



# Incorporation of Recycled Polypropylene and Fly Ash in Polypropylene-Based Composites for Automotive Applications

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## Abstract

The principal aim of this study is to investigate the utilization feasibility of waste materials, i.e., recycled Polypropylene (PP), and fly ash in the formulation of auto parts which are commonly designed based on PP. The typical formulation of auto parts consists of PP, elastomer, filler, and compatibilizer. Various recycled PP/virgin PP ratios were considered as the matrix of the composites. It is found that the increasing concentration of recycled PP in the composite leads to a reduction in mechanical properties, particularly ductility and impact strength. However, due to a lower viscosity at the presence of recycled PP, better dispersion of filler particles, observed by SEM analysis, leading to the improvement of tensile strength. Besides, the employment of recycled material lowers the percentage of crystallinity and melting temperature. It is deduced that the deployment of 20/80 to 40/60 ratios of recycled PP/virgin PP in the composites reasonably meets the requirements for auto parts with advantages in the environmental and economic aspects. Compared to the composites filled with talc, the ductility of fly ash-filled composites is higher. The simultaneous utilization of talc and fly ash as a hybrid system brings about higher ductility, but lowers impact strength compared to talc-filled samples. Using recycled PP mixed with fly ash provides advantages in cost reduction and sustainable and environment-friendly production.

**Keywords** Polymer recycling · Mechanical properties · Polypropylene-based composites · Fly ash · Talc

## Introduction

Polypropylene (PP) is a thermoplastic polymer, which is commonly used in various industrial applications due to its advantageous properties at a reasonable price [1–6]. Due to high and ever-growing consumption, a significant amount of PP is found in the industrial and municipal wastes [7]. From economic and environmental viewpoints, recycling of the post-consumer polypropylene parts is considered attractive [8, 9], which reduces the use of virgin PP and the environmental repercussions. However, the recycling rate of plastic wastes so far is not satisfactory yet. In 2015, for instance, merely 20–25% of global plastic waste was recycled, and the reminders were sent to landfills to get disposed [10, 11].

Among all defined recycling techniques for thermoplastics [12], mechanical recycling has been proposed as the most beneficial and preferred technique due to its low amount of green gas emissions and energy efficiency [13]. However, it should be considered that mechanical recycling causes deterioration of the plastic parts during the reprocessing to have a worse aesthetic appearance and mechanical properties [14]. It is worthwhile to note that the mechanical degradation is the outcome of applied shear forces during the grinding and processing steps, while the thermal degradation is due to the oxygen and high-temperature during the melting and reprocessing [15]. Recently, many studies have been carried out to investigate the effects of mechanical recycling on the properties of PP.

It has been reported that the structure of polymeric chains, morphology and crystallinity, rheological parameters, and mechanical properties of PP are all affected by successive reprocessing. Aurrekoetxea et al. [16] indicated that the main degradation mechanism of PP after several recycling cycles is a chain scission but not oxidation. The same findings were reported in many related studies later [17–19]. In a study performed by Incarnato et al. [20] successive

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reprocessing resulted in shortened polymeric chains with no significant chemical changes implying the lower molten state viscosity of recycled PP [17, 21]. As an outcome of the mechanical recycling process, morphologic alterations of PP are inescapable. The chain scission makes shorter chains, which improves the freedom and foldability of the chains and consequently results in a higher content of crystals [21]. A slight decrease in the crystallinity of recycled PP was reported by Ghahri et al. [19], which can be due to a high amount of chain ends acting as imperfections. Small spherulite and high crystallization temperature of recycled PP are related to impurities that act as nucleation agents [8]. da Costa et al. [14] reported that increasing the number of re-extrusion cycles of PP decreases the melting temperature, but in another study, increased melting temperature of PP was also reported [8]. In association with the mechanical properties of recycled PP, no changes in the elastic modulus and the yield stress indicate that small-strain properties are hardly affected by recycling [15]. However, recycling affects the impact strength and high-strain properties, where strain at break is measured, considerably. The reduction in the ductility and the impact properties of PP are reported several times in the literature [14, 16, 17, 21]. A huge drop in the toughness and the impact strength of recycled PP is predominantly correlated to the chain scission or molecular weight depletion leading to the low density of chain entanglements. Also, high crystallinity, which curbs the flexibility and ability of chains to rotate freely, plays a decisive role in the loss of impact strength and ductility [21].

Needless to say, the deterioration of the properties of recycled materials narrows their reusability. Due to the economic and environmental importance of this limitation, many researchers recently have attempted to provide appropriate solutions for the use of recycled materials in various industrial applications. Additives such as mineral fillers [22–24], impact modifiers [25–28], and coupling agents with fillers [29–32] enhance some mechanical properties of recycled PP, but it is at the cost of the other properties. Besides, the simultaneous use of virgin and recycled PP in the literature [33] and the use of other polyolefins such as PE [9, 34, 35] with recycled PP are introduced as a practical method to reduce the use of virgin material. The focus of recent researches has been mostly on the effect of reprocessing and also different additives on the properties of recycled PP. However, very few publications are available in the literature that discusses the impacts of recycled PP utilization on the properties of virgin PP-based composites containing filler, impact modifier, and coupling agent simultaneously, which are extensively used for various applications, particularly for auto parts. To the best of the authors' knowledge, this is the first study investigating the properties of PP-based composites modified with talc, polyolefin elastomer (POE), and maleic anhydride grafted polypropylene (PP-g-MA) in

the presence of recycled PP in the formulation. In the present study, the effect of recycled PP utilization, at various recycled PP/virgin PP ratios, on the properties of the formulation for auto parts are evaluated. Consequently, the optimized amount of RPP that can be incorporated with a minimal reduction in the properties of the composite is found. Furthermore, by substituting talc by fly ash, a waste material of thermal power plants [32, 36], and applying the hybrid system of talc/fly ash in the compounds, the potentiality of fly ash utilization in auto parts are investigated. The importance of such studies gets highlighted when we take into account that the use of recycled PP and fly ash is an environmental and economic solution especially in the auto industry, which can help to the efficient elimination of waste materials from the biosphere to some extent.

## Experimental

### Material

Polypropylene considered in this study has a commercial heterophasic copolymer grade (Jampilen EP 440L) with MFI 6 g/10 min (ASTM D1238 at 230 °C/2.16 kg), supplied by Jam Petrochemical Co., Iran. The maleic-anhydride-grafted polypropylene (PP-g-MA) with the trade name Aria Couple 1405 with an MFI (ASTM D1238 at 230 °C/2.16 kg) of 10 g/10 min from Aria Polymer Pishgam Co. (Iran) was used as a coupling agent. The POE (Solumer 875L) used in this study was an ethylene-octene copolymer grade and was purchased from SK Global Chemical Co. in Korea with an MFI (ASTM D1238 at 190 °C /2.16 kg) of 5 g/10 min. The talc (Talc A1) with a mean particle size of 5 microns was obtained from Omya Pars Co. in Iran. F-class fly ash with the trade name of Pozzo 100 and the mean size of 20 microns was purchased from Dirk group Co. in India. The recycled PP was purchased from a local supplier with an MFI (ASTM D1238 at 230 °C/2.16 kg) of 8.1 g/10 min. It was stated by the supplier that the employed RPP was mechanically recycled from auto battery cases, which is commonly fabricated from a heterophasic copolymer PP as well.

### Sample Preparation

To prepare compounds, Coperion Werner Pfeleiderer ZSK-25 WLE co-rotating twin-screw extruder with  $L/D=40$  and the screw diameter of 25 mm was utilized. Two loss-in-weight feeders were used to convey raw materials to the extruder. The side feeder was allocated to the filler, and other pellet-form materials according to the recipes were fed into the hopper through the main feeder. The speed of screw was set on 600 rpm and the feeding rate was 30 kg/h. The temperatures of different zones of the barrel are shown in Table 1, from the

hopper to the die. To prepare tensile and Izod impact test specimens, an injection-molding machine manufactured by Poolad Co. in Iran was employed. The temperature was ranged from 180 to 210 °C, from the hopper to the nozzle.

The samples used in this study are listed in Table 2. The nomenclature T30 and FA30, for example, indicate the samples filled with 30% Talc and fly ash respectively. Also, TFA shows the hybrid system of both Talc and fly ash fillers. The letter “R” at the beginning of some samples intimates the presence of recycled material in the formulation. It should be noted that in the same authors’ previous study [37], the optimized state of PP/Talc/POE/PP-g-Ma composites was acquired with 30% Talc, 6.2% POE, and 2.5% PP-g-MA, in accordance to the requirements for auto interior parts.

## Characterization

To determine the tensile properties according to the ASTM D638 test method, a Zwick/Roell Machine (model z100) at a crosshead speed of 50 mm/min was used. The impact properties of the samples were measured according to ASTM D256A by a Zwick/Roell B5102 pendulum impact tester. The melt flow rate measurement was conducted according to ASTM D1238 by Ceast Junior Melt Flow Index tester at 230 °C. It should be noted that the reported values for tensile properties, impact strength, and MFI analysis are the arithmetic mean of five replications for each sample. To check the exactness of the filler content in the compounds, a laboratory scale furnace was used. According to the ATSM D5630 test method, samples were kept approximately at 800 °C for 45 min.

Scanning electron microscopy (SEM) images taken from the fractured surface of the Izod impact test specimens were used to assess the microstructure and morphology of the samples. The SEM images were obtained using VEGA-II TESCAN apparatus. To avoid electrical charges accumulated during the test, the surfaces of the specimens were sputtered with a thin layer of gold. The differential scanning calorimetry (DSC) analysis was used to evaluate the thermal characteristic of the samples. The DSC analysis was performed with an apparatus manufactured by Sanaf Co. in Iran. First, the samples were heated from ambient temperature to 210 °C with a rate of 10 °C/min. To erase previous thermal history, the samples were kept at 210 °C for 5 min, and then the crystallization and cooling took place with the same rate and thermal scope. Data were collected from the second heating process performed in the same way. The data obtained from the cooling and heating curves were used to extract the melting temperature, the crystallization temperature and the degree of

**Table 2** The code of samples

Sample	Recycled PP (%) / virgin PP (%)	POE%	PP-g-MA %	Talc%	Fly ash%
T30	0/100	6.2	2.5	30	0
R20T30	20/80	6.2	2.5	30	0
R40T30	40/60	6.2	2.5	30	0
R60T30	60/40	6.2	2.5	30	0
R80T30	80/20	6.2	2.5	30	0
R100T30	100/0	6.2	2.5	30	0
FA30	0/100	6.2	2.5	0	30
R40FA30	40/60	6.2	2.5	0	30
R100FA30	100/0	6.2	2.5	0	30
TFA	0/100	6.2	2.5	15	15
R40TFA	40/60	6.2	2.5	15	15
R100TFA	100/0	6.2	2.5	15	15

crystallinity of the samples. It should be noted that the degree of crystallinity was estimated using Eq. (1).

$$X_{c,PP} = \frac{\Delta H_{m,PP}}{(1 - \omega)\Delta H_{100\%,PP}} \quad (1)$$

where  $\omega$  shows the weight fraction of mineral filler in the compound,  $\Delta H_{m,PP}$  is the area under the melting peak of PP in DSC thermogram and  $\Delta H_{100\%,PP}$  is equal to 209 J/g that is the standard enthalpy of PP when it is 100% crystalline [22], while this value for Polyethylene (PE) is reported close to 290 J/g [38].

To obtain the infrared spectra of the samples, a Perkin–Elmer spectrophotometer equipped with an attenuated total reflectance (ATR) was used. Each spectrum was recorded from 4400 to 400  $\text{cm}^{-1}$  at the 4  $\text{cm}^{-1}$  resolutions.

## Result and Discussion

### FTIR

The FTIR spectra of the recycled and virgin PP are presented in Fig. 1. As a result of oxidation reactions during the processing or reprocessing of PP, the presence of the carbonyl group, which is composed when the material is exposed to excessive thermal and mechanical strains with the presence of oxygen [15], can be tracked somewhere between 1600 and 1900  $\text{cm}^{-1}$  in the FTIR spectrum [19]. In Fig. 1, except for the peak around 1700  $\text{cm}^{-1}$ , related conceivably to the carbonyl

**Table 1** Temperature profile from hopper to die head

Zone no.	1	2	3	4	5	6	7	8	9	10
Temp (°C)	195	205	210	215	205	200	195	195	205	210

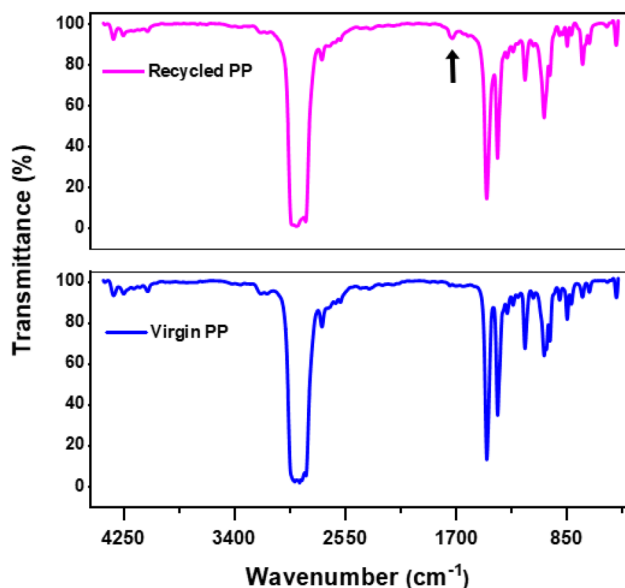


Fig. 1 FTIR spectrum of virgin PP vs. recycled PP

group in recycled PP, no significant difference in spectral bands can be observed.

### Scanning Electron Microscopy

The SEM micrographs of some composites are presented in two different magnifications in Fig. 2. These images belong to the T30 (Fig. 2a and d), R60T30 (Fig. 2b and e) and R100T30 (Fig. 2c and f) samples. In all the images, platelet like particulates of Talc are oriented towards the injection flow and perpendicular to the fractured surface. Despite the high concentration of the reinforcement, agglomerations are rarely found, which indicates that the rigid phase in the polymeric matrix is well distributed. The rough fractured surface of T30 sample indicates the plastic deformation and the semi-ductile behavior of the composite [39]. Addition of recycled PP leads to smoother fractured surfaces implying the ductile-brittle transition in the fracture mechanism of samples. This transition is due to the lower molecular weight of the recycled material [21]. However, due to the lower viscosity of the recycled material, the dispersion of the filler throughout the matrix in the R100T30 sample is more desirable than the T30 (Fig. 2c). Better dispersion of the filler removes agglomerations and make a greater filler-matrix

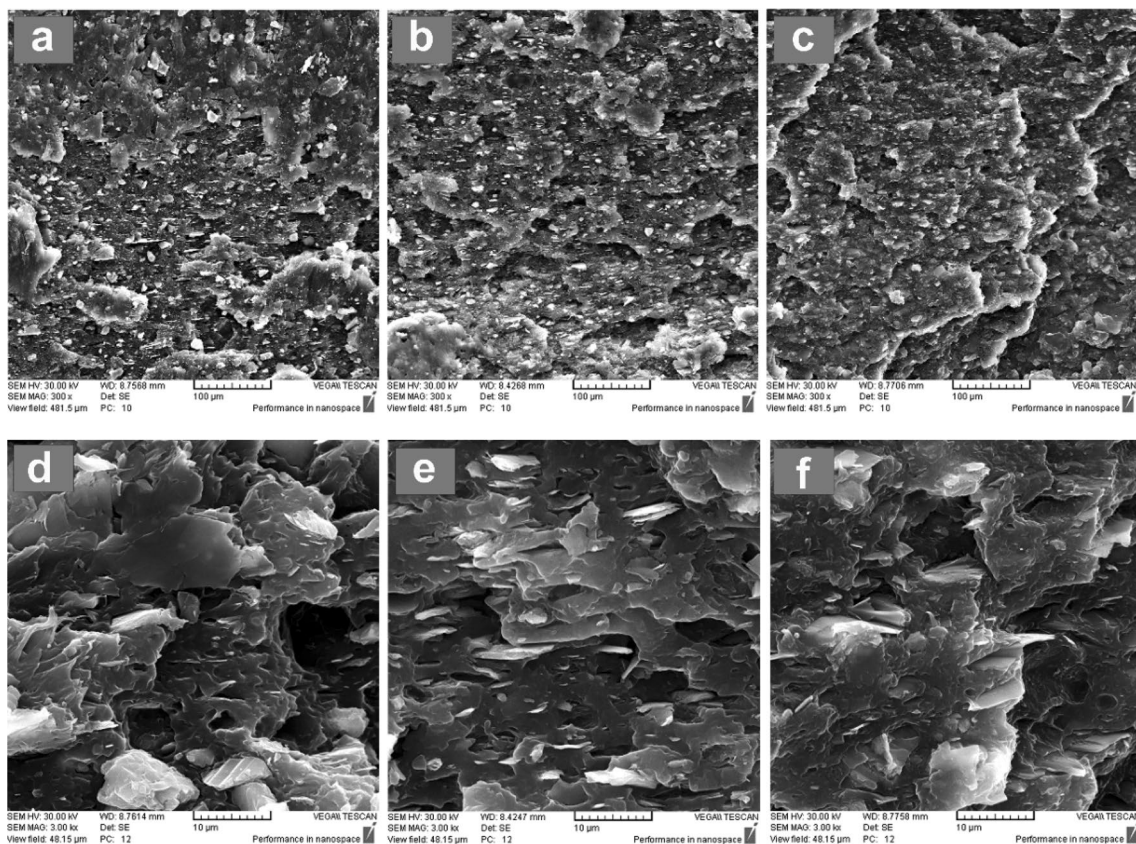


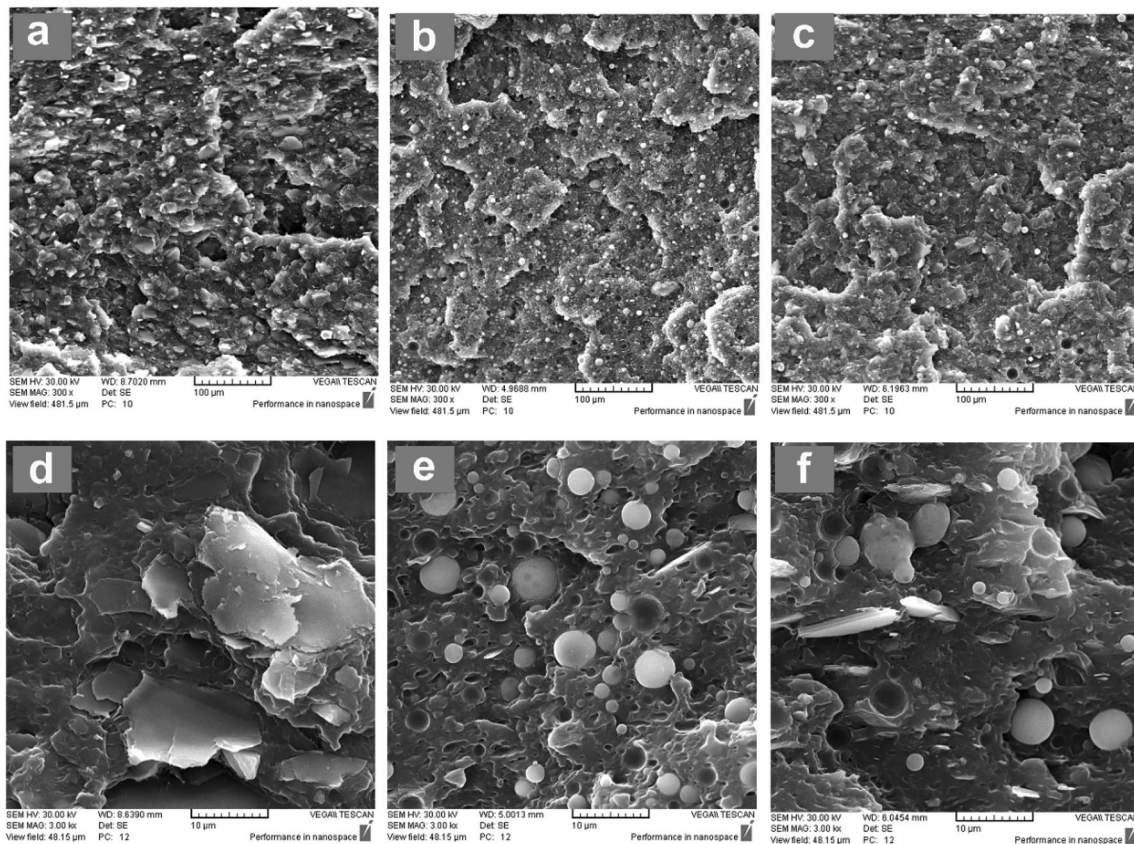
Fig. 2 SEM images of samples with various recycled PP/virgin PP ratios: T30 (a, d), R60T30 (b, e), and R100T30 (c, f)

interfacial surface area, and therefore, the applied stress can be transferred more efficiently from the matrix to the reinforcement [40].

The SEM micrographs of the R40T30, R40FA30, and R40TFA samples are compared in Fig. 3. The rough fractured surface of the R40FA30 sample (Fig. 3b) indicates the plastic deformation and the ductile behavior of the composite, which causes higher deformability compared to the R40T30 sample. The matrix residue on the surface of particles implies a good filler-matrix interfacial interaction [41].

Based on Fig. 3d and e, contrary to R40T30 sample, the lack of matrix residuals on the surface of fly ash particles and the presence of vacant spaces around spheres indicate the not well-formed bonds between the polymeric matrix and fly ash particulates. The incompatibility of polymeric segments and fly ash particulates is chiefly due to the unidentical chemical characteristics of phases [42]. Thus, silane functionalization has been introduced as an efficient and practical chemical surface treatment method bringing about a significant improvement in the mechanical properties of fly ash-filled composites [36, 43–45]. In this context, silane groups can act as chemical

bridges among hydrophobic polymer and hydrophilic fly ash particles leading to the enhancement of the interfacial adhesion [42]. Compared to talc-filled samples, the unseemly interfacial interactions between PP segments and fly ash particulates is conceivably due to the lower density of hydroxyl groups on the surface of fly ash particulates [46]. Insufficient hydroxyl groups on the surface of the fly ash particles reduces the effectiveness of the employed compatibilizer agent noticeably. Also, the higher aspect ratio of talc particles, which is due to its platelet geometrical shape, can be a determining factor in this regard. Besides, the number of surface microvoids of fly ash-filled composite (Fig. 3b), which are commonly found due to pulling out of the filler particles [39] or cavitation of dispersed POE droplets [47] verify this weak connection. At the simultaneous presence of Talc and fly ash particles, the context can be much more complicated (Figs. 3c and f). Due to the interference of the fly ash particles, the interfacial connections between the Talc and polymeric segments may not be formed perfectly resulting in adverse impacts on the mechanical properties of the hybrid composites. Besides, inhomogeneity in the hybrid systems, such as different shapes of fillers and bond strengths, can be another source of defects.



**Fig. 3** SEM images of samples at the presence of various fillers: R40T30 (a, d), R40FA30 (b, e), and R40TFA (c, f)

### DSC Analysis

In this section, the thermal behavior of virgin and recycled PP-based samples is evaluated using DSC analysis. The DSC thermograms are provided in Fig. 4. The appearance of two distinguishable peaks in the melting curve of recycled PP shows the presence of PE together with PP, which could be

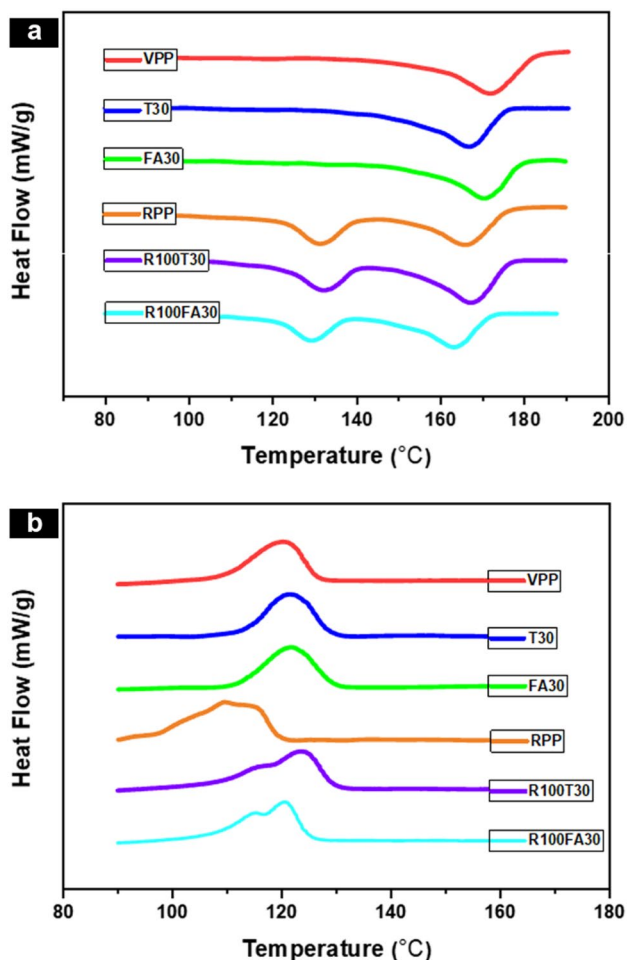


Fig. 4 Melting (a) and crystallization curves (b) of virgin and recycled PP-based samples

blended mechanically during the recycling process and was immiscible with PP. The peak near 130 °C is related to the melting of PE, and that around 160 °C is related to that of PP. There is a correlation between the content of the PE in the PP/PE blend and the area under its melting peak [13].

The results of the DSC analysis are presented in Table 3. As in the table, the melting ( $T_m$ ) and crystallization ( $T_c$ ) temperature and the crystallinity ( $X_c$ ) of recycled PP are lower than those of virgin PP. The lower melting temperature of the recycled material can be the outcome of the lower molecular weight of the polymeric chains forming weaker crystals [22]. Due to the immiscibility of PE and PP chains and the interference of PE chains in the crystallization of PP segments, the crystalline proportion of PP and the total crystallinity are smaller in the recycled material compared to the virgin PP. Besides, a fraction of available nucleation sites is held by the PE chains, which can be another reason for a smaller crystal content of PP in the recycled material. The impurities in the recycled material are inescapable, and they can act as the nucleation agents increasing the crystallization temperature [8, 20]. However, as shown in Table 3, the crystallization temperature of the recycled PP is lower due to the presence of PE in the system.

The higher crystallization temperature of T30 and FA30 samples compared to VPP is due to the nucleating effect of the fillers. By using fly ash instead of Talc, the percentage of crystallinity increases slightly. With 40% recycled PP in the system, the context becomes complicated due to the interference of PE. The increased crystallization temperature and total crystallinity of the composites when 40% of recycled PP is used can be due to the low molecular weight and low viscosity of the system, which facilitates the crystallization [20]. Incorporation of 40% recycled PP caused an approximately 3% rise in the total crystallinity compared to the virgin PP-based samples. The low molecular weight results in a low melting temperature of PP crystals in the recycled PP-included samples. Remarkable drops in the crystallinity of R100T30 and R100FA30 samples compared to the virgin PP-based samples (T30 and FA30) are also noticeable. The

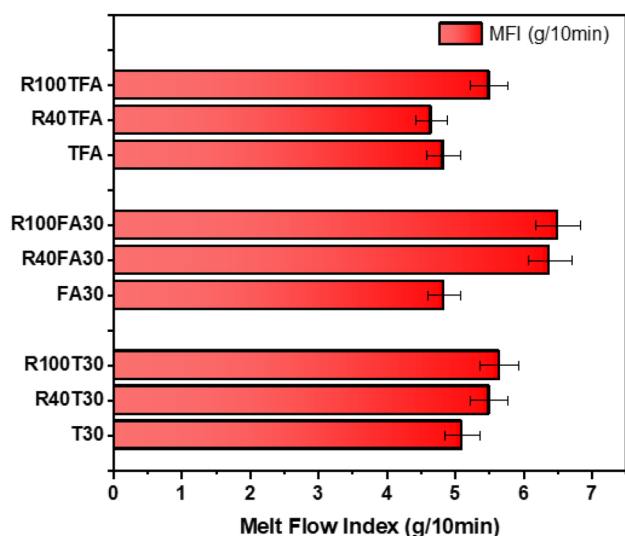
Table 3 DSC analysis of samples with different fillers and Recycled PP concentration

Sample	$T_{m,PE}$ (°C)	$T_{m,PP}$ (°C)	$T_{c,PE}$ (°C)	$T_{c,PP}$ (°C)	$\Delta H_{m,PE}$ (J/g)	$\Delta H_{m,PP}$ (J/g)	$X_{c,PE}$ (%)	$X_{c,PP}$ (%)	$X_{c,Total}$ (%)
VPP	–	167.2	–	124.5	–	64.76	–	30.98	30.98
T30	–	165	–	126.9	–	41.34	–	28.31	28.31
FA30	–	166.2	–	126.4	–	41.93	–	28.72	28.72
RPP	127	162.1	110.3	116	19.86	38.2	7	18.27	25.27
R40T30	123.2	164.7	123.6	128.1	3.775	43.435	1.86	29.75	31.61
R40FA30	124.4	165.1	123.8	126.8	4.75	42.48	2.34	29.1	31.44
R100T30	127.6	164.1	120.5	127.9	12.65	26.58	6.23	18.21	24.44
R100FA30	127.2	161	119.8	125	15.026	28.7	7.42	19.66	27.08

total crystallinity of the R100FA30 and R100T30 samples are 27.08 and 24.44%, respectively.

## Rheological Assessment

The impact of different fillers on MFI is illustrated in Fig. 5. MFI is an index of the viscosity and the molecular weight of the system. As seen in the figure, aside from the types of fillers, the increasing amount of recycled PP raises MFI. This is an indication of the lower molecular weight of recycled PP caused by the chain scission. Due to the better interfacial interaction of Talc with PP chains compared to fly ash, MFI of the system is lower when Talc with its platelet-like shape, is employed. Also, it is reported that fly ash is prone to improve the flow ability of base polymer [43]. In the hybrid system of fillers, due to the complexity in the interactions and uncharacterized interferences, it is difficult to observe a trend for MFI against the percentage of the recycled PP.

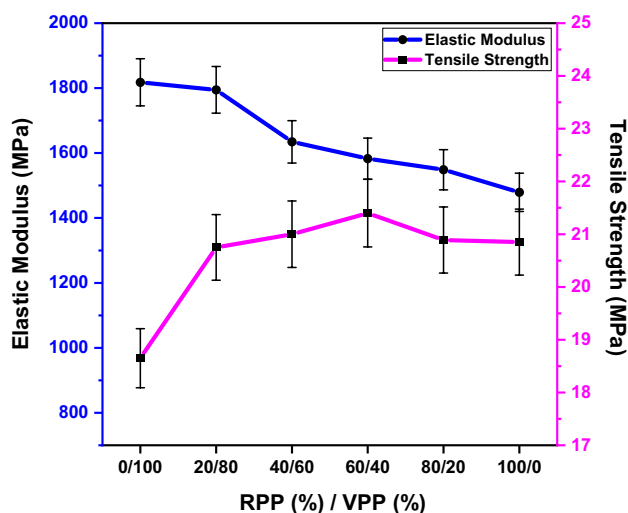


**Fig. 5** The impacts of different fillers and recycled PP concentration on MFI

## Mechanical Properties characterization

The effects of recycled PP on the mechanical properties of PP/Talc/POE/ PP-g-MA composites are evaluated here. The comparison between the mechanical properties of neat PP (virgin and recycled) and composites with different Recycled PP/Virgin PP ratio are presented in Table 4. There is a slight difference between the elastic moduli ( $E$ ) and tensile strengths ( $\delta_y$ ) of neat recycled and virgin PP. However, the Izod impact strength and the elongation at break ( $\epsilon_b$ ) of recycled PP are noticeably small.

The elastic moduli and tensile strengths of the Talc-filled samples are plotted in Fig. 6 against the recycled PP/virgin PP ratio. This figure determines that an increasing percentage of recycled PP lowers the elastic modulus, especially when the content of the recycled PP in the matrix is higher than 20%. The employment of 100% recycled PP resulted in a decrease of 18.6% in the elastic modulus. The low crystallinity and molecular weight of the recycled material can account for the less elastic modulus of the R100T30 sample.



**Fig. 6** The trends of elastic modulus and tensile strength alterations against recycled PP/virgin PP ratios

**Table 4** The impacts of diverse Recycled PP concentration on mechanical properties of PP/Talc/POE/ PP-g-MA composites

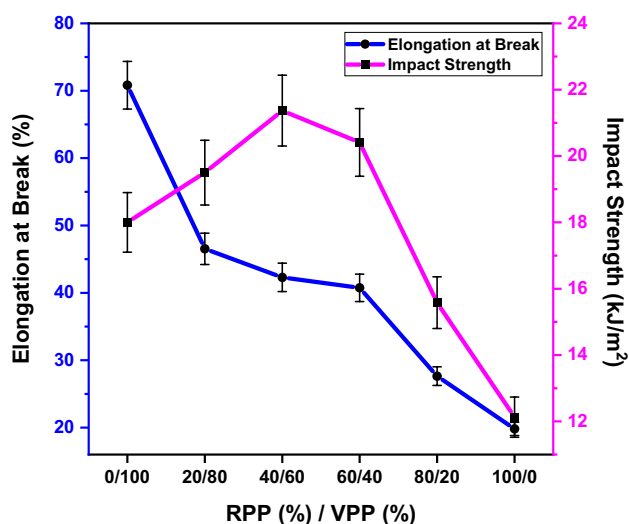
Sample	RPP (%) / VPP (%)	E (MPa)	$\delta_y$ (MPa)	$\epsilon_b$ (%)	Impact Izod (kJ/m <sup>2</sup> )
VPP	–	1115.88 ± 44.6	21.41 ± 0.9	275 ± 14	23.38 ± 1.17
RPP	–	1107.07 ± 42.2	19.47 ± 0.82	22.58 ± 2.3	15.97 ± 0.8
T30	0/100	1817.88 ± 72.7	18.65 ± 0.75	70.82 ± 3.5	18 ± 0.95
R20T30	20/80	1794.72 ± 65.3	20.75 ± 0.88	46.54 ± 2.9	19.5 ± 1
R40T30	40/60	1634.48 ± 62	21 ± 0.9	42.3 ± 2.1	21.37 ± 1.1
R60T30	60/40	1582.66 ± 54	21.4 ± 1.1	40.75 ± 1.8	20.41 ± 1.18
R80T30	80/20	1548.62 ± 61.5	20.89 ± 0.95	27.64 ± 1.4	15.58 ± 0.7
R100T30	100/0	1479.01 ± 59.1	20.85 ± 1.03	19.81 ± 1	12.13 ± 0.52

Also, PE, which is immiscible with PP, can cause smaller elastic modulus [13].

Because of the lower viscosity of the employed recycled PP compared to virgin PP, the better distribution of filler is expected. The uniform dispersion of filler and the lack of agglomerations have a great impact on the improvement of the tensile strength [48]. Although some variations, the high content of recycled PP increased the tensile strength of the composite.

The elongation at break and the impact strength figures of samples against the content of recycled PP are plotted in Fig. 7. It is seen that recycled PP reduces the ductility of the composite. By fully replacing the virgin PP by the recycled PP, the elongation at break decreased by 72%, from 70.82 to 19.81%. This is due to short chains and low molecular weight of the recycled PP caused by excessive thermal and mechanical stresses during the mechanical recycling process [15], leading to a low density of entanglements. Besides, such a reduction in the ductility at higher content of recycled material might be because of the adverse effect of the immiscibility between PE and PP chains, even if considering that POE can improve the miscibility of PP and PE [13].

Due to the lower viscosity and crystallinity, the use of recycled PP can facilitate cavitation and shear yielding, which are the leading modes of energy dissipation in semi-crystalline materials [49, 50]. Also, the low viscosity of recycled PP makes a better distribution of the filler and conceivably reduces the number of agglomerations, which are the stress concentration points [51]. However, the high molecular weight of macromolecules is considered to improve the impact strength of the composites [21]. By using more recycled material, the fraction of chains with lower molecular weight goes up in the system resulting in a loss



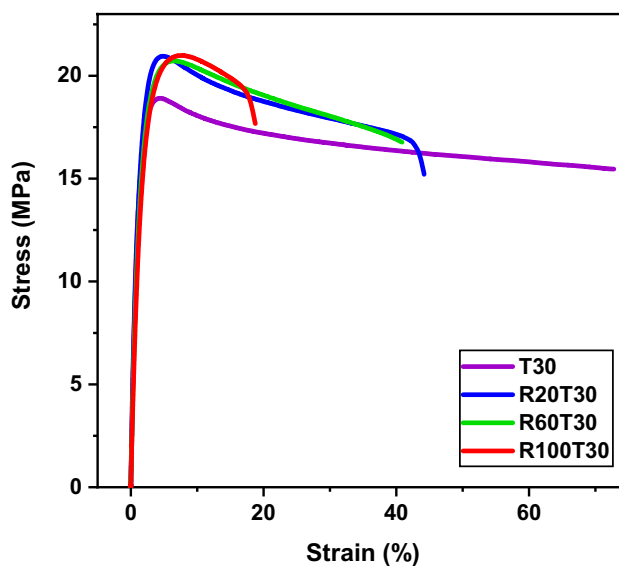
**Fig. 7** The trends of elongation at break and impact strength alterations against recycled PP/virgin PP ratios

in the impact strength. Based on DSC data in Table 3, due to a rise of 3% in the total crystallinity compared to the T30 sample, lower impact strength was expected for R40T30. However, it is seen in Fig. 7 that the impact strength has a peak of 21.37 kJ/m<sup>2</sup> at 40% of recycled PP in the matrix, and afterward, a decreasing trend is observed.

The stress-strain curves of the composites with respect to the percentage of recycled PP are presented in Fig. 8. It is noted that by adding more recycled PP, the ductility is significantly reduced.

Figure 9 shows the impact of different percentages of recycled PP on the stiffness and the tensile strength of the composites with various fillers (Talc, fly ash, and Talc/Fly ash hybrid). The use of fly ash is examined in this study as the fly ash is expected to bring further economic and environmental advantages in addition to the use of the recycled PP. Aside from the types of fillers, due to the lower molecular weight and the crystallinity, increasing the content of recycled PP decreases the elastic modulus of the composites. The elastic modulus of R40T30 and R40FA30 are 1634 MPa and 1169 MPa, respectively. The high stiffness of Talc-filled samples is due to the injection flow-oriented platy particles and a more efficient stress transfer from the matrix to the reinforcements due to well-formed interfacial bonds.

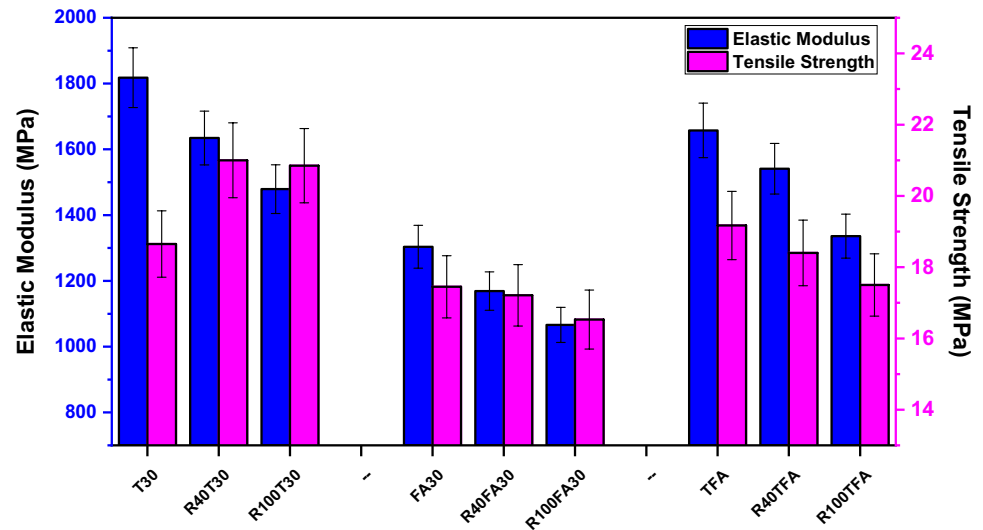
The increased amount of recycled PP proportionally increases the tensile strength of the Talc-filled composites. However, due to the weak interactions between the polymeric chains and the fly ash particles, the improved fluidity cannot enhance the tensile strength in fly ash-filled systems. The use of 100% recycled PP resulted in a drop of 6% in the tensile strength of the fly ash-filled composites. In the



**Fig. 8** Stress-strain curves of samples at the presence of various recycled PP content



**Fig. 9** The impacts of different fillers and recycled PP concentration on stiffness and tensile strength



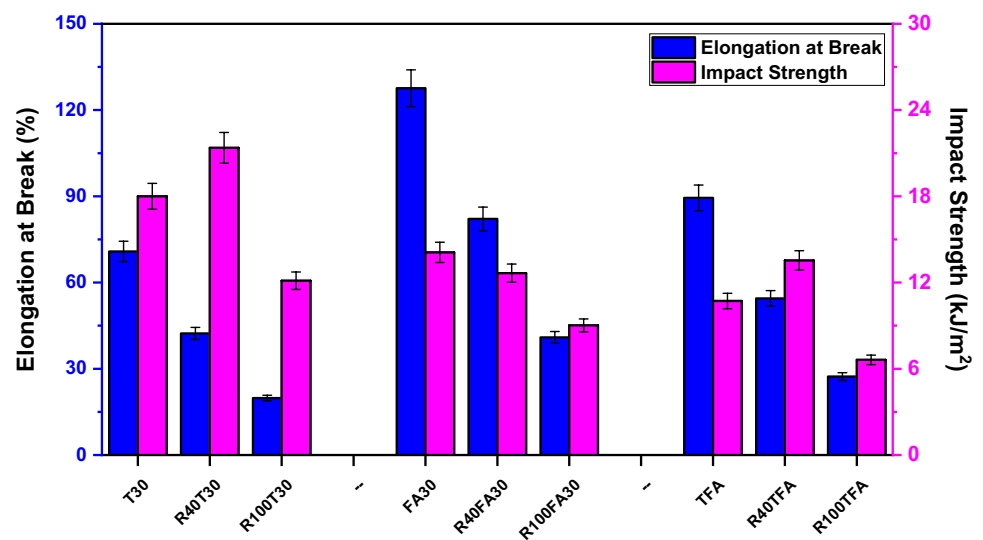
hybrid system, the elastic modulus and tensile strength both decreases with the increasing proportion of recycled PP. The elastic moduli of TFA, R40TFA, and R100TFA samples are 1657.5 MPa, 1540.94 MPa, and 1336 MPa, respectively. As can be seen, the stiffness of the hybrid system is higher than fly ash-filled and lower than Talc-filled samples. The same trend can be detected for the tensile strength, as well.

The influence of the fillers and the recycled PP contents on the impact strength and the elongation at break is presented in Fig. 10. The flexibility of Talc-filled and fly ash-filled composites dropped by approximately 70% after using 100% recycled material because of a lower density of the entanglements of polymeric chains. In comparison to the Talc-filled systems, the fly ash causes much higher ductility. The elongations at break for R40T30 and R40FA30 are 42.3% and 80.2%, respectively, close to twofold. This is presumably because the interfacial interactions of fly ash and

PP are not very strong (Fig. 3e). In the simultaneous use of the two fillers, the ductility is higher than those of the Talc-filled samples and lower than those of the fly ash-filled samples.

As in Fig. 10, the impact strength of composites reinforced with Talc is higher than those with fly ash. Such an observation is seemingly due to the good interfacial interaction between Talc particles and polymeric chains. It can be seen that if there is good interaction between the filler and the matrix (e.g., T30 sample), recycled PP can improve impact resistance to some extent. Utilization of 100% recycled PP instead of virgin PP resulted in a 35% drop in the impact strength of the Talc and fly ash-filled samples. The impact strengths of TFA, R40TFA, and R100TFA are 10.72 kJ/m<sup>2</sup>, 13.54 kJ/m<sup>2</sup>, and 6.62 kJ/m<sup>2</sup>, respectively. In the hybrid composites, the sophistication of the interactions causes more restrictions to the freedom of polymeric chains,

**Fig. 10** The impacts of different fillers and recycled PP concentration on ductility and fracture toughness



which can be verified from the MFI values in Fig. 5. Consequently, polymeric chains lose their capability to neutralize the external forces resulting in low impact strength. It seems that the more restricted chains and an unequal strength of the interfacial connections between polymeric chains and two fillers make the impact strength of hybrid composites lower than the figures of Talc and fly ash-filled samples.

## Conclusion

To sum up, due to the thermal and mechanical degradations of polymeric chains in the recycling process, the mechanical properties of recycled PP is not satisfactory, and the recycled PP cannot easily be used for various applications. The reduction in the performance of recycled PP is clearly shown in the ductility and the impact strength caused by the low molecular weight and the density of entanglements. The inevitable presence of PE with PP can be another reason for defects resulting in an underperformance of recycled material when it is under loading. The simultaneous use of recycled PP and virgin PP can be a proper solution to achieve a good performance/cost balance. However, a high amount of recycled material in the compounds consisting of PP/Talc/POE/PP-g-MA significantly reduces the elastic modulus, the elongation at break and the impact strength while improves the tensile strength. It is important to note that the impact strength reaches a peak at 40% recycled material. The impact strength reduction after this peak is related to the morphological and rheological alterations and the role of impurities and PE chains. With 20–40% recycled PP in the composites, the economic and environmental advantages can be achieved while meeting the requirements for auto parts. By substituting Talc by fly ash, an improvement in the elongation at break figures is observed while the other mechanical properties are decreased. This trend is because of the morphological mutations and the inappropriate interfacial interaction between fly ash particulates and polymeric chains. This is also observed in SEM images. By using Talc and fly ash together as a hybrid system, unclearly characterized trends are observed due to the complexity of the system. Consequently, unless the economic and environmental advantages, the utilization of fly ash along with Talc in the formulation of auto polymeric parts requires more examinations in the term of the mechanical properties.

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