



## Mixed Polymeric Wastes as a Promise Path to Green Roads

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## Abstract

In this study, asphalt binder was loaded with 5, 10, 15 and 20 wt.% of grinded mixed waste of plastics. The polymeric waste comprised mainly from a mixture of PE/EPDM (60/40 wt.%). To study the effect of these wastes on asphalt binder performance; the physical properties of asphalt binder were studied including penetration, viscosity, and softening point. The mechanical characteristics of modified asphalt binder were studied using Dynamic Mechanical Analyzer (DMA). Two types of sweep tests were conducted on virgin and modified asphalt binder to study the effects of temperature and frequency on binder performance. Elastic recovery of modified asphalt binder was also investigated using the stress relaxation mode in DMA. In addition, the fatigue properties of modified asphalt samples were estimated from the obtained data of loss modulus and rutting factor ( $G^*/\sin\delta$ ). It is observed that mixed plastic waste positively enhances the performance of asphalt binder.

*Keywords:* Mixed plastic wastes, Asphalt binder, Viscosity, Dynamic properties, Elastic recovery, Fatigue.

## 1. Introduction

Huge quantities of polymeric waste materials are growing exponentially in industrial activities and contemporary society everywhere. Accumulations around the globe are a consequence of the accumulation of industrial waste due to the steady increase in production and demand on products, and increase the volume of waste from developing business activities. Polymeric waste is growing with the growth of consumer communities in the world. These wastes are due to several reasons including the over-consumption of plastic materials used for packaging and a lot of plastic wastes and other waste materials. Therefore, recycling of polymer wastes allows for many benefits, including: preserving the natural wealth of raw materials and reduction of land allocated for dumps. In addition, recycling of these wastes will enhance the protection of the environment from ground water pollution, air pollutants emitted from incineration processes; soil pollution from toxic substances and non-biodegradable materials. Several research activities

cover a wide range of topics including biodegradable composites[1].

It is well known that most of polymer wastes do not undergo bio-decomposition and consequently it will pollute land and atmosphere if it is land filled or incinerated. However, the discovery of the binding property of polymers in its molten state in road laying helped to well manage these waste plastics [2]. Using of plastic wastes for bitumen modification was widely studied using a wide variety of polymer wastes including wastes of low density polyethylene (LDPE) [3], high density polyethylene (HDPE) [4], polypropylene (PP) [5], ethylene-vinyl acetate (EVA) [6,7], acrylonitrile-butadiene-styrene (ABS) [8], polyethylene terephthalate (PET) [9] and poly vinyl chloride (PVC) [10,11]. In addition, thermal oxidative aging of bitumen has been also investigated using specific antioxidants [12]. Most of the studied research proved that each waste type affected on a certain characteristic of asphalt binder performance; either if it has been added as filler or as a binding agent. Modifying asphalt binder with styrene butadiene rubber (SBR) improves its low temperature

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performance (resistance to cracking), elastic recovery, and ductility of binder [13].

In this study, mixed polymeric wastes including (PE and EPDM rubber) were wholly grinded and mixed with asphalt binder, to get the maximum benefit of the binding property of these plastics waste on asphalt binder performance. The physical properties of treated asphalt binder were studied by performing penetration test, viscosity measurement, and softening point determination. In addition, the mechanical characteristics including rutting factor  $G^*/\sin\delta$ , phase angel, elastic recovery, and fatigue properties were investigated using dynamic mechanical analyzer (DMA).

## 2. Experimental Methods

### 2.1. Materials

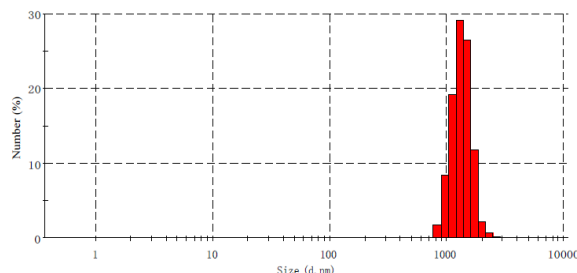
Asphalt binder (Bitumen 60/70 paving asphalt) was obtained from Suez Company, Egypt. Mixed plastic wastes comprised from a mixture of (PET, PE, PP, and EPDM rubber) were collected from accumulation points of industrial wastes in Egypt and were manually segregated [14]. Polymer wastes used in this work were selected based on our previous published work [15,16]. Recycled LDPE in crushed form was obtained from the Egyptian Saudi Company for Plastic Products; Industrial Zone, 10th of Ramadan, Egypt. The rubber material was obtained from the same company in the shape of grinded particles with maximum particle size of 0.6 mm. Based on the characteristic certificate of the company, the density of LDPE is 0.922 gm/cc and its melt flow index is 6 g/10min at (190°C, 2.16 kg). The EPDM rubber has 72 wt.% ethylene and 4.8 wt.% ethylidene norbornene contents, respectively, with a Mooney viscosity of ML(1+4)<sub>125</sub> oC=59.

### 2.2. Methods

#### 2.2.1. Asphalt Modification

Firstly, each plastic waste was oven dried for 2 hours at 80°C and was grinded afterwards. Each polymeric waste was cryogenically grinded by immersing in liquid nitrogen followed by crushing of the frozen brittle material through impact force utilizing a hammer mill [17]. The particle size and its distribution for each of the grinded waste were automatically determined using dynamic light scattering (DLS) [18] as shown in **Fig1**. In this paper, a mixture of plastic waste composed of PE/EPDM (60/40 wt. %) was used in the modification of asphalt binder. This specific mixture was found to be the optimum percentage for enhanced mechanical properties of its TPO composite

[19]. Therefore, this optimum mixture was used through all samples of asphalt binder modification.



**Figure (1) Statistics graph of particle size distribution of grinded mixed wastes**

For the fabrication of the modified binders, asphalt 60/70 was heated to 150°C while mechanically stirred at a speed of 2500 rpm, and then the grinded plastic waste was slowly added in small doses until reaching the specified amounts namely 5%, 10%, 15%, and 20% by weight of the total asphalt binder. The waste addition was performed in such a way that each dose was well mixed for 1 min before adding the next dose. The mixing time was conducted for two hours using a hot surface to ensure a constant temperature of the binder during the process.

#### 2.2.2. Testing procedure

##### - Physical Tests

The physical properties of virgin and modified asphalt binder were performed, including penetration test at 25°C according to ASTM D5 [20], softening point (Ring and Ball) according to ASTM D36 [21] and dynamic viscosity (poise) at 135°C according to ASTM D-4402 [22]. The penetration index (P.I.) values were calculated from penetration values and softening point temperatures [23].

##### - Dynamic Shear Rheometer test

The dynamic shear rheometer test was performed by Dynamic Mechanical Analyzer (DMA) using Triton Technology-TTDM, following ASTM D-7175[24]. The asphalt binder was heated up to 150°C for two hours, poured into silicon rubber molds to prepare 8-mm diameter specimens, and allowed to cool down for about an hour. Details of sample preparation and testing for dynamic shear analysis are given by A.M.M, Abd El-Rahman et al. [25]. Specimens were loaded at temperature of 6°C below the testing temperature, and they were pre-shared to minimize any historical load associated with sample preparation, loading and handling. The following sub-sections summarize the selected sweep tests (temperature and frequency) conducted on the asphalt binder as illustrated in **Table 1**.

**Table (1):** Test Matrix for DMA-based Sweep Tests performed on asphalt binder.

Sweep type	Testing Temperature (oC)	No of Tests	Strain (%)	Frequency Hz
Temperature	25 to 70	3	12 %	10
Frequency	60	3	12 %	0.0 to 10

**- Temperature Sweep test**

Specimens were loaded and trimmed at 25oC, followed by a 30-second pre-shearing at a strain of 12% and a frequency of 10 Hz. The testing temperature ranged from 25oC to 70oC, and the data was collected at 3oC interval. Time equilibrium of 3 min was maintained at each data point. The rutting factor  $G^*/\sin\delta$ , loss modulus (fatigue property), and phase angle (tan delta) for each data point was plotted against the testing temperature range.

**- Frequency Sweep test**

Specimens were loaded at 58oC, and were preconditioned at a frequency of 0.01 Hz by maintaining a constant strain of 12% at a temperature of 60oC. Following 5-min temperature equilibrium, frequency sweep tests were conducted at a frequency ranging from 0.015 Hz to 10 Hz, with a confining strain of 12% and a constant temperature of 60oC.

**- Elastic Recovery by the Dynamic Shear Rheometer (ER-DSR)**

The elastic recovery (ER) in the DSR testing for asphalt binder and mastics has been performed according to Clopotel et al.[26]. At the end of the relaxation step, the elastic recovery was calculated as follows:

$$\text{ER-DSR} = \frac{\epsilon_2}{\epsilon_1} \times 100 \quad \text{Eq. (1)}$$

Where:

$\epsilon_2$  is the recovered strain at the end of relaxation step  
 $\epsilon_1$  is the strain at the end of loading step

**3. Results and discussion****3.1. Physical properties**

Physical characteristics of the modified asphalt binder are illustrated in **Table 2**. Four specimens of modified asphalt binder with 5, 10, 15 and 20 wt.% of plastic waste were presented for comparison with neat asphalt binder. The penetration index (PI) is used to classify bitumen. It is well known that PI values can be used to determine the stiffness of bitumen at any temperature and loading time. It may also be used to identify a particular type of bituminous materials in a limited extent. Typical

values of PI are shown in **Table 3** [27]. It is clearly seen that PI values increased from 2.478 to 13.57 with increasing the plastic waste content from 5 wt.% to 15 wt.% in asphalt binder formulations. As it is observed in **Table 2**, the PI value of the specimen of 15wt.% modified asphalt binder attains 12.5 which is five times larger than that of the neat asphalt binder.

**Table (2):** physical properties of virgin and modified bitumen

Physical Characteristics	Blank	Asphalt with (% waste)			
		5 %	10 %	15 %	20 %
Penetration (@ 25C°, 100g,5s) 0.1mm	62	40	27	16	8
Softening point (ring and ball) C°	54	70	72	74	86
Dynamic viscosity (cp)	115	235	320	540	890
Specific gravity (@ 25 C°) (pycnometer)	1.05	1.05	1.05	1.05	1.05
Penetration Index (P.I)	2.47	13.57	11.14	12.46	9.517

**Table (3):** Typical values of penetration index (PI).

Bitumen type	Penetration index
Blown bitumen	> 1
Conventional paving bitumen	-1 to 1
Temperature susceptible bitumen (tars)	< -1

By comparing the physical test results of neat and modified asphalt binder, it is noticed that the added plastic wastes decreased the penetration values and increased both the softening temperature and the viscosity of asphalt binder. This could be attributed to the strong interaction between the entangled polymeric chains of waste networks which restricts the relative movement of asphalt molecule, increases the capacity to resist external forces, and also increases the viscosity of the modified asphalt [28]. This behavior stems from the very low temperature susceptibility of the added plastic wastes. In turn, this results in a higher resistance against thermal cracking of the pavement at low temperatures. In addition, these results indicate that these asphalt binder/plastic wastes formulations will also withstand against high temperatures plastic deformation which appears on the road surface under high traffic loadings [29].

**Dynamic characteristics of asphalt binder**

Characterizing the rheological properties of asphalt binders is highly important for prediction of major pavement damages such as rutting, raveling and stripping. A laboratory study was conducted to examine the viscoelastic properties of modified asphalt binder using a Dynamic Mechanical Analyzer (DMA). Two types of sweep tests were conducted on

all asphalt binder samples to examine the effects of temperature and frequency on both neat and modified asphalt binder.

### -Temperature Sweep

The temperature sweep test results showed that the rutting factor  $G^*/\sin(\delta)$  decreases with temperature as shown in Fig. 2 and the phase angle ( $\tan \delta$ ) increase as shown in Fig. 3 with an increase in temperature, which is expected [30]. At a low temperature, the asphaltenes in asphalt binder are able to form a compact structure, whereas at high end of the testing temperature they are dispersed as free particles. Thus, asphalt binder becomes a liquid like material at high temperature, and the time lag (loss tangent) between applied stress and resultant strain increases with an increase in temperature.

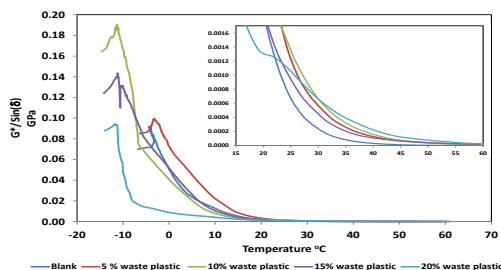


Figure (2) Rutting factor  $G^*/\sin(\delta)$  against temperature

From Fig. 2, it is also observed that these modified asphalt binders satisfy the Superpave® acceptance criterion since  $[G^*/\sin(\delta) \geq 1 \text{ KPa}]$  at any temperature up to 60°C. Also these results proved that adding mixed plastic wastes as modifiers improve the rutting factor [i.e.  $(G^*/\sin(\delta))$ ], since it is increased with increasing the content of waste. In addition, the value of  $\tan \delta$  decreases with increasing the percentage of waste mixtures which reveal that modified asphalt binders satisfy the high grade temperature for un-aged condition.

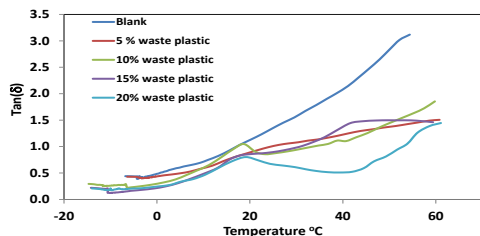


Figure (3) phase angle of virgin and modified asphalt samples

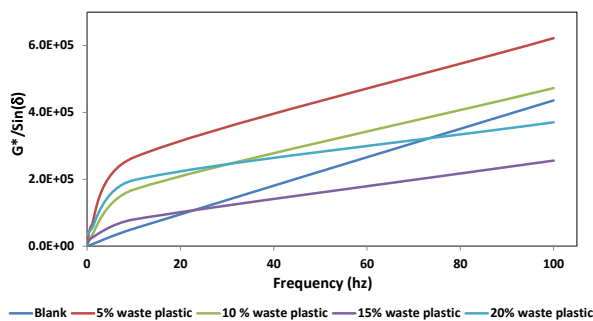


Figure (4) Rutting factor of base and modified asphalt binder with frequency at 60°C

### -Frequency sweep

As shown in Fig.4, the rutting factor is significantly lower for lower loading frequency (slow moving vehicle) for all asphalt samples but still satisfies the Superpave® rutting factor criterion  $[G^*/\sin(\delta) \geq 1 \text{ KPa}]$  for a frequency of as low as 0.1 Hz (vehicle speed from 0.5 mph to 0.58 mph) i.e. the vehicles are almost stopped. Therefore, these modified binders will be recommended at pavement sections that experience frequent congestion or at the intersection of major highways where the traffic speed is bound to be lowered.

### -Elastic Recovery

The elastic recovery of base and modified asphalt samples is evaluated using DMA at thermal equilibrium 25°C, and constant strain rate 0.023/sec is applied for 2 minutes under strain controlled mode. Then a constant zero shear stress is applied for a period of 1 hour under stress controlled mode. The elastic recovery % is calculated from equation 1. Then, a curve was plotted as shown in Fig. 5. It is noticed that addition of 5, 10, 15 and 20 (wt. %) mixed wastes of plastic increase the value of recovered strain  $\epsilon_2$  at the end of relaxation and consequently increase the elastic recovery % compared with base asphalt. This recovery is due to the addition effect of elastic properties of these wastes.

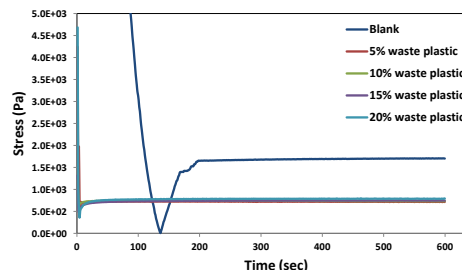


Figure (5) Stress relaxation of virgin and modified samples

### - Fatigue parameter

Fatigue cracking is one of the most serious distresses for asphalt pavement. Generally, the initiation and propagation of cracking are always related to the magnitude of dissipated energy produced by outer loading. The loss modulus ( $E^* \sin \delta$ ) could be an effective parameter to characterize the resistance to fatigue cracking of asphalt mixture as ( $G^* \sin \delta$ ) is used for asphalt binders [31]. The high value of ( $G^* \sin \delta$ ) is correlated to the poor resistance to fatigue cracking of asphalt mixture. Therefore, in this study ( $G^* \sin \delta$ ) was defined as a fatigue parameter to evaluate the fatigue resistance for modified asphalt binder. The fatigue parameters for all asphalt samples were compared at 60°C with frequency 10 Hz as shown in Fig. 6. It could be seen clearly that the fatigue parameters of the modified asphalt binder were reduced significantly when plastic wastes were added. Consequently, it could be estimated that the dissipated energy produced from cyclic loading can be reduced for modified asphalt binder, where the waste distracts the stress produced in asphalt binder and prevents the initiation and propagation of cracks.

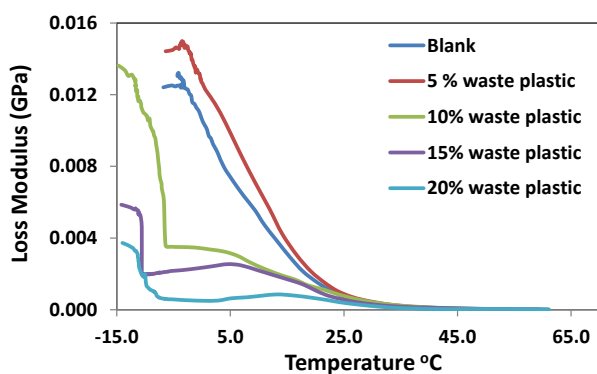
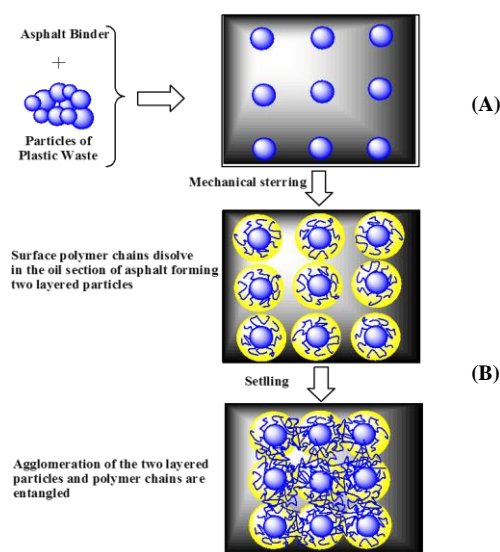


Figure (6) Loss Modulus for virgin and modified asphalt samples

As can be seen in Fig. 6, the fatigue parameter for control mixture at 10 Hz is 1526 [KPa], and for four modified binders are 1487 [KPa], 1146 [KPa], 1033 [KPa] and 549 [KPa] for 5%, 10%, 15% and 20 wt% of plastic waste, respectively.

Zhu et al (2014) discussed some academic debates about the mechanism of polymer dispersion and its modification enhancement on asphalt binder [32]. However, based on the significant enhancement of the mechanical properties of the modified asphalt binder shown here in this work, the overall mechanism of waste particles dispersion could be clearly explained as follows. Asphalt material consists mainly of three components, oils, resins and asphaltenes. When the grinded waste plastics are mixed with asphalt and under the action of mechanical force, the polymeric particles absorb the oil content in asphalt and get swollen. As a result, the

particles of swollen polymer become well dispersed and evenly distributed in asphalt matrix, and the base asphalt becomes thicker and sticky and consequently, the sizes of the swallowed polymer particles are enlarged. This could be attributed to the diminishing of the oil content which has been absorbed. After stopping the stirrer, the polymer chains at the surface of the swallowed polymer particles are entangled forming a network. After the oil content has been absorbed by the polymer network, the number of macromolecule in asphalt relatively increases. Moreover, polymer disperses in asphalt in the form of particles and get close to each other due to aggregation action, forming a network interlock structure as shown in Scheme 1.



Scheme 1: Dispersion of polymeric waste particles in asphalt binder

Consequently, the strong interaction between networks restricts the relative movement of asphalt molecule, increases the capacity to resist external forces, and also increase the viscosity of the modified asphalt [33]. As a result, these waste particles improved bitumen properties including enhancement of the percent of elastic recovery, significant lowering in the rutting factor, and longer fatigue life; which are consistent with published research articles in polymer modification of bitumen [34], and [35].

### Conclusion

In this study, the physical, and mechanical properties of virgin and modified asphalt binder with mixed polymeric wastes comprised from a mixture of PE/EPDM (60/40 wt%) by four doses (5%, 10%, 15%, and 20wt%) were studied.

The main findings of this work are:

- Modified asphalt binder showed an increase in softening point and viscosity, decrease in penetration, and increasing the penetration index (PI) of binder which is considered as a good indicator for a higher resistance against thermal cracking of the pavement at low temperatures, and less permanent (plastic) deformation at high temperatures on the road surface under traffic loadings. DMA is an efficient tool to study the dynamic mechanical characteristics of modified asphalt binders subjected to temperature, and loading frequency sweep.
- From temperature sweep results, it is concluded that modified asphalt binders satisfy the Superpave® acceptance criterion since  $[G^*/\sin(\delta) \geq 1 \text{ KPa}]$  at any temperature up to 60°C. Also the results revealed that adding plastic waste as modifier to asphalt binder improves the rutting factor.
- From frequency scan results, we conclude that modified asphalt binders could be recommended at pavement sections that experience highly frequent congestion or at the intersection of major highways where the traffic speed is bound to be low.
- The parameter of  $G^*\sin\delta$  (loss modulus) is comprehensively studied to predict the fatigue properties of asphalt binder performance. The results assured that mixed waste of plastics have clear impact in improving the fatigue resistance of asphalt binder.

Overall, these wastes will produce asphalt binder with better properties than base one, and can be provided as an option for the partial replacement of the non-renewable asphalt binders used in the construction of road infrastructure. Moreover, using the plastic wastes in pavement is considered as an economical resource and a promise path for green roads.

#### Conflicts of interest

There are no conflicts to declare.

#### Acknowledgements

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