# Article

INVESTIGATION OF ADDING POLYMERIC AND INORGANIC MIXTURES TO IMPROVE POLYPROPYLENE PROPERTIES

Seyed Javad Ahmadpanah<sup>1</sup>, Saeid Shokri<sup>1</sup>, Behrouz Nonahal<sup>2</sup>, Sorood Zahedi Abghari<sup>1</sup>

<sup>1</sup> Process & Equipment Technology Development Division, Research Institute of Petroleum Industry (RIPI), 14665-137, Tehran, Iran

<sup>2</sup> Petroleum Refining Technology Development Division, Research Institute of Petroleum Industry (RIPI), 14665-137, Tehran, Iran

Received July 8, 2017; Accepted October 19, 2017

#### Abstract

Some elastomers and fillers are used to optimize polypropylene properties. In this research, the effects of different fillers and elastomers are investigated on mechanical properties of polypropylene. The results show that impact and tensile strength of polypropylene are improved by applying polymeric and inorganic additives. Also, these additives improve thermal properties of polypropylene. Addition of low density polyethylene (LDPE), high density polyethylene (HDPE) or copolymer styrene -butadiene is more resistant mechanically than physical blend of polypropylene and polyethylene. Also, calcium carbonate modified by a titanate coupling agent, improves polypropylene properties. The experimental results indicate that copolymer styrene-butadiene-styrene (SBS) improves polypropylene mechanical properties more than copolymer ethylene –vinyl acetate (EVA). Finally, mechanical properties of copolymer polypropylene compounds were investigated and the results showed an improvement in mechanical properties compared to polypropylene compounds.

Keywords: Polypropylene; Fillers,; Copolymer; Alloys; Calcium Carbonate; Titanate.

### 1. Introduction

Polypropylene is vastly used as sheets and molding materials as it is resistant to water, gasoline and is considered a good chemical resistant. Besides the above advantages, polypropylene has a low volumetric mass, good mechanical properties and low production cost. But polypropylene suffers from weak impact strength at low temperatures. Therefore in order to improve and modify this property, different elastomers and polymers are added to polypropylene. For example, terpolymer ethylene-polypropylene-dn (EPDM) improves the impact strength of polypropylene<sup>[1-3]</sup>. In addition, butadiene-dn-styrene (BDS) also modifies impact strength of polypropylene. Various researches have proved that impact strength property of mixture of polypropylene with trans-polyoctonum (TOR) improves in comparison to unmodified polypropylene<sup>[4]</sup>. In combination of propylene with other polymers, compatibility is an important factor. Some light polymers create good compatibility with polypropylene. Overall, the most appropriate case in combination of polypropylene with other polymers is to create a covalent bond between polypropylene with the added polymer <sup>[5-6]</sup>. For example, although polypropylene's strength increases through mixing with polyethylene but the property of co-polypropylene – ethylene is better than property of polypropylene –ethylene alloy because in co-polypropyleneethylene, a new polymeric chain is created as a result of covalent bond <sup>[7]</sup>.

By adding some filler like talc, calcium carbonate, titanium oxide, magnesium hydroxide, and fiber glass, polypropylene applications can be developed <sup>[8]</sup>. Using fillers, the miscibility and cohesion of fillers with polypropylene matrix are influential factors on structure and property of polypropylene and any change in improving those factors will lead to an improvement in mechanical properties of polypropylene. In addition, by processing of fillers' surface, the interaction between polypropylene and filler will also be improved <sup>[9-12]</sup>. For this purpose, the following groups are mainly used:

- 1. Fatty acids (saturated and unsaturated)
- 2. Active surface compounds like cationic, anionic and nonionic active agents
- 3. Coupling agents especially titanate coupling

# 2. Experiments

## 2.1. Materials

The melt index used for polypropylene and polyethylene were 0.5 and 0.3 gr/min respecttively and the size of calcium carbonate consumed was more than 10  $\mu$ m. The molecular weight of low polyethylene (LMPE) and low polypropylene (LMPP) were 5000 and 6500 respectively which were prepared by thermal cracking. Further consumed materials were copolymer styrene-butadiene-styrene with a linear structure and molecular weight of around 9000 (rate of styrene of 65%), copolymer ethylene-vinyl acetate with molecular weight of 28000 (rate of vinyl stat of 26%) and polystyrene of 40000 molecular weight.

### 2.2. Preparation of Laboratory Samples

First, calcium carbonate and polypropylene (15% weight) were heated for 5 minutes up to 100°C in a mixer equipped with a mechanical agitator. Then, titanate coupling agent was blended with calcium carbonate to process its surface. In the later stage, polypropylene and other polymers were used, to be added to the mixture and the final compound was melted and mixed at 210°C in a rotating twin screw extruder. Finally, the samples were prepared by injection moldings to measure mechanical properties. The combinations of prepared samples are reported in Table 1.

Polymer	PP	LDPE	EVA	SBS	LMPP	LMPE	
Comps.							
A	100	0	0	0	0	0	
В	100	20	0	0	0	0	
С	100	20	6.4	0	2.5	0	
D	100	20	6.4	3.6	2.5	0	
E	100	20	6.4	3.6	0	2.5	

Table 1. Compounds of prepared different samples

### 2.3. Measurement of mechanical properties

Following preparation of samples, tensile strength was measured by XL-100 equipment according to ASTM D638 standard. Also impact strength was measured by 40-Izod Uj impact equipment according to ASTM 256.

The flexibility of samples was measured by LJ-500 equipment according to ASTM D790 and thermal deviation of samples was also measured in conformance with ASTM D648. All experiments were carried out at 23°C and the mean values were reported for each experiment. The results of experiments carried out in tabulated combinations are reported in Tables 1 and 2.

### 3. Analysis of results

As it can be seen from the Table 2, different polymers have different effects on polypropylene properties. Copolymer styrene-butadiene-styrene (SBS) and polyethylene (LDPE) give polypropylene the most powerful impact strength because low density polyethylene (LDPE) is a severely branched polymer with high flexibility. Also the interaction between copolymer styrene-butadiene-styrene and polyethylene and polypropylene is stronger between polypropylene and copolymer ethylene vinyl acetate.

Compounds	Tensile	Coefficient of	Thermal Deviation	Impact Strength
	Strength, (MPa)	Flexibility, (MPa)	(°C)	(J/M)
А	37.4	1683	117.5	24.9
В	25.6	1005	91.3	400
С	29.2	1168	105.3	36.9
D	23.6	588	86.9	456
E	23.3	806	82.0	600

Table 2. Mechanical properties of polypropylene combined with other polymers

Since stress transfer depends on the size of common surface and the thickness of the boundary layers of different phases, therefore tensile yield stress in heterogeneous polymeric systems is considered a useful property to predict interactions on common surface. Hence, tensile yield stress is loaded by reduced cross section and is determined by load borne on dispersed phase. As it can be observed from Table 2, thermal properties of polypropylene are reduced as a result of combining with other polymers. On one hand, due to low flexibility power of added polymers, the coefficient of flexibility of polypropylene compounds with other polymers is lower than unmodified polypropylene. The results show that modification of propylene impact strength can be obtained through adding low density polyethylene (LDPE) to polypropylene. But polyethylene reduces other polypropylene severely because PP has a relatively low tensile strength. Therefore, in other experiments, high density polyethylene (HDPE) has replaced low density polyethylene (LDPE). In addition, in order to improve flexibility and tensile strength of polypropylene, about 30% weight of calcium carbonate was added to the above mentioned compounds in Table 1. As it can be seen from Table 2, the effect of low polyethylene (LMPE) on the polypropylene properties is more suitable than low polypropylene (LMPP). Thus, in the next experiment, as well as calcium carbonate, the effect of addition of low polyethylene (LMPE) and co-polymer ethylene vinyl acetate (EVA) was investigated with the same weight percent as in the previous experiment for the properties of polypropylene. The prepared compounds are as follows:

 $\alpha$  - PP + CaCO3 + HDPE + EVA

 $\beta$  - PP + CaCO3 + HDPE +EVA + LMPE

 $\gamma$  - PP + CaCO3 + HDPE + EVA + LMPE +SBS

Further experiments were carried out on the above compounds and the results are shown in Table 3.

Compounds	Tensile Strength (MPa)	Coefficient of Flexibility (MPa)	Thermal Deviation (°C)	Impact Strength (J/M)
α	28.2	1490	110	40
β	31.1	1570	112	43
γ	29.5	1300	97	32

Table 3. Mechanical properties of polypropylene combined with calcium carbonate

As it can be observed in Table 3, calcium carbonate is produced due to strength of interfacial layers that is resulted from processing with a titanate coupling agent which enhances the strength of the polypropylene alloys considerably while unmodified thermal and tensile properties of polypropylene is lower. In addition, copolymers ethylene vinyl acetate, styrene-butadiene-styrene and polyethylene improve impact strength of polypropylene.

Since, no chemical reactions has taken place between polypropylene with other polymers even, low density polyethylene (LDPE), polypropylene property has not changed considerably. Thus, in the next experiment, the effect of polypropylene copolymer was investigated on the mechanical properties of different compounds shown in Table 4. The experiments were carried out on the compounds highlighted in table 4 and the results obtained are shown in table 5.

Compounds	PP	PS	HDPE	SBS	CaCO3	Titanate
	Copolymer					Coupling Agent
1	100	0	30	0	50	0.5
2	100	30	0	5	0	0
3	100	30	0	5	50	0.5
4	100	30	0	5	0	0

Table 4. Polypropylene copolymer compounds with other polymers

Table 5. Mechanical properties of polypropylene combined with calcium carbonate

Compounds	Tensile Strength	Coefficient of	Thermal	Impact Strength
	(MPa)	Flexibility (MPa)	Deviation (°C)	(J/M)
1	150	2700	156	270
2	110	2150	139	182.1
3	123	2400	145	196.3
4	861	1960	127	160.7

Since copolymer polypropylene has higher impact strength property in comparison with homopolymer polypropylene, these compounds have higher impact strength with regards to homopolymer polypropylene. By comparing the compounds of 2 and 4 in table 4, it can be seen that slight increase in copolymer styrene-butadiene-styrene with respect to copolymer propylene causes severe increase in copolymer styrene-butadiene-styrene due to an interfacial reaction between copolymer styrene-butadiene-styrene and polystyrene.

High density polyethylene (HDPE) increases impact strength of the compound 5 times of copolymer polypropylene. Also, by comparing the compounds 2 and 4 shown in tables 4, it can be observed that the styrene content in polystyrene cannot improve the impact strength as much as the styrene content within styrene-butadiene-styrene. By comparing the compounds of 1 and 3 in table 4, it can be learned that high density polyethylene has higher tensile strength with respect to styrene and copolymer styrene-butadiene-styrene. In other words, polystyrene does not blend with other polymers uniformly. Calcium carbonates in compounds 1 and 3 helps to enhance the flexibility coefficient property of polypropylene to a great extent.

# 4. Conclusions

With regards to wide applications of polypropylene, the properties of polypropylene compounds are key factors in developing applications of this polymer. Numerous studies have been carried out regarding the effect of addition of polymers and fillers on polypropylene properties. These investigations indicate that compatibilizing agents and methods of blending of different materials with polypropylene are the most important factors in improving polypropylene properties. The combination of polystyrene and copolymer styrene-butadiene-styrene improve polypropylene impact strength while polystyrene is not very effective in this regard. On one hand, the blend of low and high density polyethylene (LDPE) and (HDPE) improve the impact strength properties like the combination of polypropylene with copolymer-styrenebutadiene-styrene to a great extent.

In total, the combination of polyethylene and copolymer polypropylene is the desired combination that improves impact strength, tensile strength and coefficient of flexibility with respect to other compounds. Low polyethylene (LMPE) and low polypropylene enhance the melt flow index of the compound but have no considerable effect on mechanical properties. Copolymer polyethylene vinyl acetate (EVA) does not change the mechanical properties of polypropylene. However, with addition of (EVA) to polypropylene accompanied by acrylic resin improves print properties of polypropylene. Finally, adding calcium carbonate to above compounds improves mechanical property, thermal power of the product as well as lowering the finished price.

#### References

- [1] Watanabe R, Shinzawa H, Kunioka M, Mizukado J, Suda H and Hagihara H. Reinforcement Mechanism of Functionalized Polypropylene Containing Hydroxyl Group Nanocomposites Studied by Rheo-Optical Near-Infrared Spectroscopy. European Polymer Journal, 2017; 92: 86-96.
- [2] Yan ZL, Wang H, Lau KT, Pather S, Zhang JC, Lin G, Ding Y. Reinforcement of Polypropylene with Hemp Fibres. Composites Part B: Engineering, 2013; 46: 221-226.
- [3] Ple O, Le TNH. Effect of Polypropylene Fiber-Reinforcement on the Mechanical Behavior of Silty Clay. Geotextiles and Geomembranes, 2012; 32: 111-116.
- [4] Essabir H, Bensalah MK, Rodrigue D, Bouhfid R and Qaiss AL. Comparison Between Bio and Mineral Calcium Carbonate on The Properties Of Polypropylene. Composites Construction and Building Materials, 2017; 134: 549-555.
- [5] Suharty NS, Ismail H, Diharjo K, Handayani DS, Firdaus M. Effect of Kenaf Fiber as a Reinforcement on the Tensile, Flexural Strength and Impact Toughness Properties of Recycled Polypropylene/Halloysite Composites. Procedia Chemistry, 2016; 19: 253-258.
- [6] Serrano A, Espinach FX, Tresserras J, Pellicer N, Alcala M, Mutje P. Study on the Technical Feasibility of Replacing Glass Fibers by Old Newspaper Recycled Fibers as Polypropylene Reinforcement. Journal of Cleaner Production, 2014; 65: 489-496.
- [7] Mohammadhosseini H, Abdul Awal ASM, Mohd Yatim J B. The Impact Resistance and Mechanical Properties of Concrete Reinforced with Waste Polypropylene Carpet Fibres. Construction and Building Materials, 2017; 143: 147-157.
- [8] Wang K, Bahlouli N, Addiego F, Ahzi S, Rémond Y, Ruch D, Muller R. Effect of Talc Content on the Degradation of Re-Extruded Polypropylene/Talc Composites. Polymer Degradation and Stability, 2013; 98: 7, 1275-1286.
- [9] Lopez-Buendia AM, Romero-Sanchez MD, Climent V, Guillem C. Surface Treated Polypropylene (PP) Fibres for Reinforced Concrete. Cement and Concrete Research, 2013: 54, 29-35.
- [10] Sole BM, Ball A. On the Abrasive Wear Behaviour of Mineral Filled Polypropylene Tribology International. 1996; 29: 6, 457-465.
- [11] Szabo JS, Czigany T. Static Fracture and Failure Behavior of Aligned Discontinuous Mineral Fiber Reinforced Polypropylene Composites.PolymerTesting. 2003; 22: 6, 711-719.
- [12] Hadal R, Dasari A, Rohrmann J, Misra RDK. Susceptibility to Scratch Surface Damage of Wollastonite- and Talc-Containing Polypropylene Micrometric Composites. Materials Science and Engineering: A. 2004; 380: 326-339.

To whom correspondence should be addressed: Prof. Saeid Shokri, Process & Equipment Technology Development Division, Research Institute of Petroleum Industry (RIPI), 14665-137, Tehran, Iran, Tel: +98 (21) 48252540; Fax: +98(21) 44739713, E-mail address: shokris@ripi.ir