

INVESTIGATING THE EFFECTS OF WASTE GROUND RUBBER TIRE POWDER AND PP-g-MA COMPATIBILIZER ON THE MECHANICAL PROPERTIES OF PP/WASTE GROUND RUBBER TIRE (WGRT) POWDER/PP-g-MA COMPOSITES

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In this research, the effect of waste ground rubber tire (WGRT) powder was experimentally investigated on mechanical properties of polypropylene (PP). PP-g-MA compatibilizer was used to increase the compatibility of waste rubber tire powder with the PP matrix. All the samples were mixed in a co-rotating twin screw extruder and were formed into standard tensile and impact bars using the injection molding method. The morphology of combinations was studied by field emission scanning electron microscopy (FESEM). The FESEM micrographs taken from fracture surface of the parts indicated that PP-g-MA led to compatibility increase of tire powder with the PP matrix and better dispersion and prevented from agglomeration of tire powder in that. Adding tire powder to PP matrix in all binary and ternary combinations increased impact strength of PP. In the blends containing 5 wt% PP-g-MA, significant changes in tensile properties of the compositions occurred that may be caused by the created appropriate bond strength in this weight percent of PP-g-MA between tire powder particles and PP matrix. Young's modulus, yield stress and tensile strength of ternary combinations increased by the decrease of the weight percent of tire powder and increase of PP-g-MA, which were attributed to the increased bond strength. Also, break elongation decreased with the decrease of the weight percent of tire powder and increase of the amount of PP-g-MA, due to the reduced soft rubber phase and increased bond strength.

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1. Introduction

Low impact strength, especially at low temperatures, is one of the PP weaknesses that is a limitation for its use in many applications [1-3]. Using materials with high impact strength in PP-based compounds is considered one of the strategies for dealing with this weakness. In this regard, the use of rubbers such as waste rubber tire powder in the PP-based compounds is useful because of its low price and good environmental effects [4]. Waste ground rubber tire (WGRT) powder is a thermoset material, which include polymer chains with irreversible cross-links. These links avoids the materials from being reprocessed and reused. Adding the WGRT in powder form or vulcanized scrap into polymer compositions has lots of economic benefits [5, 6].

The technical and commercial feasibility of using WGRT powder as filler has been demonstrated in many applications like roofing and shoe soles. The properties of polymer

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composites are determined by four factors of component properties, composition, structure and interfacial interaction. Interfacial adhesion is extremely important in all the composites used in load-bearing applications. Improper adhesion leads to debonding of the components, development of voids, which may merge to large cracks, and premature failure of the part. Interfacial adhesion is the decisive factor which determines the properties of composites and appropriate adjustment of interphase properties; adhesion strength is the most important condition of achieving acceptable properties. Very strong adhesion is claimed to lead to stiff and brittle composites while, in the case of weak adhesion, the components are debonded under the effect of external load. Therefore, medium or appropriate strength is claimed to have the highest advantages which can be induced by fundamental reaction in the melt-mixing process [7, 8]. The interfacial adhesion between the WGRT and the polymer matrix is usually very weak due to the cross-linked structure of WGRT. In order to further improve the interfacial adhesion between the WGRT and the PP, not only the WGRT should be devulcanized, or at least partially devulcanized, to facilitate the molecular entanglement between the WGRT and the PP matrix, but also the compatibilizer must be added to the blend. Functionalized polymers are often added to polyolefin composites to create the necessary adhesion [9, 10].

As far as the compositions of PP and WGRT are concerned, the maleic anhydride grafted functionalized polymers are more used as compatibilizer [11-13]. The surface of the WGRT reacts with a functionalized polymer that has a compatibilizer role such that the cross links in the WGRT are broken under mechanical mixing in extruder and the MA polar group in functionalized polymer reacts with the phenolic OH group in WGRT [11-14]. This chemical interaction leads to improved compatibility and adhesion between PP and WGRT as well as better dispersion of tire powder in polypropylene matrix [1, 15 and 16].

In this research, the effect of WGRT powder was experimentally investigated on mechanical properties of PP. PP-g-MA compatibilizer was used to increase the compatibility of WGRT powder with the PP matrix. All binary and ternary composites and even pure PP and pure WGRT powder through melt mixing method were mixed in a co-rotating twin screw extruder and then were formed into standard tensile and impact bars using the injection molding machine. Impact strength and tensile tests were performed to see the effect of embedding the desired particles in the PP matrix. The results showed that, if the proper amount of these particles was used, the impact and tensile strength of the compounds might be considerably improved.

2. Experiment

2.1. Materials

In this study, PP homopolymer moplen HP550J was supplied by Arak Petrochemical Company, Iran. Its melt flow index (MFI) measured by an ASTM-D1238 was 3g/10 min (230 °C, 2.16 kg). The compatibilizer, PP grafted maleic anhydride (PP-g-MA) (MA content = 2.4 wt%, MFI = 38 g/10 min with an ISO1133), was purchased from Kimia Baspar Company, Iran. According to the manufacturer, the brand of PP in the PP-g-MA was V30S from Maroun Petrochemical Company. WGRT was produced by Taminavaran Lastik Ghadir Company in Iran. The WGRT was produced by a wet grinding method from a combination of 70% truck tires and 30% passenger tires. The material was obtained from the whole part of tires, with separated metal and polyester cords. Its particle size was characterized to be 50 meshes ($\approx 300 \mu\text{m}$) by FESEM (Fig. 1). The approximate composition of the WGRT was as follows: 29% natural rubber (NR), 26% styrene-co-butadiene rubber (SBR), 30% carbon black, 9% oils and low molecular weight additives and 6% ash. The exact composition depends on the specific type of tire and the place of the tire from which the particles are originated.

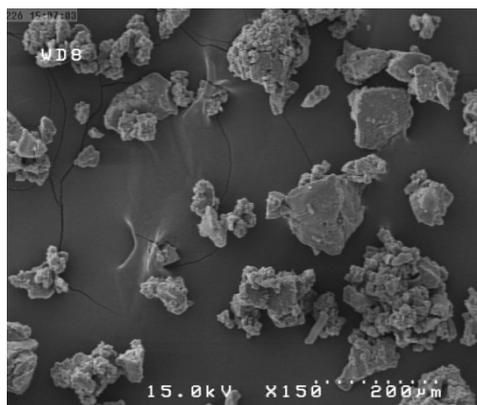


Fig. 1. FESEM micrograph of 50 mesh WGRT powder.

2.2. Sample preparation

Each of the pure PP, PP-g-MA, WGRT samples and also all binary and ternary compounds were built in the same conditions after physically mixing their components. Composite ingredients were mixed together in each case based on the weight percentage. Compounding of the materials was done using a co-rotating twin screw extruder (ZSK 25 P8.2E WLE). This extruder had six temperature control zones. Unblended pure PP, WGRT and other blends were prepared in a temperature profile of 160/165/170/175/180/190 °C. The screw speed was fixed at 250 rpm. The L/D ratio of the screws was 40 and D = 25 mm. The product was cooled in a water bath and was later granulated into a form ready for injection molding into impact and tensile test specimens. Injection molding was carried out in a temperature profile of 190/200/210 °C with an injection pressure of 90 bar. After conditioning for 24 h, mechanical properties were evaluated. Stress-strain data were determined by the Zwick/Roell machine, Z100 model, on I-type specimens according to ASTM D-638. A crosshead speed of 5 mm/min was used in the tensile tests. Izod impact strength testing of the notched specimens according to ASTM D-256 was conducted using an RESIL IMPACTOR impact tester at 1 J energy level. The average value of the impact and tensile properties were calculated using at least five samples. Their compositions are listed in Table 1. Letter “g” represents the weight percent of PP-g-MA and letter “W” represents the weight percent of tire powder in combination. No letter number indicates the weight percent of PP.

2.3. Scanning electron microscopy and phase structure analysis

Studying of composites microstructure, dispersion manner of tire particles in the PP matrix were done by a Hitachi (S-4160) field emission scanning electron microscopy (FESEM) at 15 kV working voltage. The specimens were obtained by cryogenic fracture. After immersing the samples in liquid nitrogen for more than 20 min, they were broken by hand immediately (less than 3 s) after they had been taken out of liquid nitrogen. To prevent electrical charging, the SEM samples were sputter-coated with a thin layer of gold. The coating of the fracture surfaces was done by TECHNICS machine, for 12 minutes at 6 kV working Voltage and 5 mA flow under argon gas atmosphere. To improve the contrast between PP matrix and WGRT phase, the fractured surfaces of all composites containing PP and WGRT were etched in 12 M hydrochloric acid for one week in advance.

Table 1. Compositions of the composites studied in this work^a.

Abbreviation	Component's Amounts (Weight percent)		
	PP	WRT	PP-g-MA
PP	100.0	0.0	0.0
WRT	0.0	100.0	0.0
PP-g-MA	0.0	0.0	100.0
95/5W	95.0	5.0	0.0
90/10W	90.0	10.0	0.0
80/20W	80.0	20.0	0.0
70/30W	70.0	30.0	0.0
40/60W	40.0	60.0	0.0
95/5g	95.0	0.0	5.0
90/10g	90.0	0.0	10.0
65/34W/1g	65.0	34.0	1.0
65/32.5W/2.5g	65.0	32.5	2.5
65/30W/5g	65.0	30.0	5.0
40/55W/5g	40.0	55.0	5.0
65/27.5W/7.5g	65.0	27.5	7.5
65/25W/10g	65.0	25.0	10.0
40/50W/10g	40.0	50.0	10.0
65/32.5W/2.5g	65.0	32.5	2.5

^aNote: “g”, “W” and no letter number denote PP-g-MA, WGR powder and PP in combination, respectively.

3. Results and discussion

3.1. Characteristics of the composites phase structure

Lots of parameters affect mechanical properties of nano-composite materials which include the amount of filler, form of filler, particle dispersion in the matrix and type and amount of particle coating, etc. [1, 8, 17-21]. Tire powder particles should be small. They also must have good dispersion and good adhesion with the matrix [1, 15 and 20]. In this study, the applied compounds were PP, WGR powder and PP grafts with maleic anhydride (PP-g-MA). Since tire powder was considered the filler, according to the expressed content, good dispersion and adhesion with the PP matrix were essential for obtaining good mechanical properties. Thus, PP-g-MA was used in small amounts to enhance the compatibility of tire powder with a PP matrix.

Studying the phase structure of composites on their mechanical performance analysis will be useful. So, FESEM images taken from the fracture surfaces of the samples after breaking them in liquid nitrogen were checked. Fig. 2 shows images of the broken surfaces of without filler modes with liquid nitrogen. Fig. 3 (a), (b) and (c) show the FESEM images taken from the fractured surfaces with liquid nitrogen of PP composites filled with 30 wt% of WGR powder, filled with 50 wt% of WGR powder and 10 wt% PP-g-MA and filled with 30 wt% of WGR powder and 5 wt% PP-g-MA, respectively. As can be seen in the images in Fig. 3 (c) in comparison with those in Fig. 3 (b), despite the increase of weight percentage of tire powder, tire powder particles which are white in PP matrix still had a good dispersion and small size. This result arose from the increased compatibility of tire powder with the PP matrix due to the presence of PP-g-MA. In fact, the surface of tire powder reacted with PP-g-MA, which had a compatibilizer role so that the cross links in the WGR were broken under mechanical mixing in the extruder, the MA polar group in PP-g-MA reacted with the phenolic OH group in WGR and non-polar part of PP-g-MA reacted with non-polar PP [11-14]. This chemical interaction led to improved compatibility and adhesion between PP and WGR as well as better dispersion of tire powder in PP matrix [1, 15, 16].

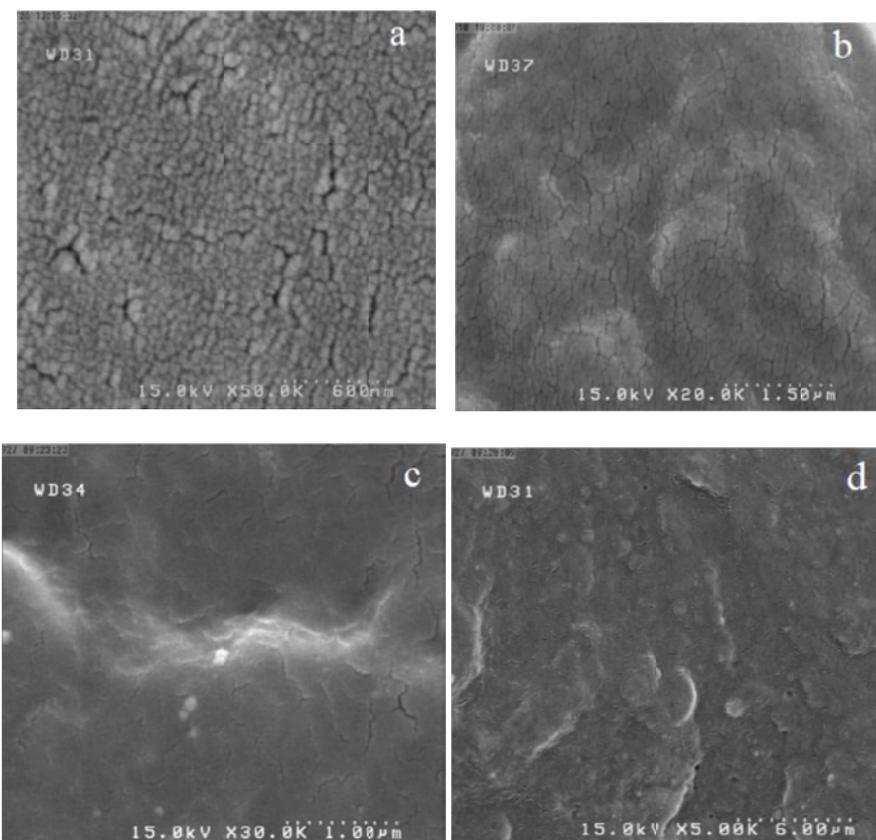


Fig. 2. FESEM images taken from the fractured surfaces with liquid nitrogen of modes without filler: a) PP, b) PP-g-MA, c) 90/5g and d) 90/10g.

The comparison between Fig. 3 (c) and Fig. 3 (b) indicates the less weight percentage of tire powder in matrix volume unit. Apparently, decreasing weight percentage of PP-g-MA led to the dispersion reduction of WGRT. Vectors in the figure point to the larger tire particles and weak dispersion of tire particles. In order to see the rubber phase in PP matrix, surfaces of all combinations, including tire powders, were broken in nitrogen and were etched to remove the rubber phase. Previous studies using periodic acid were suitable for this work [22, 23]. In the present work, first, according to daCosta et al.'s [23], the compounds containing WGRT were immersed in 1M hydrochloric acid for 24 h. But, after doing this process in the FESEM images of fracture surface of pieces, there was no trace of etching rubber particles. Therefore, the time interval of the process increased to 3 days and then a week. However, in both recent periods, the desired result was not achieved. In the next step, the 3M hydrochloric acid was used instead of 1M acid in the timeframe of a week. Also at this stage, elimination of rubber particles from the PP matrix was not possible. Finally, immersing parts including WGRT in 12M hydrochloric acid for a week made observation of rubber particles in FESEM images possible. Although rubber particles were not removed from the matrix, seemingly, only the reaction of the carbon black existing in the structure of WGRT with the acid only led to observing rubber particles in the matrix. Periodic acid can removed unsaturated rubber through the reaction with double bonds [22].

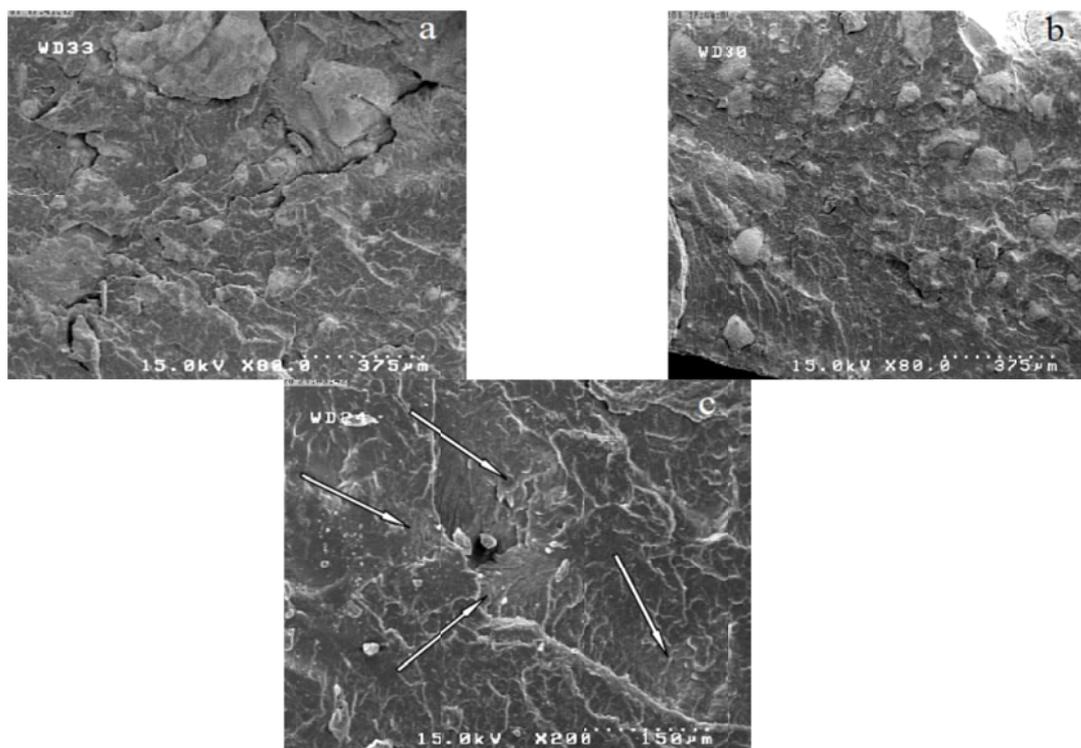


Fig. 3. FESEM images taken from the fractured surfaces with liquid nitrogen of composites filled with tire powder and PP-g-MA: a)70/30W, b)40/50W/10g and c)65/30W/5g.

3.2. Experimental analysis

3.2.1. Effect of WGRT

Figure 4 displays Izod impact strength, yield stress and tensile strength of pure PP, PP-g-MA, WGRT and also binary blends of PP/PP-g-MA and PP/WGRT in different weight percentages. As can be seen, the addition of waste tire powder to the PP matrix leads to an increase of the impact strength. Rate of increase in higher percentages of filler becomes more. This shows the positive impact effect of tire powder in increasing the impact strength of the PP. In general, the elastic nature of rubber particles must increase the impact strength of PP matrix. In addition, because of the elastic nature of the rubber particles they also may increase impact strength through hole creation after particle separation mechanism. Thus the particle separation provides free volume in the order of particle size and the stress state around the particle can change. This results in the plastic yielding of bonds around the particles through the shear yielding of polymer matrix and increases the energy absorption capacity of material [24].

Generally, in compounds containing WGRT, poor adhesion of these particles to the matrix and insufficient dispersion of particles (because of their large size) may be reasons for decreasing of the impact strength [1]. An increase in impact strength even at high percentages of tire powder indicates good dispersion of particles in the matrix. According to the elastic properties of the WGRT that is a factor for increasing the matrix impact energy, formation of strong bonds between the particles and matrix may also make good interfaces for stress transfer between the matrix and the fillers. Generally particle size, good distribution and dispersion of particles in the matrix and the good adhesion between the particles and matrix are affective in raising the impact energy of the particles filled compounds.

Yield stress and ultimate tensile strength of binary combinations of PP and WGRT can be seen in Fig. 4. In both cases, the properties show a decreasing trend with increasing WGRT weight percent. Reduction of yield stress and ultimate strength of rubber-filled PP compared with pure PP

mode can be caused by separation of the rubber particles before yielding and rubber plasticize property. Also the poor adhesion between tire particles and matrix can reduce both yield stress and ultimate strength due to separation of particles from the matrix.

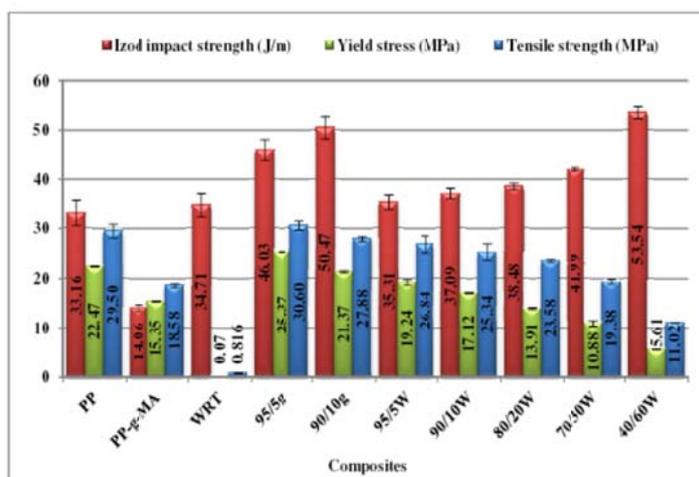


Fig. 4. Izod impact strength, yield stress and tensile strength of pure PP, PP-g-MA and waste rubber (w) and also binary blends of PP/PP-g-MA and PP/w in different weight percentages.

Fig. 5 displays Young's modulus and elongation at break of pure PP, PP-g-MA and WGRT and also binary blends of PP/PP-g-MA and PP/WGRT in different weight percentages. Low modulus of the pure WGRT suggests that this material is too soft. Hence the addition of this soft material to PP matrix leads to a reduction of the matrix modulus. The more tire powder is added to the matrix, the more softness appears in matrix. The poor adhesion between matrix and rubber particles may be one of the reasons for this reduction. As can be seen in Fig. 5, compared to the pure PP, binary combinations have extreme reduction of elongation at break. This originates from the cross linking structure of the rubber and its incompatibility and poor adhesion to the PP matrix and also stress concentration around the rubber particles. These may cause that WGRT shows a weak capacity for stress transfer from PP matrix to the filler particles. In fact cross linking rubber is connected to the other compounds existing in other rubber particles such as carbon black and zinc oxide. This prevents moving of binary combinations compared to pure PP that is a barrier for elongation at break [19].

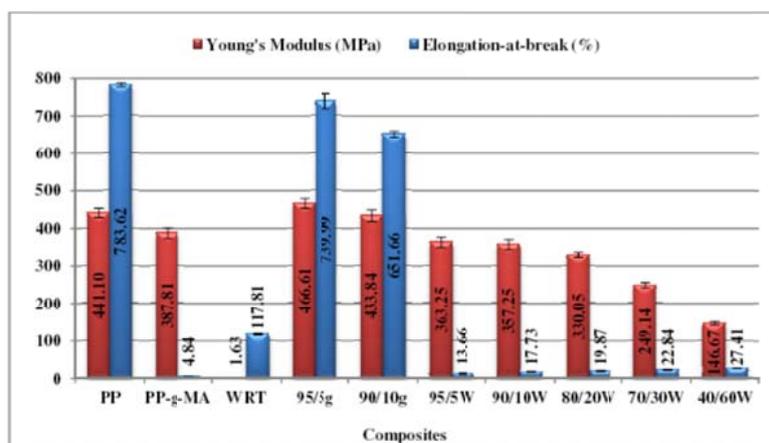


Fig. 5. Young's modulus and elongation at the break of pure PP, PP-g-MA and waste rubber (w) and also binary blends of PP/PP-g-MA and PP/w in different weight percentages.

Tensile strength and elongation at break are related inversely. An increase in tensile strength results in a reduction of elongation at break. In binary combination by increasing the weight percentages of WGRT, elongation at break increases which is because of the elastic property of rubber (as an elastomer). This also increases the softness of the matrix that is caused by using of the soft tire powder.

3.2.2. Effect of PP-g-MA

In this study, the aim of adding PP-g-MA to the matrix was to increase the compatibility of waste WGRT and the PP matrix [15]. To see the effect of adding this material on the mechanical properties of PP matrix, 5 and 10 wt% PP-g-MA was added to a PP matrix. Then, the mechanical properties of binary compounds were compared with the pure PP-g-MA and PP. Impact strength, yield stress and tensile strength of pure PP and PP-g-MA binary compounds are shown in Fig. 4. As can be observed, by increasing PP-g-MA weight percent in the PP matrix, its impact strength increased. It is evident that, in 5wt% of PP-g-MA, impact strength increased by about 39% and, in 10wt% of PP-g-MA, it increased by about 52%. This increase can be caused by the compatibility of PP used in manufacturing PP-g-MA with the PP matrix that established appropriate links between them and formed a uniform matrix. Although the impact strength of PP-g-MA was low, its composition with PP led to the increase in the impact strength.

Yield stress and ultimate strength bars showed the same trend of changes such as the Young's modulus. Apparently, in 5wt% PP-g-MA, tensile properties of PP matrix showed a good improvement that could be attributed to the formation of strong links between PP-g-MA and matrix. But, by increasing PP-g-MA in PP matrix, the increased percentage of maleic anhydride was derived which resulted in the increase of PP-g-MA and led to earlier yielding of compound. This also resulted in the reduction of ultimate strength of the compound.

Young's modulus and elongation at break of pure PP and PP-g-MA binary compounds can be seen in Fig. 5. As can be observed, modulus of PP-g-MA was about 12% lower than PP. This was a piece of evidence for the low stiffness of the PP-g-MA with respect to PP.

Apparently, the addition of small amounts of PP-g-MA increased the binary compounds' modulus. This might be due to the creation of strong links between the PP and PP-g-MA. The more PP-g-MA was added to the PP matrix, the less the modulus of the compound would be reached, which was because of the low modulus of PP-g-MA.

3.2.3. Effect of simultaneous presence of WGRT and PP-g-MA

Fig. 6 shows impact strength, yield stress and tensile strength of pure and ternary combinations of PP, WGRT powder and PP-g-MA. Accordingly, adding WGRT to the PP matrix led to an increase in the impact strength. All binary and ternary compounds had higher impact strength than pure PP. The binary combinations also had higher impact strength than ternary ones. Also, in all ternary compounds, increase of PP-g-MA simultaneously with the reduction of WGRT reduced the impact strength of the samples. Although this reduction was low, in the ternary blends containing 5wt% PP-g-MA, substantial changes occurred which could be caused by strong bonding between rubber particles and PP matrix. The presence of PP-g-MA increased compatibility of WGRT powder with PP matrix and particle adhesion to the matrix and, as noted earlier, this compatibility was coming from reactivity of MA groups in PP-g-MA and phenolic OH groups in the WGRT powder. Indeed, the PP-g-MA was placed in the interface of tire powder and PP matrix. As mentioned above, in ternary compounds, reducing WGRT powder or increasing PP-g-MA decreased impact strength of the samples. This might be due to the reduction of rubber phase, as a factor of increasing the impact energy, and also because of increasing PP-g-MA brittle phase in compounds.

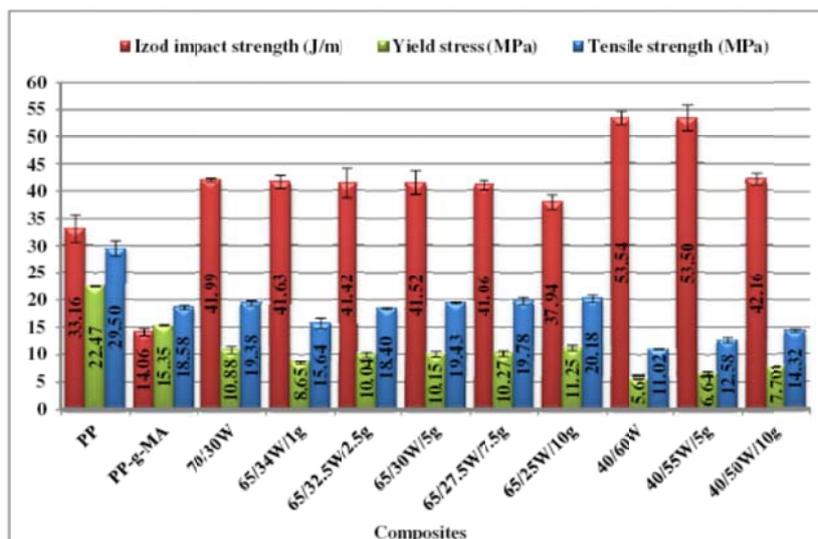


Fig. 6. Izod impact strength, yield stress and tensile strength of pure and ternary combinations of PP, W and PP-g-MA, in different weight percentages of tire powder and PP-g-MA.

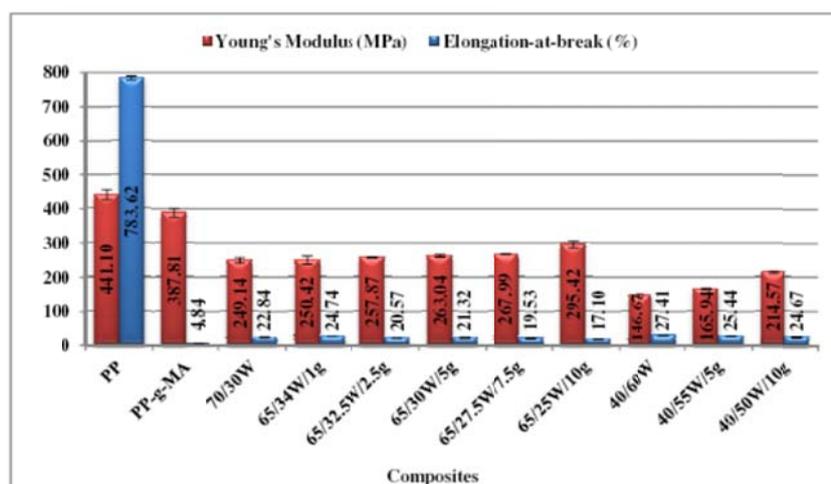


Fig. 7. Young's modulus and elongation at break of pure and ternary combinations of PP, W and PP-g-MA, in different weight percentages of tire powder and PP-g-MA.

Also, by increasing PP-g-MA simultaneously with the decrease of WGRT powder, strong bonds were created between rubber particles and matrix, which impaired the matrix toughening mechanism through particle separation and may also decrease impact energy. In Fig. 7, both yield stress and tensile strength increased in ternary combinations. This was because of reducing rubber powder and increasing the PP-g-MA. But, this increase was more pronounced in higher weight percentages of tire powder.

Fig. 7 shows Young's modulus and elongation at break of pure and ternary combinations of PP, W and PP-g-MA. It can be seen that, in ternary compounds, reduction of WGRT powder and increase of PP-g-MA resulted in the increase of modulus of the compound due to the reduction of soft rubber phase in the matrix and also creation of strong bonds of rubber particles with the matrix. Generally, by reducing rubber and increasing PP-g-MA in compounds, a reduction happens in elongation at break. This is due to increase of compound brittleness because of the creation of strong bonds and also for brittle nature of PP-g-MA and reduction of the compound softness due to the reduction of soft rubber phase from the matrix. Only in the modes containing 1

and 5 wt% PP-g-MA, an increase in elongation at break was observed that could be resulted in the creation of strong bonds between rubber particles and the matrix due to the consistent effect of PP-g-MA.

4. Conclusions

In this research, the effect of WGRT powder on mechanical properties of PP was experimentally investigated. The FESEM micrographs indicated that PP-g-MA led to the compatibility increase of WGRT powder with the PP matrix, better dispersion of tire powder and preventing from their agglomeration in the matrix.

Adding tire powder to PP matrix in all binary and ternary combinations yielded increase of impact strength of the compounds. The rate of this increase was more in higher filler percentages. Increase of the impact strength even in high percentages of the WGRT powder indicated good dispersion. In the ternary compounds consisting of PP, wear WGRT powder and PP-g-MA, increase of PP-g-MA led to the decrease in the impact strength of the compounds. Due to the brittle nature of PP-g-MA and establishing strong bonds between WGRT powder particles and the matrix, the impact strength of the compounds decreased. By decreasing the WGRT powder or increasing the amount of PP-g-MA, Young's modulus, yield and tensile strength increased and elongation at break decreased. Generally, in 5 wt% of PP-g-MA, mechanical properties had significant improvement, which could be attributed to the formation of appropriate bonds between PP-g-MA and matrix. But, by increasing PP-g-MA, a loss in mechanical properties of the compound happened because of the increase of maleic anhydride in PP matrix.

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