

## ENHANCING THE PHYSICO-MECHANICAL/RHEOLOGICAL PROPERTIES OF ASPHALT 60/70 IN DIFFERENT MEDIA VIA MA-PP COMPATIBILIZED POLYPROPYLENE/ACRYLONITRILE BUTADIENE RUBBER BLENDS (PP/NBR)

M. M. Badr<sup>1</sup>, M. El-Shafie<sup>2</sup>, A. A. Ragab<sup>2</sup>

<sup>1</sup> Petrochemical Department, Egyptian Petroleum Research Institute (EPRI), Nasr City, Cairo, Egypt

<sup>2</sup> Petroleum Applications Department, Egyptian Petroleum Research Institute, Nasr City, Cairo, Egypt

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### Abstract

Elastic reverse deformation and enhance the mechanical / rheological properties at low temperatures are desired in asphalt modification. So, thermoplastic elastic blends 3, 5, 7 and 10% PP/NBR with and without using PP-MA were used. PMAs were evaluated using Penetration Index (PI), temperature susceptibility, shear stress / shear rate, dynamic viscosity and mechanical properties in different media air and water, tensile shear strength and elongation. The results showed that the addition of polymers to asphalt reduces its thermal susceptibility at different temperatures since the asphalt become more elastic, PMAs have thixotropic property, high yield stress, good gunnability and lack of flow on vertical surfaces. It is found also, the tensile shear strength for the obtained blends increases with polymer content with using PP70/MA more than PP70. In both water and air PP70 & PP70/MA enhancing the mechanical properties of asphalt but in air is the best, while the suitable ratio is 7 & 5% according to workability. Finally, PMAs have a higher tensile strength means asphalt can tolerate higher strains before failing (i.e. cracking). Additionally, the moisture susceptibility of the asphalt mixture can be evaluated by comparing the tensile strength of asphalt mixtures exposed to wet and dry conditions.

**Keywords:** Asphalt; thermoplastic elastic; polypropylene, enhancing; susceptibility; rheological.

### 1. Introduction

The history of polymer blends is quite long and strictly correlated with that of polymers themselves [1]. Along with over increasing growth in the application of engineering polymers such as polymer blends [2] composites [3-5] and nanocomposites [6], understanding of their mechanical behavior under long-term loading conditions becomes more significant and challenging.

Indeed, blending two or more polymers may give rise to new polymeric systems with new, interesting properties, different from those of the two components, thus eliminating the long and expensive route of synthesizing new polymers [7-10]. Nevertheless, an important shortcoming in blending different polymers is that the polymers often form immiscible and incompatible blends, displaying a coarse morphology with particles of dispersed phase badly adherent to the matrix and badly distributed. Consequently, a compatibilization process is necessary in order to modify the interfacial properties in the immiscible polymer blend, thus leading to a reduction of the interfacial tension coefficient and the formation and stabilization of the desired morphology. Several compounds have been used as compatibilizers including, more recently, nanoparticles that can modify the final morphology of the polymer blend [1]. A recent outstanding book [11] outlines the behavior of many polymer blends used in several applications or just investigated.

The simple blending of crystalline thermoplastic and elastomeric polymer results in a new class of material termed a thermoplastic olefin (TPO). The properties of the resultant TPO will be derived from the properties of each of the two polymers and dependent on the composition

and the interaction between their phases [12–15]. An approach to further improve the performance of TPO is to develop dynamically vulcanized TPO [16]. During dynamic vulcanization, the rubber is vulcanized in the presence of the molten thermoplastic under shear forces. Since crosslinked rubber is unable to coalesce, rubber particles are dispersed in the thermoplastic matrix even at high rubber content [16]. The effect of the dynamic vulcanization on morphology and mechanical properties has been extensively studied, and some important reviews are available in the scientific literature. Acrylonitrile butadiene rubber (NBR) is a commonly used oil-resistant rubber. Polypropylene (PP)/NBR-based TPO combines the oil resistance of NBR as well as excellent chemical and moisture resistance, good ductility and stiffness, low density, and easy processing characteristics of PP [17]. The large difference in the solubility parameter between the polar NBR and the non-polar PP leads to poor compatibility of blends. As a result, PP and NBR are highly incompatible and usually exhibit phase-separated morphology because of poor physical, mechanical and chemical interactions across the phase boundaries [18]. The interfacial adhesion and mechanical properties of PP/NBR blends can be improved by the addition of compatibilizers or interfacial agents. Phenolic-modified PP and maleic anhydride-modified PP are two main types of a compatibilizer for PP/NBR blends. George *et al.* [19] have reported several works about the effect of phenolic-modified PP and maleic-modified PP as compatibilizers on the properties of PP/NBR blends.

Thermoplastic elastomers (TPEs) are a class of copolymers or a physical mixture of polymers (usually a plastic and a rubber) which consist of materials with both thermoplastic and elastomeric properties. TPEs have attracts more attentions in the industry since they combine the properties of vulcanized rubber with the ease of processing of thermoplastics [20–25]. Styrene-butadiene-styrene copolymer (SBS) is a typically synthesized copolymer and the largest TPE being used as asphalt modifier because of its good compatibility with asphalt, workability and high performance.

However, the poor oxidation resistance caused by the presence of double bonds in the backbone of SBS and high cost are the main challenges before selecting it as asphalt modifier [26–30]. Therefore, more competitive TPEs are deserved to develop for asphalt modification.

Maleic anhydride and irradiation approach are also used to improve compatibility [31]. Magioli [20] has reported using dynamic vulcanization technology to enhance the properties of TPE.

In modification of a paving asphalt, it is desirable to increase its softening point, make penetration less temperature-dependent, make asphalt less susceptible to elastic reverse deformation, increase the ductility (especially at low temperatures), enhance the adhesion of the asphalt to mineral fillers of asphalt concrete, and extend the range of working temperatures [32]. To this end, it is first necessary to ensure good compatibility of rubbers with asphalts. However, elastoplastics are not all compatible with asphalts. Their compatibility depends on the chemical composition of both asphalts and elastoplastics.

It is known that high-molecular-weight rubbers to be compounded with asphalt require, for swelling and subsequent dissolution, a larger amount of oils than that contained in paving asphalts. Furthermore, these rubbers (elastoplastics) can induce coagulation of asphaltenes, which are the major structuring components of asphalts; this is inadmissible in the production of high-performance paving asphalts [33–34].

In this paper, thermoplastic elastic blends (PP/NBR in percentages 3, 5, 7 and 10 wt/wt%) use to achieve desired properties in the presence of maleic anhydride (10 wt/wt%) as a compatibilizer for good compatibility blends. PMAs were evaluated via Physical characterization (PI, temperature susceptibility, and routine tests), rheological (shear stress/ shear rate, dynamic viscosity) and mechanical properties in different media (air and water) (Tensile shear strength and elongation). The results showed that as expected the addition of polymers to asphalt reduces its thermal susceptibility at temperatures and guarantees a greater constancy of its properties since the asphalt become more elastic. Increasing the ratio of polymer added found to improve the physical and rheological properties, rheological data PMAs indicates that the PMAs have thixotropic property and high yield stress, good gunnability and lack of flow on vertical surfaces. It is found. Also, the tensile shear strength ( $\text{N/cm}^3$ ) for the obtained blends

increase with polymer % content and for that forming with using PP70/MA more than that with using PP70 so, addition of TEPs (PP70& PP70/MA) enhancing the mechanical properties of virgin asphalt 60/70 in both water and air but in air is the best. Finally, polymer modified asphalt samples have a higher tensile strength means asphalt can tolerate higher strains before failing (i.e., cracking). Additionally, the moisture susceptibility of the asphalt mixture can be evaluated by comparing the tensile strength of asphalt mixtures exposed to wet and dry conditions.

## 2. Experimental

### 2.1. Materials

Local virgin asphalt cement of penetration grade (AC 60/70) produced by El-Nasr Petroleum Company in Suez, Egypt. PP70/ NBR30, PP70/NBR30/10% MA which prepared according to Motawie *et al.* [35].

### 2.2. Experimental procedure

The testing program included the following steps.

#### 2.2.1. Characterization of virgin asphalt 60/70

The virgin asphalt sample was tested as illustrated in Table 1 for penetration (ASTM D5), softening point (ASTM D36), specific gravity (ASTM D70), Brookfield viscosity (ASTM D4402) and n-heptane insoluble (ASTM D3297). The results are illustrated in Table 1.

Table 1. Physical and chemical properties of asphalt samples AC 60/70 used for modification

Physical properties of asphalt samples AC 60/70					
Penetration 0.01mm 25°C	Ductility, cm	Kinematic viscosity 135°C, C.st	Absolute viscosity 60°C	Softening point, °C	Specific gravity, g/cm <sup>3</sup>
62	150	259	1088	50.6	1.1763
API	Penetration temperature susceptibility	Penetration index	Specific heat Btu pound per °F	Thermal conduc- tivity	
-11.20	0.0273	-0.51	0.391	0.6330	
Chemical properties of asphalt samples AC 60/70					
Wax %	Oil %	Resin %	Asphaltene content %		
6.88	28.90	55.10	16.41		

#### 2.2.2. Preparation and evaluation of PP/NBR blends

PP/NPR blends of various weight compositions with and without compatibilizers were prepared with a Brabender plasticorder (PL 2100, 350S). At the start, PP was introduced into the Brabender plasticorder preheated to 180°C for 2 min, the tested compatibilizer was incorporated, and NBR was added thereafter. The mixer was operated at 180°C; the rotor speed is maintained at 60 rpm for 6 min. Upon using MAH, benzoquinone (0.75 parts) and dicumyl peroxide (3 parts) were added in a Brabender plasticorder at 180°C with the above mentioned technique 5,16 giving maleic acid-modified PP (MA-PP) as a compatibilizer.

#### Property evaluation

The tensile strength (rR), elongation at break (eR), and young's modulus (E) were determined with a universal tension testing machine (type M-10 Hunge Ta Instruments). The compressed sheets were cut into dumbbell-shaped specimens with appropriate punching dies with a width of 4 mm (DIN 53504). The specimens, with a width of 4 mm, a neck length of 50 mm, a thickness of 1–1.5 mm, were tested at a crosshead speed of 50 mm/min.<sup>17</sup> The hardness of test specimens at least 6-mm thick was measured with a shore A durometer (PTC instrument of Model 306L18). Mass swell percentages of the test pieces were carried out in the

benzene/acetone solvent mixture. About 0.1–0.2 g of each specimen (square test pieces with 5-mm dimensions and 1-mm thick) was weighed in a weighing bottle, which was covered with benzene/acetone solvent mixture 50/50 for 24 h. The swollen samples were weighed and then dried in an oven to a constant weight. The last weight was taken as the correct weight of the sample free from dissolved matter. The mass swell percentages  $Q$  of the samples were calculated as follows:

$$Q = [(m - m_0)/m_0] \times 100 \quad (1)$$

where  $m$  and  $m_0$  represent the weights of the samples after swelling and original one, respectively.

All these tests were performed at room temperature ( $25^\circ\text{C} \pm 1^\circ\text{C}$ ), and the reported results were averaged from a minimum of five specimens.

### 2.2.3. Evaluation of polymer blends carried out according to the following subsections

#### 2.2.3.1. Mechanical measurements for polymer blends with and without MA as a compatibilizer

Tensile strength ( $T_s$ ) MPa, elongation at break ( $E_b$ ) %, absorbed energy Kgf.mm, Young's modulus MPa, and hardness, were measured for the all prepared samples.

Table 2. Mechanical Properties of PP/NBR and PP70/10 MA Binary Blend Samples with Different Ratios of Blending

Sample	Ratio PP : NBR		Tensile Strength (MPa)	Elongation at Break (%)	Absorbed Energy (Kgf.mm)	Young's Modulus (MPa)	Hardness Shore A
	PP	NBR					
PP <sub>100</sub> (Pure PP)	100	0	33.2	15.2	206.6	518.6	96
PP <sub>90</sub>	90	10	25.0	66.6	182.6	437.5	96
PP <sub>80</sub>	80	20	21.3	80.1	164.9	366.3	96
PP <sub>70</sub>	70	30	17.2	96.3	137.7	278.5	96
PP <sub>60</sub>	60	40	13.7	69.8	75.5	219.6	95
PP <sub>50</sub>	50	50	8.7	45.7	47.9	161.3	94
PP <sub>40</sub>	40	60	7.1	41.9	25.6	91.7	90
PP <sub>30</sub>	30	70	3.6	38.6	11.1	52.7	83
PP <sub>20</sub>	20	80	2.7	244.9	5.3	28.1	75
PP <sub>10</sub>	10	90	1.6	663.8	1.8	5.2	64
PP <sub>0</sub> (Pure NBR)	0	100	0.8	1250.0	0.2	1.6	31
PP70/ 10% MA	70	30	20.3	75.0	217.6	576.0	99

### Stress-strain measurements

Five individual dumbbells shaped specimens for the tensile test were cut using a steel die of standard width 4 mm and length of 50 mm by Punch Press model 1701 Germany. The stress strain behavior of the test specimens prepared from vulcanized rubbers film samples with a thickness approximately 1 mm. Thickness measured by Digital thickness gauge, a gauge graduated to one hundred of mm.

Tensile strength ( $T_s$ ), elongation at break ( $E_b$ ), absorbed energy, and Young's modulus was determined directly according to ASTM D638-98, at room temperature using (Tension Machine Type M-10 HUNG-TA, connected with data analysis, at cross-head speed 100 mm/min.

#### a. Tensile strength

It is defined as the force per unit area of the original cross sectional area which is applied at the time of rupture of a specimen and is expressed in MPa [36-37]. The tensile strength of the specimen at break can be calculated as follows:

$$\text{Tensile Strength } (T_s) = (L/T.W) \times 10 \text{ MPa} \quad (2)$$

where:  $L$  is load in kgf, which necessary to cause a break;  $W$  is width of the specimen in mm;

T is thickness of the sample in mm.

### b. Elongation at Break

Elongation at break is an indication of material elasticity, and expressed as the percent elongation of the original distance between benches marks attained at the moment of rupture and is calculated according to:

$$E_b (\%) = (L - L_0)/L_0 \times 100 \quad (3)$$

where: L is length of the specimen at the moment of rupture;  $L_0$  is length between bench marks.

### c. Absorbed energy

The absorbed energy is an indication of the material toughness, which is the amount of energy that material must absorb before breaking. Absorbed energy value determined by experiment according to ASTM D182 and D2289, toughness can be calculated as the area under stress strain curve.

### d. Young's Modulus

Young's modulus of a specimen is a physical constant of the material, gives an indication of material stiffness, and determined by experiment in the same units of stress, i.e., MPa, and can be calculated from values of stress and strain of the initial straight part of the curve, by using the following equation:

$$\text{Young's modulus} = \text{Stress} / \text{Strain} = \text{MPa} \quad (4)$$

## 2. Hardness measurement

Hardness determined for all prepared samples which must have a flat surface, at room temperature. The measurement was applied according to, (ASTM-D 2240), using Durometer of Model 306 L Type A. The unit of hardness is expressed in (Shore A).

### 2.2.4. Modification of asphalt using prepared polymers (PP70 and PP70/MA)

In this step; asphalt was modified via using four percent of PP70 and PP70 + 10% MA (3, 5, 7 and 10 percent by weight of asphalt). The samples were prepared in suitable cans using a high shear mixer rotating at 3000 rpm. The polymer was added gradually at a rate of 5 g/min. The temperature was kept within  $180 \pm 1^\circ\text{C}$  during the polymer addition and subsequent mixing. Then, stirring was performed for 2 hours after complete addition of polymer [38-39].

### 2.2.3. Characterization of polymer modified asphalt samples (PMAs)

#### 2.2.3.1. Physical properties

#### Penetration Index (PI) and temperature susceptibility

The penetration index represents a quantitative measure of the response of bitumen to variation in temperature. Knowing the penetration index of particular bitumen, it is possible to predict its behavior in an application. Therefore, asphalt binders with high penetration numbers (called "soft") are used for cold climates while asphalt binders with low penetration numbers (called "hard") are used for warm climates. All bitumen display thermoplastic properties, i.e. they become softer when heated and harden when cooled. Several equations exist that define the way that the viscosity (or consistency) changes with temperature. One of the best known is that developed by Pfeiffer and Van Doormaal which states that: If the logarithm of penetration, P, is plotted against temperature, T, a straight line is obtained such that [40]:

$$\log P = AT + K \quad (5)$$

where: A is the temperature susceptibility; P is penetration at temperature T; K is constant.

The value of A varies from 0.0015 to 0.06 showing that there may be a considerable difference in temperature response. Pfeiffer and Van Doormaal developed an equation for the temperature response that assumes a value of about zero for road bitumen. For this reason, they defined the penetration index (PI) as:

$$\frac{20-PI}{10-PI} = 50A \quad (6)$$

or explicitly,

$$PI = \frac{20(1-25A)}{1+50A} \quad (7)$$

The value of PI ranges from around -3 for high temperature susceptible bitumens to around +7 for highly blown low temperature susceptible (high PI) bitumen. The PI is an unequivocal function of A, and hence it may be used for the same purpose. The values of A and PI can be derived from penetration measurements at two temperatures, T1 and T2 using the equation:

$$A = \frac{\log \text{pen at } T1 - \log \text{pen at } T2}{T1 - T2} \quad (8)$$

Pfeiffer and Van Doormaal (year) found that most bitumens had a penetration of about 800 dmm (deci-millimetre) at the ASTM softening point temperature. Replacing T2 in the above equation by the ASTM softening point temperature and the penetration at T2 by 800 they obtained equation (9):

$$A = \frac{\log \text{pen } T1 - \log 800}{T1 - \text{ASTM softening point}} \quad (9)$$

Substituting equation (9) in equation (7) and assuming a penetration test temperature of 25 °C gives:

$$PI = \frac{1952 - 500 \log \text{pen } T1 - 20 \text{ softening point}}{50 \log \text{pen} - \text{softening point} - 120} \quad (10)$$

Equations (7) and (9) were applied in this study to calculate first for A (temperature susceptibility of bitumen) and PI (penetration index). These were calculated from the measured softening point temperatures and penetrations.

### 2.2.3.2. Rheology of asphalt using Brookfield viscometer (ASTM D4287, 1994)

The Brookfield DV-III Ultra Programmable Rheometer measures fluid parameters of shear stress and viscosity at given Shear Rates. Viscosity is a measure of a fluid's resistance to flow. The principle of operation of the DV-III Ultra is to drive a spindle (which is immersed in the test fluid) through a calibrated spring. The viscous drag of the fluid against the spindle is measured by the spring deflection. Spring deflection is measured with a rotary transducer. The viscosity measurement range of the DV-III Ultra (in centipoise or cP) is determined by the rotational speed of the spindle.

### 2.2.3.3. Mechanical characteristic; tensile shear strength and elongation measurements for modified asphalt samples using PP70 and PP70/MA

To examine adhesion properties of asphalt and asphalt-polymer blends, shear strength was measured for standard 10 cm length and 2 cm width plain steel plates (bonded area 2.5 cm) solvent degreased after 14 days curing. The tensile shear strength was carried out by Instron testing machine (model 1026) at 23±2°C with cross head speed 100 mm/min, chart speed 200 mm/min and load cell range 0 - 500 Newton according to ASTM D638-77a. Measurements were carried out for samples after being suspended in Air and immersed in water for 24 hrs, at room temperature.

Test procedure [41]:

1. Cut or injection molds your material into one of the five "dumbbell" shapes. The exact shape you use is dependent upon your material's rigidity and thickness.
2. Load the specimen into tensile grips.
3. Attach the extensometer to the sample
4. Begin the test by separating the tensile grips at a constant rate of speed. Speed depends on specimen shape and can range from 0.05 - 20 inches per minute. The target time from start of the test to break should be from 30 seconds to 5 minutes.
5. End the test after sample break (rupture)



Analysis obtained: tensile strength, elongation at yield, elongation at break, nominal strain at break (grip separation), modulus of elasticity, secant modulus, Poisson's ratio (requires transverse extensometer).

### 3. Results and discussion

#### 3.1. Evaluation of blending ratios of PP/ NBR for choosing optimum ratio

##### 3.1.1. Optimization of blending ratios of PP/NBR

Different mixing ratios of PP and NBR are required to study the characterization of PP/NBR blends. Prepared blends of PP/NBR, respectively take these ratios, [(pure PP = PP<sub>100</sub> = 100/0), (PP<sub>90</sub> = 90/10), (PP<sub>80</sub> = 80/20), (PP<sub>70</sub> = 70/30), (PP<sub>60</sub> = 60/40), (PP<sub>50</sub> = 50/50), (PP<sub>40</sub> = 40/60), (PP<sub>30</sub> = 30/70), (PP<sub>20</sub> = 20/80), (PP<sub>10</sub> = 10/90), and (pure NBR = PP<sub>0</sub> = 0/100)], formulations are presented in Table 2.

Mechanical properties measurements were used to evaluate the prepared blend samples. Results of tensile strength, elongation at break, absorbed energy, young's modulus and hardness are presented in Table 2. Mechanical properties measurements were used to evaluate the prepared blend samples. Results of tensile strength, elongation at break, absorbed energy, young's modulus and hardness are presented in Table 2.

From the data obtained it is clear to see that PP has the highest values of tensile strength, absorbed energy, and young's modulus combined with the lowest value of elongation at break, while NBR gives lowest values of tensile strength, absorbed energy, young's modulus, and hardness combined with highest value of elongation at break. These behaviors may be due to the high crystalline content of PP and very low content of crystalline content in NBR, which is an amorphous polymer [42], observed that the crystallinity of the PP/NBR blend was decreased by the incorporation of NBR, from 55.3 for pure isotactic PP to 33.9 and 13.7 for both incorporation 30 and 70 wt/wt% NBR, respectively.

The strength of PP/NBR blends depends on the strength of the PP phase, which in turn depends on the extent of crystallinity of the blend, was decreased by the incorporation of NBR. It is also observed that increasing NBR content in PP/NBR blend decreases absorbed energy and Young's modulus, which may be due to the presence of the soft rubber phase and fall in crystallinity of the PP phase. Hence, decreasing the crystalline content of the blend occurred [43]. have shown that the spherules growth of PP in blends with rubber is hindered by the presence of the rubber phase. It can also be deduced that the tensile strength, absorbed energy and young's modulus values for blends will show a negative deviation, i.e., blend properties lie below the additive line. The negative deviation is due to poor interfacial adhesion between the non-polar PP and polar NBR phases, which causes poor stress transfer between matrix and the dispersed phase. A clear change in the slope of this tensile composition curve is seen between the composition ranges 50/50 to 30/70 PP/NBR. The observed change in mechanical properties composition curves have been reported by [44] for the PP/EPDM system.

In immiscible blends, the elasticity usually depends on the particle size of the dispersed phase. Data indicate that elongation at break slightly increases with the addition of NBR with ratios 10, 20 and 30% in the region of PP<sub>90</sub>, PP<sub>80</sub> and PP<sub>70</sub> blends. It can be considered that NBR with ratio  $\leq 30\%$  act as a plasticizer to PP, that is where particles of NBR in this ratio of blending formed small spheres, which dispersed in the continuous phase of PP. Increasing ratio of NBR from 30 to 40, 50, 60 and 70 increase growth of the particle size of the NBR dispersed phase, which hinders blend to give elasticity, this is clearly described by the continues reduction in values of elongation at break from PP<sub>70</sub> to PP<sub>30</sub>. This decrease in elongation at break at higher rubber content is due to the bigger particle size of the dispersed NBR phase and poor interfacial adhesion between the polymers have shown that elongation at break decreases with increase in particle size of the dispersed rubber phase of PP/EPDM blend. The poor interfacial adhesion causes premature failure as a result of the usual crack-opening mechanism. Smaller and uniformly distributed particles are more effective at initiating crazes and terminating them before they develop into catastrophic sizes. Increasing rubber content

over 70% accompanied with sharply increase in elongation at break continuously with the further addition of NBR. This sharp increase in elongation at break may be due to the continuous nature of NBR phase, which forms a co-continuous structure with the plastic phase; similar results were reported in the case of the PP/EVA system [45]. NBR give the highest value of elongation at break. The elongation at break also shows negative deviation from the additive line.

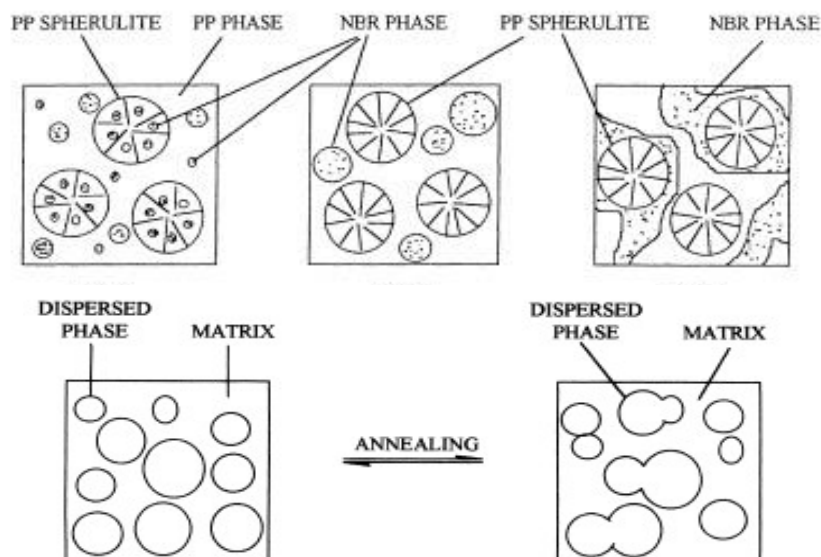


Fig 1. Representation of dispersed phase (NBR) in the matrix of the continuous phase (PP), (George et al. [42]).

It is also observed that hardness values do not give any change with a low ratio of NBR addition (from 10 to 30%), which may be due to high crystallinity of PP not affected with the small particle of NBR formed in the PP matrix as shown in Fig 1. Low rate decrease in values of hardness was observed with increasing NBR concentration from 30 to 70%. Here the particle size of NBR dispersed in the matrix of PP increased, but PP still the continuous phase and formed the matrix. More rate of reduction in hardness values and the slope change in the curve at higher proportions of NBR ( $\geq 70\%$ ) can be explained by the phase inversion of NBR from dispersed to the continuous phase to form co-continuous phase of both PP and NBR on passing from the 50/50 PP/NBR to the 30/70 PP/NBR blend. It is interesting to see that the hardness values show a positive deviation.

It can be deduced clearly from all the above mentioned data of mechanical properties for all ratio of PP/NBR blends that; the sample of PP70 with blending ratio PP/NBR (70/30) has the most reasonable required collection of mechanical properties that is in means of high values of tensile strength, absorbed energy, young's modulus, and hardness combined with high value of elasticity. It is noteworthy that any another sample will have deficient in one or more value of mechanical properties than PP70.

### 3.1.2. Determination of suitable type and ratio of compatibilizers, for PP/NBR (70/30) Blends [35]

The mechanical properties of immiscible polymer blends are affected by the addition of compatibilizers. Data obtained in Table 2 show the variation of tensile strength, elongation at break, absorbed energy, young's modulus, and hardness of 70/30 PP/NBR blend and copolymers with the presence of compatibilizer,

In the case of MA-PP compatibilized copolymers, the increase in tensile strength may be due to the increased dipolar interaction between the MA-PP and NBR phase, which causes an increase in interfacial adhesion between PP and NBR phases, although there is no reduction in particle size with the increase in MA-PP concentration beyond 1%.



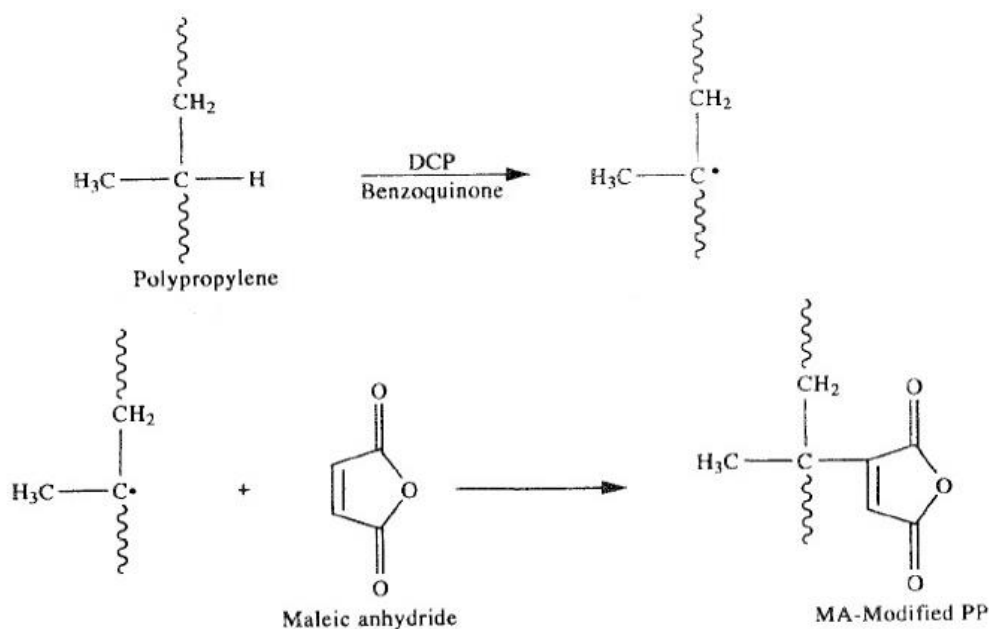
The **tensile strength** of the blends could be predicted by Coran's equation where the MA-PP and Ph-PP are found to act as compatibilizers in PP/NBR system. With the increase in the concentration of compatibilizer, the domain size of the dispersed NBR phase decreases, followed by a leveling off at higher concentrations, which is an indication of interfacial saturation. The theories of Noolandi and Hong predict a linear decrease of interfacial tension with compatibilizer volume fraction for concentrations less than the CMC. Considering the fact that the interfacial tension is directly proportional to the domain size, it is demonstrated that the experimental data are in agreement with these theories.

It is also seen that the **absorbed energy, young's modulus, and hardness** of the blend is found to increase significantly with the addition of compatibilizer. The rate of increasing in these parameters found to increase with a number of  $H_2O_2$  moles added per each mole of oil in an epoxidation process. MA-PP used as compatibilizer gives the highest values of absorbed energy, young's modulus, and hardness

Values of **elongation at break** give noticeable decrease with the addition of MA as a compatibilizer. Further decreases of elongation at break are obtained as a result of the further addition of MA.

### 3.1.3. The mechanism of compatibilization

The mechanism of compatibilization process using PP-MA as a compatibilizer for PP/NBR (70/30) blend, maleic anhydride is the leading monomeric candidate for reactive compatibilization. Particularly in the production of the graft copolymer (PP-MA) that is the effective compatibilizer, this graft copolymer acts as an emulsifier at the interface and thus reduces interfacial tension. The maleic anhydride (MA) groups are grafted onto the PP chain backbone to form maleic anhydride grafted polypropylene MA-PP as shown in Scheme (1) [35].



Scheme 1. The Reaction of maleic anhydride (MA) with polypropylene (PP) to produce maleic anhydride modified polypropylene (MA-PP)

### 3.2. Characterization of polymer modified asphalt samples (PMAs)

PMA samples were prepared as previously mentioned using 3, 5, 7 and 10% PP70 and PP70/MA. The characteristics of all samples will be detected as follows:-

In Table 1 the properties and performance of asphalt 60/70 dramatically change with the polymer modification.

### 3.2.1. Physical characteristics modified samples

Generally, data in Table 3 and Figure 2 indicated that the modified samples are more hardening than the virgin one as there was an increase in softening point, specific gravity, and dynamic viscosities and decrease in penetration value. These results are attributed to the effect of addition both of PP70 and PP70/MA which blended with asphalt. Also, it is noticed from Tables 4 and 5) that, hardness in case of using PP70/MA is more than that recorded in case of using PP70.

With the increase of PP70 levels as 3, 5, 7 & 10 wt/wt% noticed that comparing to virgin AC properties the penetration value of PMAs decreased in percentages of 32.26, 54.8, 62.9 and 72.6%, while softening point increased in percentages of 10.67, 26.48, 34.4 and 44.27% respectively. Also, the dynamic viscosity at 60°C increased in percentages of 338.5, 393.0, 456.4 & 892 and at 130°C increased in values of 6.7, 73.24 & 258.53 respectively. While the penetration indexes increased from -0.51 to -0.21, 0.38, 0.65 & 0.85 respectively, as well as the specific gravity, increased in the same manner.

While, increasing the PP70/MA content from 3 to 10 wt/wt% penetration of PMAs decreased in percentages of 32.3, 56.45, 72.6 and 88.7%, while softening point increased in percentages of 10.87, 42.29, 58.1 and 73.9% respectively with the addition of RRNP as 3, 5, 7 and 10%. Also, the dynamic viscosity at 60 °C increased in percentages of 473.0, 546.4, 678.5 & 1036.5 and at 130 °C increased in percentages of 8.6, 68.24, 196.53 & 221.64 respectively. While the penetration indexes increased from -0.51 to -0.21, 2.83, 3.18 & 4.3 respectively, as well as the specific gravity, increased in the same manner.

This is attributed to nature and accordingly the chemical molecular composition of the PP70/MA which react with asphaltene in the matrix.

### 3.2.2. Temperature susceptibility of all asphalt samples

The temperature susceptibility of the modified samples was investigated by calculating PI values. From Table 3 and Figure 3 with comparing to virgin AC the PI values also, increased from -0.51 for virgin AC to -0.21 & 0.38 & 0.64 & 0.85 for 3, 5, 7 and 10% of PP70 content respectively and -0.21 & 2.83 & 3.12 & 4.3 for 3, 5, 7 and 10% of PP70/MA content respectively. Also, in case of using rubber materials the increase in P.I will increase the resistance of asphalt samples to cracking at low temperature.

The penetration index of all modified asphalt samples from Table 3 the PI proved a rather powerful indicator for classifying asphalt rheological behavior, and it was observed that blown asphalt had typically  $PI < 1$  when straight-run asphalt had  $-1 > PI > +1$ . Only very susceptible materials such as coal tar gave  $PI > -1$ . Therefore, PI was thought to be a good indicator of asphalt type, with  $PI < 2$  being indicative of a gel asphalt whereas  $PI > 0$  is typical of a sol. In general, PI varies between -2.6 and 8 when including all types of asphalt (including blown ones) but typically lies between -2 and +2 for paving grades asphalt. Solvent deasphalted asphalt could give PI values between -2 and +2, based on the exact process used (especially type of solvent and yield. Still, negative values were more frequent, and deasphalted asphalt is generally considered more susceptible than straight run ones. Van der Poel showed that asphalt with the same PI had similar rheological master curves. Since a mathematical model turned out to be quite difficult to propose, Van der Poel developed a nomograph from which the complex (or creep) modulus at whatever temperature and frequency (or time) can be predicted for given asphalt, knowing only its PI and Ring and ball softening temperature. This device still gives acceptable values.

Hardness and stability of modified samples are obviously noticed for PP70/MA addition rather than in case of using PP70.

Table 3. Physical properties of PMAs using PP70 and PP70/MA

Asphalt sample	Penetration 0.01 mm 25 °C	Ductility cm	Kinematic viscosity 135 °C, C.st	Absolute viscosity 60 °C	Softening point °C	Specific gravity gm/cm <sup>3</sup>	(1) API	2* PTS	3* PI	*4 C, Btu pound per °F	*5 thermal conduc- tivity	Dynamic viscosity mpa.s, 60 °C 1198.08 S <sup>-1</sup>
3	44	129	375	1645	56	1.023	-8.563	0.0361	-0.21	0.385	0.6530	1176.54
5	28	120	435	1732	64	1.0910	-7.713	0.0281	0.38	0.377	0.667	1560.69
7	23	80	450	1820	68	1.1431	-3.008	0.0247	0.64	0.2320	0.834	2201.12
10	17	50	495	1921	73	1.1510	-1.254	0.0201	0.85	0.2291	1.021	3301.56
With MA												
3	42	125	450	1720	56	1.024	-6.415	0.0344	-0.21	0.360	0.748	1350.23
5	27	115	503	1850	72	1.028	-2.568	0.0305	2.38	0.342	0.862	1820.36
7	17	75	560	1930	80	1.030	-0.652	0.0250	3.12	0.202	1.025	2830.56
10	7	30	630	2140	88	1.032	1.542	0.0184	4.3	0.160	1.458	4238.14

\*2 - penetration temperature susceptibility; \*3 - penetration index; \*4 - Specific heat; \*5 - Thermal conductivity

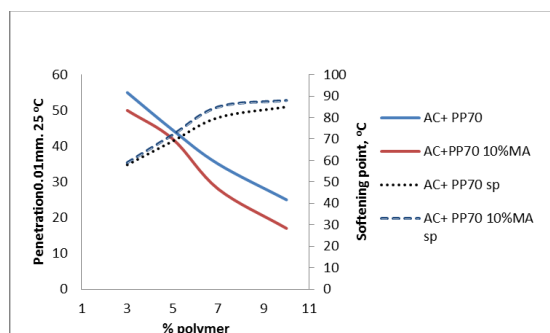


Fig. 2. Penetration and softening point of PMAs with percentages of blends addition

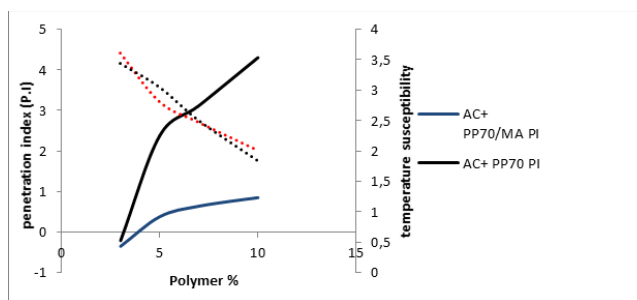


Fig. 3. Penetration index and temperature susceptibility of PMAs with percentages of blends addition

### 3.2.3. Rheological properties

#### 3.2.3.1. Viscosity and temperature

In this test, the effect of temperature variation as sever conditions for which the coating may be exposure on the viscosity for all samples is detected. The test is also used to develop temperature-viscosity charts. As shown in Figures 4,5 it can be observed that the viscosity values reduce as the test temperature increases regardless of the polymer type and content. The blend viscosity increased with the increase in polymer content at the chosen temperatures from 60 to 150°C, and this increase was observed in both the viscous and elastic moduli. As shown in Figures 4,5 the viscosity of PMAs samples are highly remarkable decrease as compared to the unmodified sample. This is due to the presence of PP/ NBR particle which causes improvement in rheological properties of virgin asphalt. dynamic viscosity decreases in case of using PP70/MA & PP70 at temperature 60 °C in percentages of 3, 5, 7 and 10% wt/wt in percentages of 100, 200, 225, 250% & 300, 350, 360 and 600% respectively while at 130°C dynamic viscosity decreases in percentages 47, 50, 52 & 53 and 20, 30, 43 & 45 respectively. As expected, there is an increase in the asphalt viscosity with the polymer addition, and it is important to stress that samples with compatibilizer agents, which have a plasticizer effect, showed higher viscosities than the pure asphalt [46].

The high differences between the two modifiers is due to presence of MA-PP as a compatibilizer in PP70/MA which interact with asphalt to form very strong network causes the asphalt to become highly viscous.

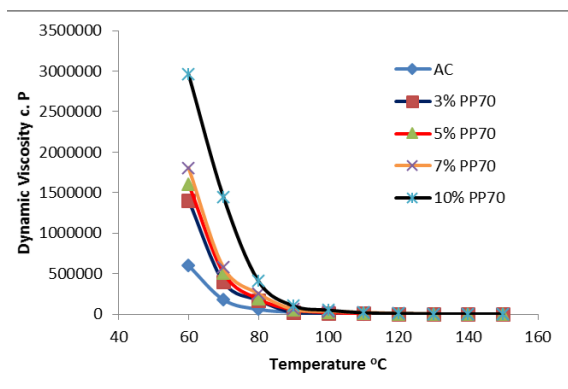


Fig. 4. Dynamic viscosity with temperature of PMA using PP70

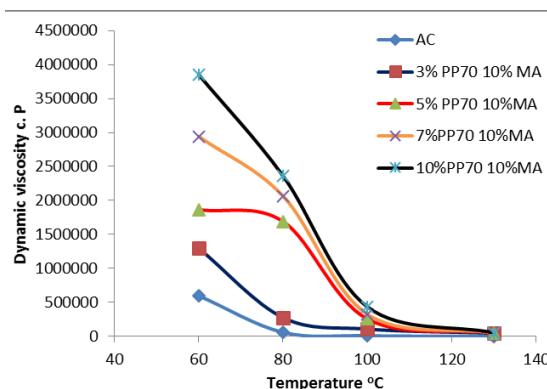


Fig. 5. Dynamic viscosity with temperature of PMA using PP70/MA

### 3.2.3.2. Shear rate and shear stress

Figures 6-9 show that the virgin asphalt and PMAs samples do not undergo stress rates proportional to the applied shear stress that's means that all samples are undergo non-Newtonian matters.

The viscosity of such fluids will therefore change as the shear rate is varied. Thus, the experimental parameters of Viscometer model, spindle and speed all have an effect on the measured viscosity of the non-Newtonian fluid. Non-Newtonian flow can be envisioned by thinking of any fluid as a mixture of molecules with different shapes and sizes. As they pass by each other, as happens during flow, their size, shape, and cohesiveness will determine how much force is required to move them. At each specific rate of shear, the alignment may be different and more or less force may be required to maintain motion. There are several types of non-Newtonian flow behavior, characterized by the way a fluid's viscosity changes in response to variations in shear rate. As shown in Figures 6-9 it is noticed that the elasticity modulus of PMAs increase with increasing the polymer addition level in the order of 3, 5, 7 and 10% wt/wt for both modifiers namely PP70 & PP70/MA but in case of using PP70/MA, the elasticity modulus is remarkable highly increased. This attributed to that the increased dipolar interaction between the MA-PP and NBR phase, which causes an increase in interfacial adhesion between PP and NBR phases. This bond should prevent phase separation and improve storage stability [38].

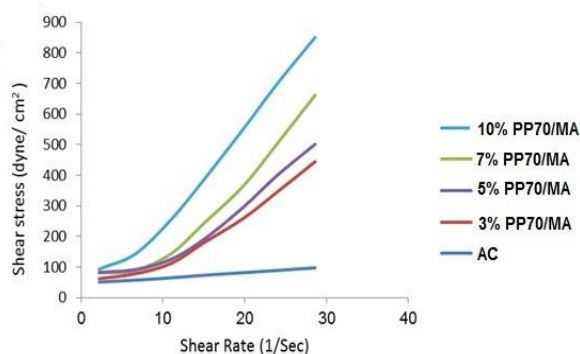


Fig.6. Shear stress shear rate curves of PMA using PP70/MA at 130°C

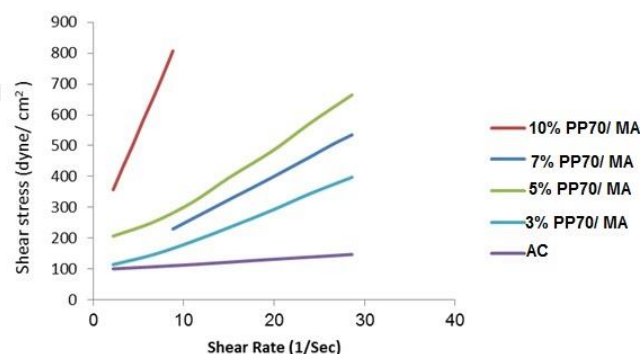


Fig. 7. Shear stress shear rate curves of PMA using PP70/MA at 80°C

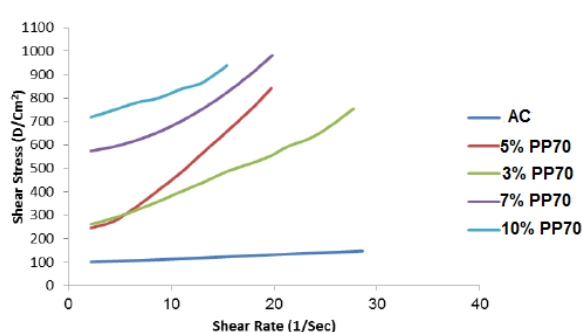


Fig. 8. Shear stress shear rate curves of PMA using PP70 at 60°C

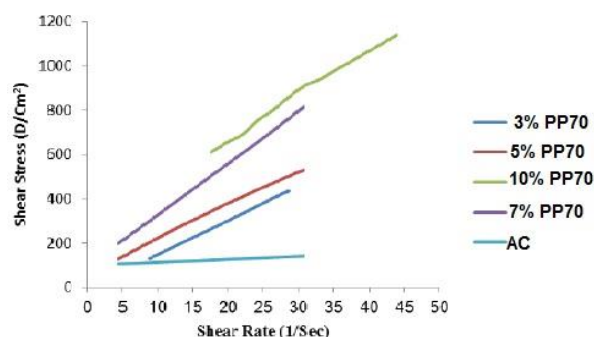


Fig. 9. Shear stress shear rate curves of PMA using PP70 at 130°C

### 3.2.4. Tensile shear strength and elongation measurements for modified asphalt samples using PP70 and PP70/MA

The purpose of tensile strength testing is to evaluate asphalt mixtures fatigue potential and moisture susceptibility. Chapuis *et al.* [47] has indicated the tensile strength of hot-mix asphalt is related to fatigue cracking.

A higher tensile strength means asphalt can tolerate higher strains before failing (i.e., cracking). Additionally, the moisture susceptibility of the asphalt mixture can be evaluated by comparing the tensile strength of asphalt mixtures exposed to wet and dry conditions.

From Figs 10-13 and Table 4, the data revealed that the tensile shear strength values measured for different samples in air and water give interesting behavior. The tensile shear strength for all samples is slowly increasing with increasing the percentages of polymer added. It is noticed from the results that the PMAs with using PP70/MA acquire better tensile strength as compared with those based on PP70 without MA. Comparison of PMAs using PP70/MA in the air and in water shows that the order of increase in tensile shear strength follows the sequence 10% polymer > 7% > 5% > 3%. The tensile strength ratio (TSR) [48] for samples conditioned with water is shown in Figures 10-13 and Table 4. The value of TSR is calculated as: Tensile strength ratio = Tensile strength of conditioned sample/Tensile strength of the dry sample.

In Figures 10-13 and Table 4 TSR testing would have a value of less than 1.00 in case of blended PP/NBR with and without MA because it is expected that the conditioned samples would suffer moisture damage and exhibit lower tensile strength with comparing to PMAs; this phenomenon was observed in the control sample. However, it was found that all polymer modified asphalt samples exhibited TSR values greater than 1.00 [49]. This indicated that the sample after conditioning has higher tensile strength. Overall, it was concluded that the addition of TEPs (PP70& PP70/MA) would reduce a mixture's moisture susceptibility in most cases.

The different ratios added from PP70 & PP70/MA were found to have the potential to improve material strength and fatigue characteristics while enhancing ductility and other durability properties of engineering materials in different media.

Table 4. Tensile shear strength, (N/cm<sup>3</sup>) and elongation (El., %) for PMAs with PP70 & PP70/10% MA after 24 hrs immersion in different media, at room temperature

Sample	In air		In H <sub>2</sub> O	
	N/cm <sup>3</sup>	El, %	N/cm <sup>3</sup>	El, %
PP70	825	40.4	800	80
PP70 /MA	1370	41.7	1165	100
3% PP70	420	60	523	140
5% PP70	523	80	623	120
7% PP70	795	100	795	100
10% PP70	823	120	1180	80
3% PP70/MA	623	80	573	160
5% PP70/MA	707	100	1180	140
7% PP70/MA	975	120	1225	120
10% PP70/MA	1115	140	1295	100



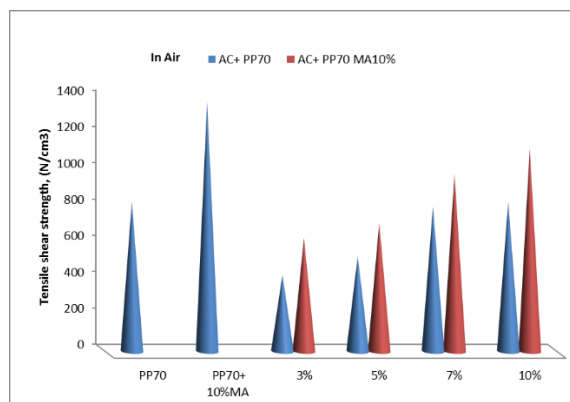


Fig. 10. Tensile shear strength of PMAs with % blends added in air

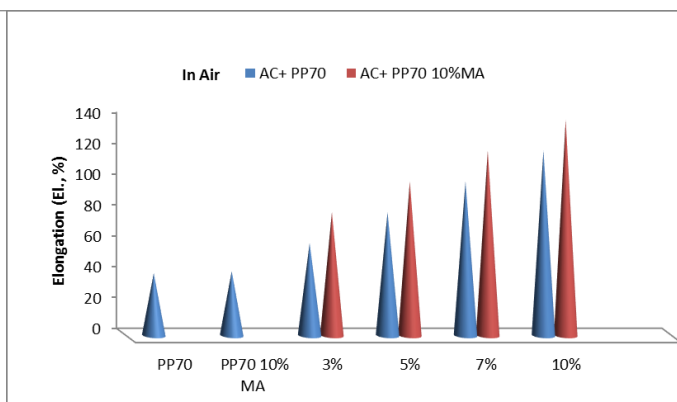


Fig. 11. Elongation of PMAs with % blends added in air

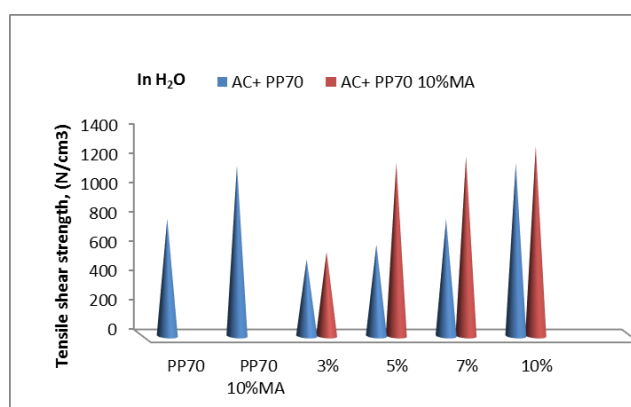


Fig. 12. Tensile shear strength of PMAs with % blends added in water

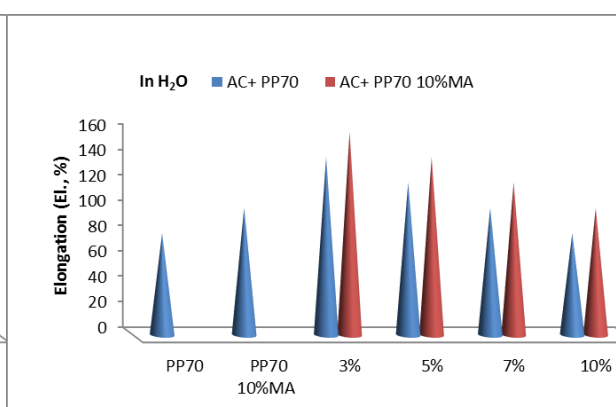


Fig. 13. Elongation of PMAs with % blends added in water

#### 4. Conclusion

This research aims to use Thermoplastic elastomers (TPEs) which are a class of copolymers or a physical mixture of polymers (usually a plastic and a rubber) which consist of materials with both thermoplastic and elastomeric properties in the modification of asphalt 60/70 to produce very specific asphaltic material for use in infrastructure applications in different media. TPEs have attracts more attentions in the industry since they combine the properties of vulcanized rubber with the ease of processing of thermoplastics. The new asphaltic material is low cost and has a high quality performance at low temperature. To achieve the aim of the study, blend PP/ NBR with and without compatibilizers (MA) were used as additives to modify asphalt 60/70 in percentages of 3, 5, 7 & 10 wt/wt% The results of the study concluded that;

- The compatibility of PP and NBR is poor and can be enhanced by the addition of compatibilizers.
- In this study, PP/NBR (70/30 wt/wt%) blends were modified by the addition of compatibilizers, that is, MAH. Ternary blends with compatibilizers (10 wt/wt%) showed an improvement in mechanical and rheological properties compared with uncompatibilized ones
- As expected the addition of polymers to asphalt reduces its thermal susceptibility at temperatures and guarantees a greater constancy of its properties since the asphalt becomes more elastic.
- Increasing the ratio of polymer added found to improve the physical and rheological properties, and the suitable ratio is 7% according to workability
- Rheological data PMAs indicates that the PMAs have thixotropic property and high yield stress

- The phenomenon of low viscosity at a high shear rate, and lack of flow at low shear rate which is gained by prepared blends is very important from the practical point of view since the application requirements for sealants necessitate both good gunnability and lack of flow on vertical surfaces.
- The tensile shear strength ( $\text{N/cm}^3$ ) for the obtained blends is found to increase with polymer % content and for that forming with using PP70/MA more than that with using PP70.
- Addition of TEPs (PP70 & PP70/MA) enhancing the mechanical properties of virgin asphalt 60/70 in both water and air but in the air is the best.
- Polymer modified asphalt samples have a higher tensile strength means asphalt can tolerate higher strains before failing (i.e., cracking). Additionally, the moisture susceptibility of the asphalt mixture can be evaluated by comparing the tensile strength of asphalt mixtures exposed to wet and dry conditions.

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*To whom correspondence should be addressed: Dr. A. A. Ragab, Petroleum Applications Department, Egyptian Petroleum Research Institute, Nasr City, Cairo, Egypt, [chemragab83@yahoo.com](mailto:chemragab83@yahoo.com)*