Improved asphalt binder using recycle polyethylene terephthalate polymer

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Abstract
Roads exposure to a range of natural factors, like the difference between the temperatures between the seasons between the high temperature and drought to the sharp decline in winter temperature and the impact of water and high humidity in addition to the mechanical factors, which is high load pressure and increased traffic. In asphalt binder such as cracking and crawling and the cracks of all types due to fatigue and aging due to the loss of asphalt material Association of some of its important components as the matter of bitumen is the main material bonding components of paving asphalt and therefore emerged the need to improve their specifications through additives, in this study the recycle polyethylene terephthalate of drinking water bottles by a percentage of (5%, 10%, 15%) to improve the properties of mechanical and rheological properties of asphalt and overcome pavement problems as well as the disposal of excess polymer waste and environmental pollution. Also, to the improper treatment of incineration due to the release of harmful gases such as carbon dioxide and environment-polluting chemicals. Bitumen tests such as sutures, softening point and penetration test, linear viscosity (K.V) , rotational viscosity (R V) and (FTIR) spectra were studied. The result showed that of modified asphalt binder better than the unmodified.

Introduction
Bitumen is a hydrocarbon organic chemical of various proportions which is a major component of petroleum and is produced from refinement oil as byproducts [1].The chemical structure of Bitumen is shown as liquid at high temperatures with low homogeneity and at low temperatures becomes very brittle [2]. The bitumen structure includes ratios of elements composed mainly of carbon and hydrogen with lower ratios of nitrogen and sulfur and many minerals such as nickel, vanadium, iron and magnesium in the form of oxides and inorganic salts [3].The heat of the weather has an important effect in the paving asphalt cement, especially when the rise and fall of the temperature, which leads to the destruction of roads and reduce the life time and make the maintenance higher cost [4].One of the most important defects of asphalt roads is the rapid deterioration of the pavement structure due to the process of permanent deformation as it reduces the time of operation of the road as well as the convenience and safety of the user of the road [5].Asphalt cement is characterized by high temperature and low temperature cracking properties. Moreover, the most common deformities are drilling, trenching, rippling and surface cracks [1].Asphalt paving process
depends on the type of asphalt used and the amount of containment of the heavy materials as well as the ratios of the materials and the composition of it, so one of the solutions is the process of improving the final asphalt link by adding hosts with desirable characteristics that improve the resistance to permanent deformation due to high load and increasing traffic and especially in hot climates and high temperatures [6-8]. Polymers as additives use to achieve better resistance to water, toughness. Fatigue resistance, noise retention and cracking resistance in cold areas; High viscosity and high softening point increase the bond strength of modified asphalt components compared to conventional asphalt [9]. On the other hand, the development of modern life and the diversity of living methods led to the presence of large quantities of plastic waste, which is not biodegradable, causing harm to the environment for a long period of time. Therefore, there was a need for a method of waste disposal in an innovative and sustainable way to converted into useful products [10].

The use of polymer additives in asphalt improves performance by resisting fatigue and enhancing the rheological properties at high and low temperatures and reducing the breakage and thermal breakage [11]. Different types of additive-polymer have been used to improve asphalt specifications in order to increase fatigue resistance. Polymers and fibers are considered the best additives in improving asphalt properties [12].

Two ways were used to adding additives to the asphalt mixture. The wet method is to add additive to bitumen before adding other asphalt mixtures. This process requires a high cut mixer to obtain a homogeneous mixture [13]; While the second (dry method) additive adding to the bitumen with the ingredients of the mixture and one other batch during the preparation of hot asphalt [14]. In a dry way, a greater proportion of plastics can be used in the asphalt mixture by replacing part of the metal aggregates with an equal size of plastic waste polymers [11]. However, there is a weak compatibility between polymers and asphalt documented due to the different structure of polymer and the structure of Asphalt, which causes the separation of phase, especially at high temperatures, which is one of the most common problems in improving the performance of asphalt polymer, called this problem high temperature storage stability [15].

Polymers are classified into thermoplastic and thermosetting polymersto determine their mechanical properties. Polyethylene terephthalate (PET) is a type of thermoplastic polymers that allow for re-molding and formation for many times due to the possibility of reversing the resulting structure heating [16]. (PET) Is a thermoplastic polymer from the family of polyesters manufactured and is in the normal state of polymer solid, strong, semi-crystalline, colorless, and these characteristics depend on the method of preparation is lightweight [17]. There are three ways to recycle (PET) and the best mechanical method includes a range of processes such as separation, washing, drying, milling and processing, as well as entering into other products such as polystyrene fiber and containers and covers and carpets due to the thermal, optical, tensile, and tensile stresses of the polymer [18]. In this work, the recycle (PET) was used to improving the performance of asphalt will help in the environmental solutions of solid waste through a series of simple operations with formation of new materials with more durable specifications [19].
Experimental

A. Materials

I. Plastic

Polyethylene Terephthalate produced from drinking water bottles produced by Al Waha Mineral Water Plant - Babil - Iraq. Table (1) shows the physical properties of the Polyethylene Terephthalate.

Table 1: Physical properties of the polymer

| Properties          | Methods                          | Class requirements | Values     |
|---------------------|----------------------------------|--------------------|------------|
| Density             | 1.37 g/cm³                       |                    |            |
| Thermal conductivity| 0.15 – 0.24 W/m.K                |                    |            |
| Melting point       | 254 °C                           |                    |            |

A quantity of water bottles were collected after use, washed and dried. It was indirectly treated thermally and then cut into small pieces and then grind by a mill machine. The analysis of the sieve revealed that %38.37 of the PET passed from a (0.300 mm) and %31.3 from (0.600mm) and % 22.5 through (0.425mm) (Figure 1).

II. Asphalt Binder (Bitumen)

The binder was penetration grade (40-50), which is often used in pavement (surface layer) in Iraq; Table 2 show the physical properties of the asphalt binder.

Table 2: Physical properties of the asphalt binder

| Properties                        | Methods                      | Class requirements | Values     |
|-----------------------------------|------------------------------|--------------------|------------|
| Penetration (25 °C, 100 g, 5 min )| Iraqi Standard No. 9         | 40 - 50            | 46 (0.1 mm)|
| Softening point (ring & ball)     | Iraqi Standard No. 1987/10    |                    | 44         |
| Ductility (25 °C, 5 cm/min)       | Iraqi Standard No. 160        | 100 <              | 120 cm     |
| Flash point (cleveland open – cup)| AASHTO T-48                  | 232 °C <           | 245 °C     |
| K. Viscosity (mm²/sec, 130 °C)    | AASHTO T 201                 |                    | 544        |

B. Methods

I. Modified Bitumen Preparation

There are many tests for asphalt binder as well as for asphalt mixtures to show the physical, mechanical and rheological properties of polymeric additives added to the asphalt binder and indicate improvement in these properties. In this paper the wet process was used. The bitumen (500 g) was heated in the oven until it became liquid and the polymer was added slowly. The mixer speed was maintained at (1500 rpm) and temperature between (165 °C and 175 °C). The concentration of (PET) ( %5, %10 , % 15 ) in weight from a mixture of asphalt
bonding continued for (60 minutes) to produce a homogeneous mixture. The polymer-modified bitumen (PMB) was then sealed in containers and stored for further testing.

C. Laboratory Testing

The K. Viscosity, the degree of Penetration, the softening point, and the ductility tests were carried out at Babylon's structural laboratory in Hilla and the rotational viscosity test was conducted in the soil laboratory at the Faculty of Engineering, Karbala University. Infrared spectroscopy at the college of Science, University of Babylon

1- Penetration (Iraqi Standard No. 9)

This method describes the way of assigning penetration to semi-solid and solid bituminous materials. This method is performed by smelting and cooling the sample under hard conditions. The penetration was measured using a standard Penetration device and needle. The amount of penetration is defined as the distance of ten millimeters that is penetrated vertically by a standard needle in a sample of the material under constant conditions of temperature. The standard test of the penetration grade was (100 g, 25 °C and 5.0 sec) on Penetrometer P734, on the bitumen base and PMB with polymer concentrations (5%, 10%, and 15%) of its weight of bitumen.

2- Softening Point: (Iraqi Standard No. 1987/10)

This method is to measure the bitumen tendency to flow at high temperatures when placed on roads; Also, method helps to control the production of bitumen in refineries and in the production of bitumen exposed to air, the ring and ball softening test is the standard test for determining bitumen strength, which is the standard temperature at which the phase transition occurs from solid to liquid. The steel ball weighs 3.55 grams and touches the base plate, which is 2.5 mm away.

3- Ductility test (Iraqi Standard No. 160)

The test is carried out to estimate the gravitational potential of the asphalt bond, which shows the evaluation of the asphalt containment of the adhesive material and thus the ability of the bitumen to adhere to the aggregate in the asphalt mixture.

4- Temperature Susceptibility:

The viscosity is directly proportional to the temperature rise. As the asphalt binder is characterized by thermal conductivity, the stability of the penetration of the asphalt must be checked at different temperatures (15, 25, 35 and 45) °C under the same conditions of pressure and time [20].

5- Rotational viscosity RV (ASTM D-4402)

The Rotational viscosity test was performed using a programmable apparent viscosity of the DV-III Ultra Programmable Brookfield, both for the non-modified asphalted binder and the polymer ratio in previous ratios. The rotational viscosity measurements were performed at different temperatures (135 °C to 175 °C) respectively.

6- K. Viscosity (AASHTO T-201)

The time required for the flow of a certain volume of liquid is measured during a precise course of a capillary viscosity measurement with a capillary tube, under precisely repeatable pressure and at a highly controlled temperature. The viscosity is then calculated by multiplying the flow time per second in the viscosity calibration coefficient. This method
covers the motor viscosity of bitumen, and road oils at 60 °C, as well as semi-steel asphalt at 135 °C.

**Results and discussion**

The degree of penetration decreases with the increase of the added percentage of (5%, 10%, and 15%). This is because the polymer material particles increase the strength of the bond between the asphalt particles, thus giving the overlapping asphalt higher toughness than the pure asphalt model which gives the permeability above to penetrate them as shown in figure 2.

![Figure 2: Penetration against %Polymer-Bitumen Ratio](image)

The bitumen gradually moves from the hardness state and becomes softening point and less viscous. The higher the temperature within a wide range of heat, where the higher the softening point is the bitumen sensitivity to heat. This experiment is useful in comparing the different asphalt types to classify the bitumen types. Figure (3) observes that the degree of elasticity increases by increasing the minutes of polyethylene terephthalate. The increase in the degree of elasticity is due to the order of molecules and their association with each other, especially when increasing the temperature to the occurrence of cohesion or lack of gaps between the chains linked to each other thus giving a better specification.

![Figure 3: Softening point against %Polymer-Bitumen Ratio](image)
The presence of a large aromatic ring in PET recycling units gives the polymer hardness and strength a noticeable form, especially when the polymer chains are identical with each other in the arrangement of the expansion regulator in the asphalt content. The semi-crystalline form makes it very resistant to deformation. Thus, the polymer-modified asphalt elongation increases with the addition of 10% of the additive and then decreases by 15% due to the high molecular weight of the sample, increasing the viscosity and reducing the thermal sensitivity of the asphalt bond (Figure 4).

![Figure 4: Ductility against %Polymer-Bitumen Ratio](image)

The improved asphalt bond is highly resistant to permanent deformation at low temperatures to reduce gradients, cracking and low hardness. The polymer has been added with certain ratios to show the thermal sensitivity of the asphalt bond at high temperature. Its work to resist cracking. The hardness has increased significantly through the decline in the value of stitches at high temperatures to a large extent, the additions have contributed to improve polymer properties at low temperatures and temperature of use (Figure 5).

![Figure 5: Penetration value for each control and PET modified asphalt binder in different temperatures.](image)

When the bitumen is mixed with the polymer, a multi-phase system of these stages consists of a low-grade phase that is not absorbed by the polymer. The viscosity increases because of a more complex internal structure [24,25]. The behavior of bitumen flow in terms of viscosity is either Newtonian or non-Newtonian depending on factors such as crude source, loading, temperature, as well as the nature of the internal structure of bitumen describes the behavior of viscous properties [26]. The state of equilibrium between the polymer particles is
a more harmonious state, which offers less resistance to flow, but increased shear rate provides higher resistance due to the aggregation and particle aggregation in the multi-spherical system as particles between forces such as Vanderfals forces become visible due to high molecular weight. Polymer is not fully distributed in the bitumen liquefaction, elasticity and viscosity in all polymer concentrations and mix. The Newtonian fluid is the fluid in which the shear rate is proportional to shear stress and the fixed ratio, the shear stress to shear rate is the viscosity of the liquid. If this ratio is not constant, the liquid is non-Newtonian (Figure 6).

![Figure 6: Rotational viscosity graph with different temperature with different percentages of PET%](image)

K. Viscosity is the ratio of the viscosity factor to the density of the liquid, a measure of resistance to the flow of liquid under gravity (Figure 7).

![Figure 7: K.Viscosity vs. polymer ratio](image)

Asphalt structure and asphalt samples with the proportion of the polymer were characterized by FTIR technique. The spectrum shows appearance of new peaks as evidence of physical interaction between polymer and asphalt. The peaks in the positions (2919, 2850, 1459, and 1375 cm$^{-1}$) represents the C-H stretching vibration band, C=C stretching vibrations in aromatics, C-H bending vibration band respectively [21,22], and a medium intensity of aromatic C=C, =C-H and C-H bond detected by stretching vibration at 1456, 860, and 723 cm$^{-1}$. Alkane C-H showed a stretching vibration at 1375 cm$^{-1}$, and the ester group bond C-O was indicated by the presence of strong stretching vibration bands at 1031 cm$^{-1}$, and aromatic bands in-plane/out-plane bending at 1031 and 723 cm$^{-1}$[17]. The shape of the asphalt link
before modification shows functional aggregates, which means that the modification of asphalt by polymer is a physical process (Figure 8).[23-26]
Conclusion

The process of modifying traditional bitumen by adding polymers has improved the physical, mechanical and rheological properties by improving the viscosity behavior using the polyethylene terephthalate (PET) and the use of the solvent. The mesh structure and the high melting point at different rates, increasing the softening point and decreasing the penetration value while enhancing the viscous viscosity and periodicity. Spectral analysis FTIR showed that there was a difference in the intensity of the peaks proportional to the ratio of polymer addition to the asphalt link where the aromatic content decreases with increasing the ratio of addition, where increases the intensity depending on the type of polymer and blending ratio. The study showed that plastic waste is a good additive to improving asphalt as well as a practical way to manage plastic waste recycling.

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