depends on the type of load, particle size of the mineral load being used and degree of dispersion of these particles in the polymer matrix. The most widely used commercial mineral loads are talcum and calcium carbonate [32–34].

This study describes the addition of glass powder to a PP matrix in order to obtain reinforcement properties and compare them with those of conventional composites. The aim is to acquire different properties in the polypropylene composites and reuse a residue (in this case, glass powder). We also assessed the effect of adding recycled polypropylene on the final properties of composites in order to reuse both industrial (glass powder) and urban residue (PP recycled from packaging).

2. The use of polypropylene in a composite or mixture

Polymers have been increasingly used in a number of applications as a substitute for traditional materials such as metal and ceramic, as homopolymers; formulated with additives, in the form of mixtures and polymer composites; or simply for their different properties, such as lightness, low transformation cost, resistance to corrosion, optimal thermal and electric insulation and easy conformation into complex shapes [33].

In general, the mechanical properties of polymers are not suitable in a number of applications owing to their lower resistance compared to metals and ceramics. However, the thermoplastic industry is growing due to ecological issues, in addition to the promising potential of these materials as mixtures or a composite matrix [35].

Compound systems formed by the combination of two polymer materials (mixtures) or a polymer material and a load (composites) are of significant technological interest due to the cost–benefit ratio. In both cases, the material consists of a continuous (matrix) and disperse phase, whose properties depend on good interaction between them [35].

The properties of interest for the automotive industry can be modified with studies on improving the polymer matrix, load, and polymer-load interface, among others. The interface is a link between the surface of the load and the matrix, and since the matrix receives the reinforcement, there is close contact between them, and there may or may not be adhesion. For a same combination of materials, different adhesion mechanisms can occur, such as mechanical, chemical, and electrostatic adhesion and by interdiffusion. The degree of reinforcement or improvement in mechanical behavior depends on a strong matrix-particle interface bond [36, 37].

The stress–strain behavior of many reinforced polymers or plastics can be changed by adhesion promoters and interfacial coupling agents (such as maleic anhydride) that alter adhesion and the nature of the matrix-load interface [38].

Polypropylene (PP) is a recyclable thermoplastic, that is, it melts when heated and hardens again when cooled, in a reversible process. Moreover, PP is easily mixed, primarily with organic reinforcing loads such as natural or inorganic fibers, including calcium carbonate, clay and talcum, and is widely used in structural applications [39–41].

The use of modified PP, especially for applications in the automotive industry, requires a suitable balance between stiffness and tenacity. In this scenario, the process of incorporating elastomeric materials, as well as mineral loads such as talcum and calcium carbonate (CaCO_3), into the PP matrix has been widely used to achieve different properties [42, 43].

Nanofillers, such as silica and calcium carbonate nanoparticles, have been added to improve the final properties of the PP matrix [44, 45].

2.1 Use of glass as an additive to the composite

The use of glass in a polypropylene matrix has been extensively studied and employed its glass fiber form in materials in which mechanical properties such as tensile strength and resistance to impact are important [46, 47].

There are several groups of glass, including silica, oxynitride and phosphate, but the first is the most important raw material used in composites [48]. Short E-glass fibers, obtained from a mixture of Si, Al, B, Ca and Mg oxides, are normally used as reinforcement for thermoplastics due to their low cost when compared to aramid and carbon [49], in addition to better impact strength and stiffness [50].

The interfacial interaction of glass composites with a thermoplastic matrix is often very weak. Particularly with polyolefin polymers such as polypropylene, there is little or no chemical reaction between the glass and the matrix. The interest in polypropylene for applications as a matrix in composites has been growing and the adhesion of this nonpolar polymer to the glass surface, which is also nonpolar, is a daunting challenge [51, 52].

In addition to the use of glass fiber, there are also glass microsphere applications [53]; however, residual glass powder remains a poorly explored load as reinforcement.

2.2 Environmental justification for polypropylene and window glass, materials contained in automobiles

Initiatives to develop more sustainable technological innovations and ecologically responsible management programs have been driven by a growth in environmental awareness and increasingly rigid legislation. The accumulation of plastic waste caused by the increase in per capital consumption of thermoplastic resins has prompted enormous research and efforts to substitute traditional thermoplastics [54].

To improve the production process, it is necessary to diagnose the flowchart of the process and manage inputs (water, energy, raw materials, etc.) and outputs (products, residues, effluents, atmospheric emissions, etc.). In general, inputs are natural resources that often cause environmental impact, such as ecosystem destruction, atmospheric pollution, etc. Outputs are environmental liabilities created by activities and residual materials (solid, liquid or gas) that, if not suitably managed, may cause permanent environmental impacts [55].

A sustainable production process contains a circular flow, where outputs are reintegrated into the process, which reduces impacts and costs in the generation of inputs and the destination and treatment of outputs. Recycling is an example of this type of sustainable production strategy and is therefore an attempt to reuse the material, natural resources and entropy expenditures in the production of a solid residue, reintroducing it into a new production process, thereby transforming the output of a process into the input of the same or another process [55].

The automotive industry is attempting to transform the car into a more sustainable and efficient product, not only in terms of the environment, but also from the consumer's financial standpoint. As such, the automotive industry has been working within the so-called DFE (Design for the Environment), that is, designing for the environment and introducing environmental variables in all the production strategies of the factory, such as product design (automobiles and parts), the process (manufacture of parts and assembly) and associated technologies (material treatment, painting, etc.) [28].

It is important to underscore that all participants in the life cycle of a product have shared responsibility. Thus, manufacturers, importers, distributors,

merchants, consumers and public cleaning concessionaires should promote the reuse of solid residues, transfer them to the production chain, reduce residue generation and encourage the development of products derived from recycled materials. The automotive industry, like all companies, is responsible for the entire process, from acquiring raw materials to discarding components, such as bumpers. Moreover, polypropylene is present in many automobile components.

3. Materials and methods

3.1 Materials

The grade of the Virgin polypropylene (PP) was H 605. The values for the PP properties presented in **Table 1** were provided by the Braskem Company.

The post-industrial polypropylene (PPr) was a washed and ground material supplied by the Poli Injet Company (Brazil), while the residue came from the packaging industry. The properties of PPr are: melt temperature (T_m) of 161°C, crystallinity degree (χ_c) of 32%, and melt flow index (MFI) of 4.83 g/10 min (230°C/2.16 Kg). The methodologies to evaluate these properties are described below.

The glass powder (GP) used in this study was supplied by the Massfix Company (Brazil) and is ground from windshield scraps. **Figure 1** shows the morphology of GP samples examined under a scanning electron microscope (SEM). The GP sample is composed of irregular-shaped particles with a broad size distribution.

Polypropylene modified with maleic-anhydride (PPMA), and Polybond 3200 with 1 wt% of maleic anhydride (MA) were supplied by the Chemtura Company (USA). The melt flow rate specified by the supplier is 115 g/10 min at 190°C under 2.16 Kg.

3.2 Blend and composite preparation

Table 2 shows the compositions of the blends and composites. GP, PP, PPr, and PPMA were dried in an oven at 60°C for 24 h before extrusion. Next, each composition was processed in a twin-screw extruder (TeckTril, L/D = 36, screw diameter = 20 mm) at a screw speed of 400 RPM and temperature profile of 90/120/150/160/185/200/220/240/260/260°C. The materials underwent injection molding to produce appropriate specimens for stress and impact strength tests, which were performed according to ASTM D638 and ASTM D256 standards, respectively. Injection molding was carried out in an Arburg 270 S injection machine, using a

Properties	ASTM standard	Property value
Density, g/cm ³	D 792	0.905
MFI (230°C, 2.16 Kg), g/10 min	D 1238	2.1
Flexural modulus (1%), MPa	D 790	1600
Yield stress, MPa	D 638	37
Rockwell hardness (R)	D 785	101
Impact strength (Izod, 23°C)	D 256	45

BRASKEM company data sheet.

Table 1.
Polypropylene H605 properties.

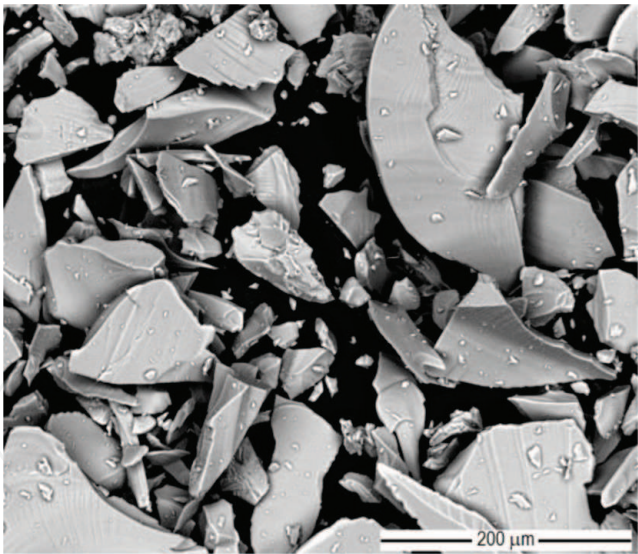


Figure 1.
SEM micrographs of glass powder (GP) particles.

Component (%wt)	PP	PPr1	PPr3	PPr1/ GP	PPr3/ GP	PPr1/ GP/ PPMA	PPr3/ GP/ PPMA
PP	100	90	70	85	65	75	55
PPr	—	10	30	10	30	10	30
GP	—	—	—	5	5	5	5
PPMA	—	—	—	—	—	10	10

Table 2.
PP, PPr, glass powder and PPMA blends and (or) composites composition.

temperature profile of 210/215/220/230/230°C. The injection and molding pressures were 1000 and 180 Bar, respectively. The mold temperature was 30°C, with a cooling time of 30 seconds.

3.3 Material characterizations

Tensile properties were measured using a universal testing machine (EMIC, DL3000) based on ASTM D-638. Izod pendulum impact resistance was determined using a CEASt Resil Impactor tester based on ASTM D256. The data related to all the mechanical properties were based on the average of eight tested specimens.

Melting temperature (T_m), melt enthalpy (ΔH_m) and crystallinity degree (χ_c) of the materials were determined using a Differential Scanning Calorimeter (STA 6000, Perkin Elmer) during the second heating scan. Samples weighing between 25 and 30 mg were heated from room temperature to 300°C at a heating rate of 10°C/min (first heating scan). The temperature was then lowered to 30°C at a heating rate of 10°C/min, and the samples submitted to a second heating scan under the same conditions as the first. Crystallinity degree was calculated using Eq. (1).

$$\% \text{ Cristalinidade} = \frac{\Delta H_f}{\Delta H_{100\%}} \times 100 \tag{1}$$

where ΔH_m is the endothermic enthalpy, ΔH_{100%} the theoretical melting enthalpy of 100% crystalline PP (209 J/g) [56], and wt% the amount of PP in the blend or composite.

The cryogenic-fractured surface morphology of the materials was examined under a scanning electron microscope (SEM, FEI, Quanta 400, accelerating voltage at 25 kV, 800X). The fractured samples were coated with gold.

3.4 Statistical analysis

The statistical analysis of the results was performed using STATISTICA 6 software. Analysis of variance (ANOVA) was applied to test for significant differences between the means. Residual normality and homogeneity of variances (Cochran C and Bartlett methods) were determined before univariate tests of significance and Fisher's least significant difference (LSD) test, using a significance level of $\alpha = 0.05$.

4. Results and discussion

4.1 Morphological and thermal characterization

Figure 2 shows the cryogenic fractured specimen for comparison between PP, PPr1/GP, and PPr3/GP. Poor interaction can be observed between the polymer matrix and glass particles due to the presence of small voids, gaps, and unattached particles.

The addition of PPMA in GP/PPr composites resulted in the smoothing of fractured surfaces (**Figure 3**). Furthermore, unattached particles, micro-voids and the gap between the matrix and the filler were slightly reduced. As such, filler-matrix interaction improved due to the addition of the coupling agent. **Table 3** shows the thermal properties of composites containing GP.

The melting temperatures (T_m) of all samples were similar to those of neat PP, except for the presence of a small endothermic peak in some composites at 127°C. Based on literature data [57], the small peak at 127°C can be attributed to the polyethylene and contamination in PPr, which is very common due to the difficulty in separating PP from PE during the recycling process.

In general, the composites showed similar crystallinity degrees (χ_c) to those of neat PP (**Table 3**). The results are noteworthy because they suggest that the presence of post-consumer materials (PPr and GP) did not disturb the crystal formation of the final composite, which leads to the assumption that the final properties of the composite are maintained, even with the addition of post-consumer materials.

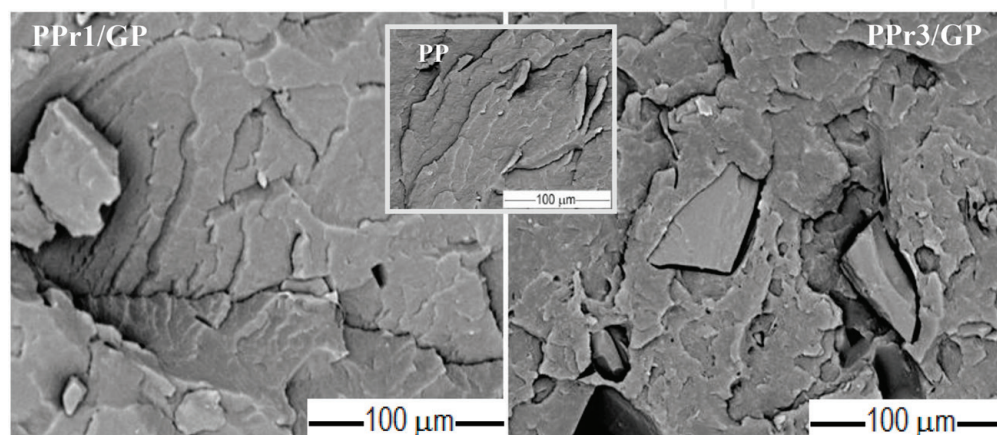


Figure 2.
SEM micrographs of neat PP, PPr1/GP (90/10%w/w) and PPr3/GP (70/30%w/w) composites.

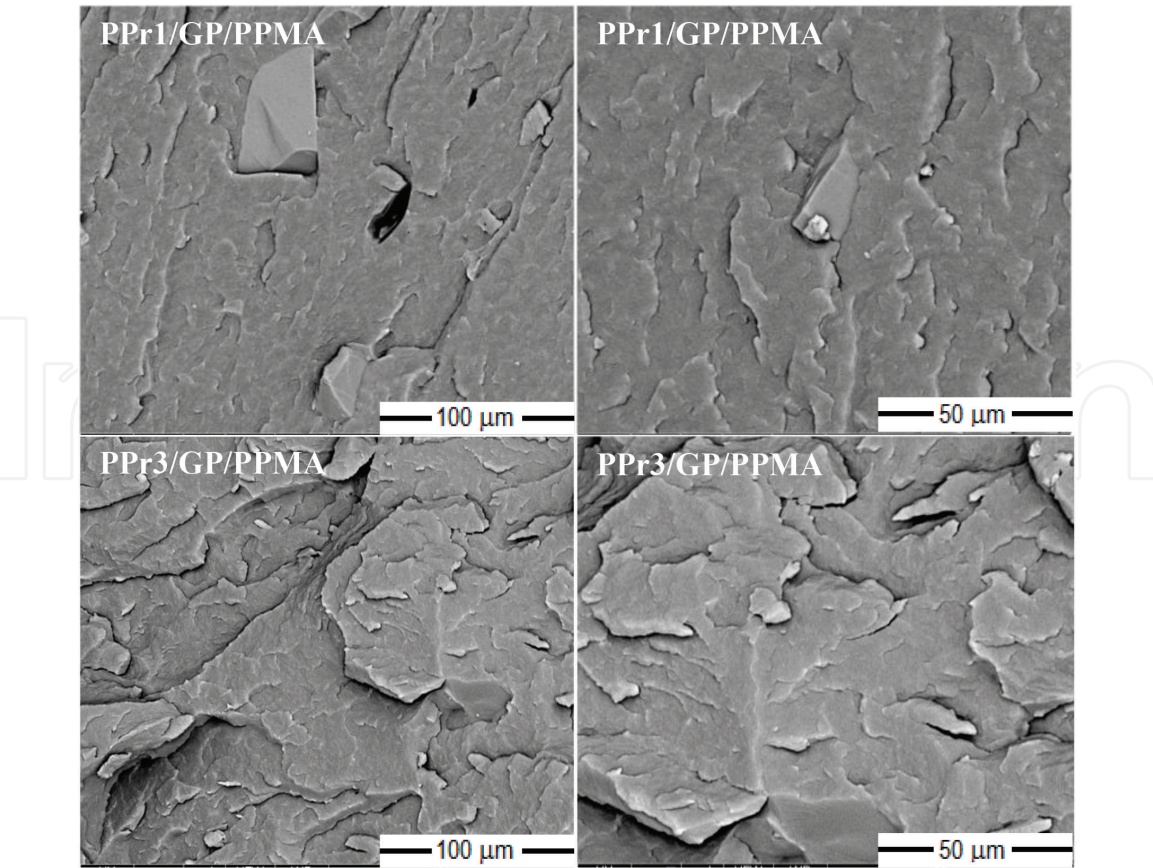


Figure 3. SEM micrographs in two sizes (100 and 50 μm) of PP/PPr1/GP/PPMA (75/10/5/10%w/w/w/w) and PPr3/GP (55/30/5/10%w/w/w/w) composites.

Property	PP	PPr1/GP	PPr3/GP	PPr1/GP/PPMA	PPr3/GP/PPMA
Tm ($^{\circ}\text{C}$)	166	165	127/165	126/165	126/164
ΔHm (J/g)	81.6	88.0	72.6	68.8	72.6
χc (%)	39	44	37	35	37

Table 3. Thermal properties of PP and PP/PPr/GP-base composites.

4.2 Mechanical properties

Material tensile properties are important for both engineering and packaging applications, since they represent the ability of materials to withstand the load transferred in the longitudinal direction. Impact strength is essential to engineering applications, due to the need to bear high loads for very short periods of time. Thus, tensile properties are vital in evaluating bags or mooring ropes, while impact strength is a critical feature in recycled plastic fencing, furniture and automobile parts. Tensile properties, mean values and confidence intervals (Fisher's LSD test) are shown in **Figures 4–7**.

With respect to elastic modulus (**Figure 4**), a comparison between PPr1, PPr3, and PP showed that elastic modulus, which is related to composite rigidity, tends to decrease with the addition of PPr, but this effect is only significant for the composite with 30%wt of PPr. However, when recycled filler (GP) was added to PP/PPr blends, the modulus rose until the mean composite modulus values were equal to those of virgin PP. This finding suggests that the previous decline in stiffness observed in PP/PPr blends can be solved by adding glass powder.

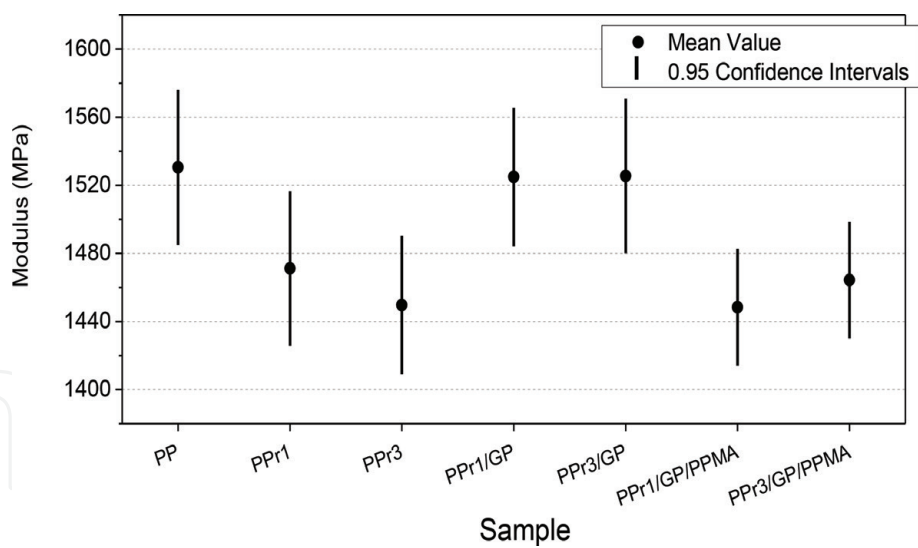


Figure 4.
Elastic modulus of PP and PP/PPr/GP-base composites.

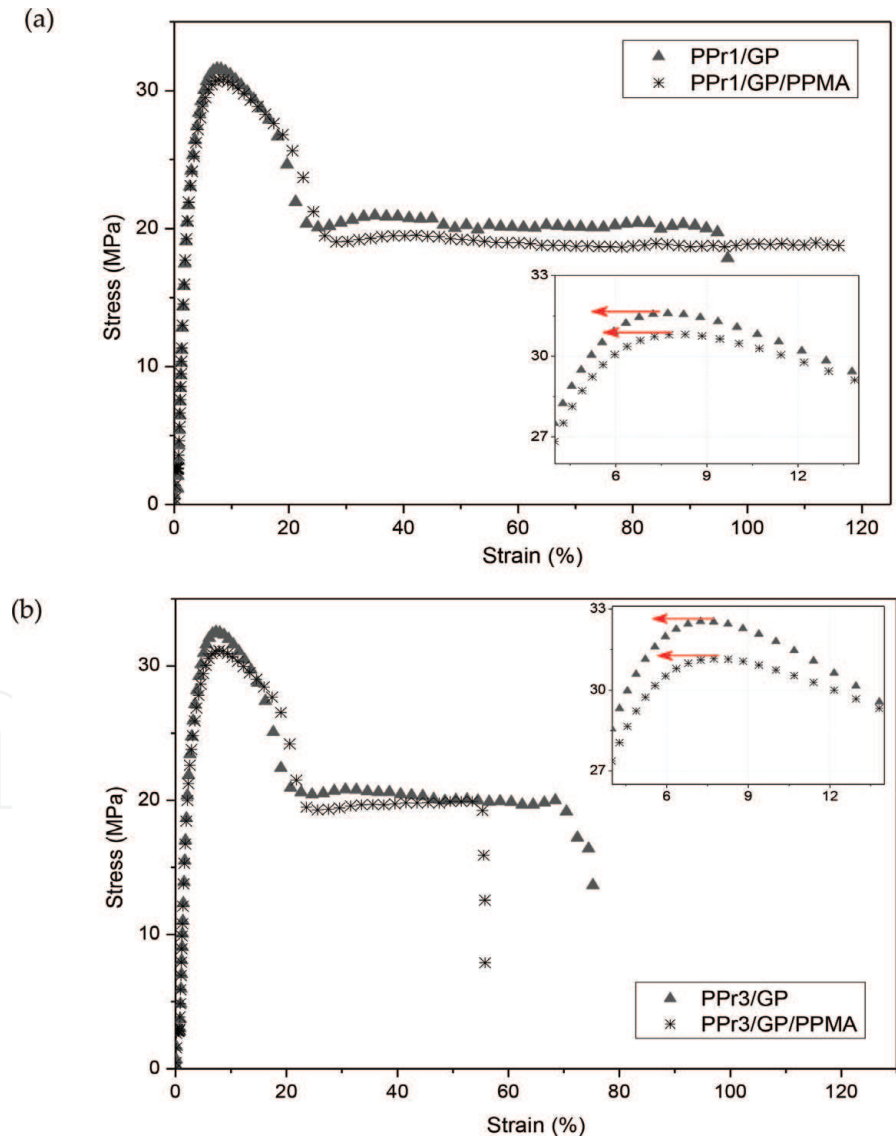


Figure 5.
Stress x strain curves of composites samples with: (a) 0% and (b) 10% of PPMA.

By contrast, the improvement in elastic modulus was reversed when PPMA was added to PP/PPr/GP composites. This is significant because good cohesion between the matrix and GP, as shown in **Figure 3**, was expected to improve the

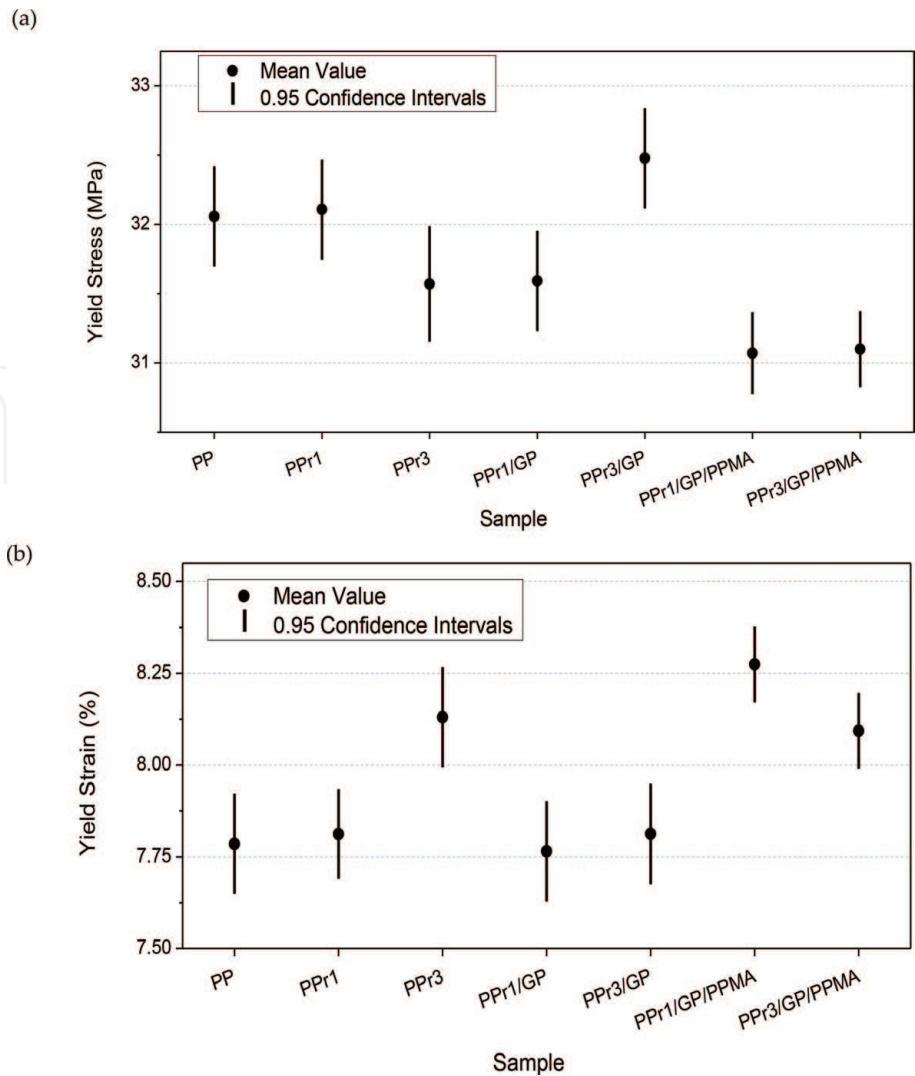


Figure 6.
Yield properties of PP and PP/PPr/GP-base composites: (a) yield stress and (b) yield strain.

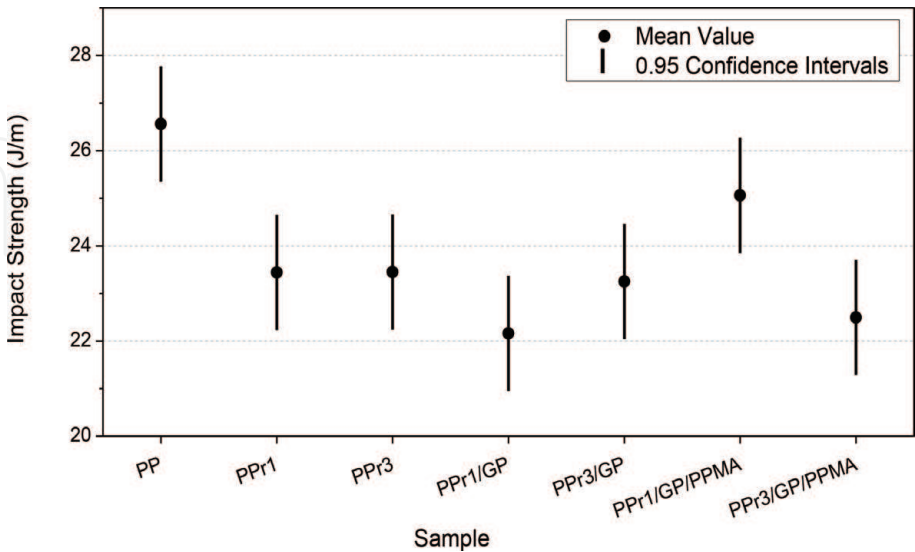


Figure 7.
Impact strength of PP and PP/PPr/GP-base composites.

tmodulus of the composite. One reason that may explain the previous undesirable result is the high amount of PPMA used as plasticizer. In other words, the amount of PPMA exceeded what was needed to coat the particle surfaces, diffusing in the

polymer matrix and influencing plasticizer properties or those of a third polymer component.

Yield stress is the maximum stress at which the material begins to exhibit permanent deformation. As the elastic limit shifts, the material does not return to its original dimensions after the applied stress is removed. This property is particularly important for automotive applications. The yield stress of composites is illustrated on median stress x strain curves (**Figure 5**), showing the PPMA effect.

Yield properties (**Figure 6**) are generally in accordance with the modulus trend, that is, the higher the stiffness, the greater the stress and lower the strain at the yield point of the material. This behavior became evident when virgin PP was compared to PPr/GP composites. For products whose performance is highly dependent on tensile properties, the PPr3/GP sample remains the best option, considering elastic modulus, yield and environmental aspects, since the properties are very similar to those of neat PP even with the addition of 30% PPr and 5%wt of GP.

With respect to the automotive applications of plastic materials, acceptable impact strength is one of the requirements and, in the case of composites, this property is highly sensitive to particle/matrix debonding during mechanical energy dissipation. **Figure 7** shows the mean values and confidence intervals of impact strength.

The PPr1/GP/PPMA sample showed somewhat better results compared to virgin PP, with a P-value of 0.0838, indicating no significant difference between results. The use of PPMA as a coupling agent for GP could be optimized to improve the impact strength of composites.

In the present study, the PPMA grade used exhibited a maleic anhydride level of 1.0% by weight, but other grades with higher levels and greater affinity to GP could be tested in future research.

Compared to virgin resin, blends of PP with PPr demonstrated poor impact properties (**Figure 7**). In addition to higher impurity levels, PPr is expected to show lower molecular weight than PP, and both factors can contribute to failure in recycled material blends. Fukuhara et al. [58] evaluated isotactic polypropylene with different molecular weight and observed less Izod impact strength in samples with lower molecular weights. Furthermore, any structural particularity in PPr able to influence PP crystalline morphology can modify mechanical properties. Xu et al. [59] studied the relationship between spherulite size and crystallinity in the impact strength of PP samples with several different nucleating agents. The authors reported that impact strength was primarily controlled by spherulite size for samples with low crystallinity. For high crystalline samples, crystallinity itself is the decisive factor in strength. The authors also observed that impact strength is greater in PP samples with small spherulites and lower crystallinity. Nevertheless, no clear relationship between the degree of crystallinity and impact strength of samples was observed in the present study (**Table 3, Figure 7**). As such, other factors, such as impurity content, may exert the greatest influence on impact results. Given that products such as furniture and automobile parts require high impact strength, suitable coupling agents should be added to recycled composites in order to enhance their properties.

5. Conclusions

The greatest challenge to plastics in the automotive industry is in recycling. Some automotive manufacturers, such as Ford and Toyota, are recycling their vehicles plastics and reusing in the new vehicles, for example, old or damaged bumpers are recycled and reused in bumper reinforcement cores [60].

According to the results presented, it can be concluded that the properties did not vary significantly as a function of composition. Since the objective was to produce lower cost composites (incorporating recycled PP and glass powder) and more sustainable materials without significant loss of properties, this result is within the parameters established. In other words, it was possible to recover post-consumer materials, replacing the virgin resin without significant loss of mechanical integrity in the final product. The addition of maleated polypropylene (PPMA) was shown to significantly improve the toughness of the material.

In conclusion, based on the properties analyzed and the sustainable appeal of the new products, the powder-based composites displayed potential for use in various applications in the automotive industry, replacing conventional materials.

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Conflict of interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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