Evaluation of Pyrolysis PET Utilization in Asphalt Binder

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Abstract: This study investigates the ability of improving the performance of the asphalt binder by using Polyethylene Terephthalate (PET). The effect of Polyethylene Terephthalate (PET) on asphalt binder properties such as penetration, softening point, ductility, the susceptibility of the modified asphalt to temperature, durability (aging) and storage stability at high temperature was also studied. The results showed that, the morphology and properties of the modified asphalt are dependent on the type of polymer and polymer content. At low polymer contents, the samples revealed the existence of dispersed polymer particles in a continuous bitumen phase, whereas at high polymer contents a continuous polymer phase has been observed. Polymer modification improved the conventional properties of the base bitumen such as; penetration, softening point, temperature susceptibility, this means better performance in hot weather.

Key words: Modified asphalt, PET, Compatibility, Storage stability.

1. Introduction

The asphalt was widely used with aggregates as a binder material since 17th century, it became the optimum choice in the structure of asphalt mixture that used in pavement, but to accommodate ever increasing traffic loadings in varying climatic environments and to resist to failures such as permanent deformation, cracking and water damage, major emphasis has been placed on improving the performance of asphalt mixtures. This approach has led to a fundamental variation in the design of long lasting asphalt pavements [1,2]. For improving the bitumen characteristics, using polymers might be a better solution. There are large types of polymers being used as polymer modified asphalt, such as polyethylene terephthalate (PET), styrene–butadiene–styrene (SBS), styrene–butadiene rubber (SBR), polystyrene (PS), polypropylene (PP) and polyethylene (PE) [3]. However, the major obstacle to widespread usage of PMAs in paving practice has been their tendency to phase separation at elevated temperatures because of the poor compatibility between polymer and asphalt [4].

Zubeck, 1999[5] studied the ability of using PMA by adding (4-6) % SBS and (2-4) % SBR processed in high speed mixer (5000 rpm) at 175 C for 30-40 minutes. He studied the storage stability of modified asphalt at high temperature and concluded that the compatibility and storage stability could be dependent on polymer type, polymer concentration, base asphalt, and method used in mixing the polymer with the asphalt.

Al-Gannam [6] found that the softening point increases as the percentage of pyrolysis polyethylene increases, while the penetration and homogeneity decreases beside the ductility remains (100+ cm) up to 8% polyethylene and began to decrease after this percentage.

Al-Hadidy [7] studied the capability of using pyrolysis polypropylene (PP) to improve the rheological properties of bitumen and concluded that penetration and ductility will be reduced while softening point will be increased with (PP) increasing, and that led to reducing of temperature susceptibility when compared with un modified bitumen.

2. Aim and Objectives of the Study

This study tries to use pyrolysis Polyethylene Terephthalate bottle (which is one of the plastic materials that is widely spread in the world) as modifier asphalt paving to achieve the following aims to:

1- Modify the rheological properties of asphalt.
2- Improve the durability of asphalt paving mixture to resist the conditions of the changing temperature.
3- Reduce life costs of pavements.
4- Reduce the environmental pollution resulted from random discarded polymers which are hardly disintegration by bacteriological creatures.

3. Materials and Laboratory Testing

3.1 Asphalt Binder

(40-50) penetration-grade asphalt cement was used in this study; it was supplied from Baiji refinery (220 Km north of Baghdad the capital). The table (1) displays the physicochemical properties of the bitumen [8-15].

<table>
<thead>
<tr>
<th>Property</th>
<th>ASTM Standard</th>
<th>Test Condition &amp; Units</th>
<th>Test Result</th>
<th>SCRB Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Penetration</td>
<td>D-5</td>
<td>25 °C, 100gm, 5 sec., 0.1mm</td>
<td>48</td>
<td>40-50</td>
</tr>
<tr>
<td>SP. Gr.</td>
<td>D-70</td>
<td>25°C/ 25°C</td>
<td>1.042</td>
<td></td>
</tr>
<tr>
<td>Softening Point</td>
<td>D-36</td>
<td>Ring &amp; ball, °C</td>
<td>54</td>
<td></td>
</tr>
<tr>
<td>Ductility</td>
<td>D-113</td>
<td>25°C, 5 cm/min.</td>
<td>148</td>
<td>&gt;100</td>
</tr>
<tr>
<td>Flash point</td>
<td>D-92</td>
<td>Cleveland open cup, °C</td>
<td>267</td>
<td>Min. 232</td>
</tr>
<tr>
<td>Loss on heat</td>
<td>D-1754</td>
<td>5hrs, 163°C, %</td>
<td>0.24</td>
<td></td>
</tr>
<tr>
<td>Solubility in trichloroethylene</td>
<td>D-2042</td>
<td>%</td>
<td>99.7</td>
<td>&gt;99</td>
</tr>
<tr>
<td>Asphaltene</td>
<td>D-4124</td>
<td>%</td>
<td>24.5</td>
<td></td>
</tr>
</tbody>
</table>

3.2 Aggregates

Al-Khazer aggregate was used in this study and it was crushed in a specialized factory to make it according to SCBR specifications, the properties of aggregates are determined according to ASTM D-127 and ASTM D-131 respectively [16,17]. Table (2) shows the aggregate sieve analysis according to SCR [18]. Table (3) displays the results of the physical properties of coarse and fine aggregates.

<table>
<thead>
<tr>
<th>Sieve size (In)</th>
<th>% (passing)</th>
<th>Research Job mix</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. 1</td>
<td>25.0</td>
<td>100</td>
</tr>
<tr>
<td>¾</td>
<td>19.0</td>
<td>90-100</td>
</tr>
<tr>
<td>½</td>
<td>12.5</td>
<td>76-90</td>
</tr>
<tr>
<td>3/8</td>
<td>9.5</td>
<td>56-80</td>
</tr>
<tr>
<td>No. 4</td>
<td>4.75</td>
<td>35-65</td>
</tr>
<tr>
<td>No.8</td>
<td>2.36</td>
<td>23-49</td>
</tr>
<tr>
<td>No.50</td>
<td>300 µm</td>
<td>5-19</td>
</tr>
<tr>
<td>No.200</td>
<td>75 µm</td>
<td>3-9</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Property</th>
<th>ASTM Designation No.</th>
<th>Coarse aggregate</th>
<th>Fine aggregate</th>
<th>ASTM limits</th>
</tr>
</thead>
<tbody>
<tr>
<td>L.A. abrasion</td>
<td>C-131</td>
<td>20.6</td>
<td>-------</td>
<td>40 max.</td>
</tr>
<tr>
<td>Bulk Sp. gr.</td>
<td>C-127</td>
<td>2.638</td>
<td>2.575</td>
<td>-------</td>
</tr>
</tbody>
</table>

3.3 Filler:

Calcium carbonate (CaCO3) was used as a mineral filler with calculated specific gravity of (2.711) and it was taken from the asphalt plant. Table (4) shows the sieve analysis of the filler used in mixes according to SCR [18].
Table (4) Sieve analysis of filler

<table>
<thead>
<tr>
<th>Sieve size (In)</th>
<th>Sieve size (mm)</th>
<th>Percentage Passing by Weight (%)</th>
<th>SCRIB Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>(No. 30)</td>
<td>0.600</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>(No. 50)</td>
<td>0.300</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>No.200</td>
<td>0.075</td>
<td>72</td>
<td>70-100</td>
</tr>
</tbody>
</table>

3.4 Polyethylene Terephthalate (PET)

The source of PET was water bottles, it is a linear and aromatic [19]. PET can be recycled, and has number “1” as its recycle symbol. Figure (1) and figure (2) show the chemical structure of PET.

![PET recycling symbol][19]

![chemical structure of PET][19]

Figure 1. PET recycling symbol[19]  
figure 2. chemical structure of PET[19]

4. Experimental Works

4.1 Preparing the polymer

PET was shredded by mechanical grinding and was cut into small pieces, then exposed to thermal degradation process by placing it in a preheated large-sized ceramic pot covered with sheets of aluminum in special electric furnace at a temperature of 350 °C for one hour, then it was driven out as a solid waxy piece and was allowed to cool until it reached room temperature, then it was grinded to a very fine powder using an electric machine grinder as shown in figure (3).

![Pyrolysis PET][3]

Figure 3. Pyrolysis PET

4.2 Modified asphalt formulation

The Pyrolysis PET was mixed in proportions (1, 2, 3, 4 and 5) % by weight of asphalt binder at temperature (155±5) °C for about (40-50) minutes at speed (2000 rpm) using electrical mixer.

4.3 Rheological tests of polymer modified asphalt

The rheological properties of PET-modified bitumen such as penetration properties, softening point, ductility, Temperature susceptibility and thin film oven were carried out according to ASTM D-5, ASTM D-36, ASTM D-113 and ASTM D-1754 respectively.

4.4 Compatibility of polymer modified asphalt

The compatibility between polymer and asphalt binder was studied by using the following techniques:
The dispersion uniformity of PET in asphalt binder was confirmed by having the binder pass through sieve No.100 at 165°C. The prepared binder can be stored for future use [20].

Also, an optical microscope was used to evaluate the compatibility between polymer and asphalt binder.

### 4.5 Storage Stability of polymer modified asphalt at high temperature

The storage stability of asphalt binder was tested as follows. The modified asphalt was poured into an aluminum tube with diameter of 30 mm and a height of 150 mm closed from one end, it was placed vertically in an electric oven at a temperature 163 °C for 48 hours. Then the tube containing the modified asphalt was cooled to ambient temperature and cut horizontally into three equal sections. The difference in softening points between the top and the bottom sections of the tube was measured, when the difference was less than 2.5 C, the sample could be regarded as having good storage stability. If the softening points differed by more than 2.5 C, the PMA was considered unstable [21,22,23].

### 5. Discussion of Results

#### 5.1 Penetration test

It is a measure of the consistency or hardness of bitumen. Figure (4) shows the variation of penetration value with the various percentages of PET modified asphalt and it shows that consistency decreases with the addition of polymer. The penetration values for the modified binder decrease as the polymer content in the mix increases. The decreases were 14.89%, 23.4% and 34.04% with the addition (3-5) % of PET as compared to the original bitumen. This means that the addition of polymer makes the modified bitumen harder which means high resistance to deformation in the road pavement but on the other hand this may affect the flexibility of the bitumen by making the asphalt much stiffer, thus the resistance to fatigue cracking can be affected [24].

![Figure 4. Relationship between asphalt content and Penetration](image)

#### 5.2 Softening point test

The softening point is a measure of the temperature at which bitumen begins to show fluidity. The results clearly show that the addition of PET asphalt binder increases the softening point value, and as the polymer content increases the softening point also increases as shown in figure (5). The increases were 7.4%, 12.96% and 15.74% with the addition of PET as compared to the original bitumen. This phenomenon indicates that the resistance of the binder to the effect of heat is increased and it will reduce its tendency to soften in hot weather. In addition to this increase in hardness indicate an improvement in temperature susceptibility with polymer modification.
5.3 Ductility test:

It is measured by the distance in cm to which the binder will elongate before breaking when a standard briquette specimen of a material is pulled apart at specified speed and a specified temperature. From figure (6) it can be seen that PET is keeping the ductility value of (+100) up to (4%) PET.

The results show the high degree of dependence of the chemical composition of the polymer, and base bitumen–polymer compatibility.

5.4 Penetration Index (PI):

It is defined as the change in the consistency parameter as a function of temperature [25]. A classical approach related to PI calculation has been given as shown in the equation (1):

\[
\frac{20-PI}{10+PI} = \frac{\log 800-\log pen}{Trb-T} \quad \text{----- Eq. (1)}
\]

T= Testing temperature for penetration (25°C)

\(Trb\)=Ring and Ball softening point
The Figure (7) shows the effects of the PET on the penetration index of asphalt binder. It is clearly seen that polymer modified have increased PI values with increased polymer content. The high value of PI means the modified asphalt is less susceptible to temperature changes and more resistant to low temperature cracking as well as permanent deformation [25,26]. Generally, it can be said that the asphalt which has suitable properties for pavement should have PI occurs between (-2 and 2).

In general, the variations were occurred on the modified bitumen samples in term of decreasing the penetration, ductility and increasing softening point were related to nature of polymers and characterized by ductility value is less than bitumen, so it is natural to be the final properties for any mixture is the result of the properties of its components. Moreover, the addition of these materials led to change the nature of equilibrium within components of bitumen system.

5.5 Short-term aging in the thin film oven test (TFOT)

The short-term aging test for modified asphalt was carried out by thin film oven test (TFOT) and the loss of weight was conducted after that. Figure (8) shows that the percent loss of weight by heat and air decreases as the PET content increase. The reason is that the polymer occupied a space of the total mix, and consequently caused the reduction of asphalt volume, which leads to a decrease in loss by dehydrogenation and oxidation of asphalt in the mix [7].
5.6 Compatibility of polymer modified asphalt result

A minimum compatibility between the polymer and the asphalt is necessary to avoid separation during storage, pumping, and applying the asphalt and to achieve the expected properties in the pavement. The morphologies of the modified binders measured at room temperature are presented in Figure (9). In the microscopic test, specimens were prepared by taking a drop of sample at 160°C on a glass plate. As expected, the addition of a polymer with a high molecular weight to the bitumen is a difficult process, and the formation of a homogeneous system is not easy. These polymers are difficult to disperse into asphalt binder with addition of 5% PET.

At a low polymer content (4% PET), the polymer exhibits dispersed phase in the binder due to its lowered oil content, the bitumen phase has a higher asphaltenes proportion. As a result, both the cohesion and elasticity of the asphalt are enhanced. In addition, the polymer phase is dispersed through the bitumen matrix. Briefly, the dispersed polymer phase enhances the properties of the binder both at low and at high service temperatures. In this case, the choice of bitumen is a determining factor. At a sufficiently high polymer content, a continuous polymer phase is formed (The polymer phase is the matrix of the system) and the asphalt is there in dispersed. The properties of such a system are depending essentially on those of the polymer [27].

![Photomicrographs of polymer modified asphalt](image)

Figure 9. Photomicrographs of polymer modified asphalt
(A= 0%, B = 4% PET, C = 5% PET).

5.7 Storage Stability Test

The storage stability of modified bitumen has been identified as an important criterion of production and usage of modified asphalts. Typical designs of storage tanks used in this field indicate that most modified bitumen is stored with continuous agitation to obtain uniform temperature and homogeneity in the material [2]. Therefore, an improvement in the storage stability would produce an important cost reduction for the bitumen industry.

Table (5) shows that the difference between the top and bottom part of tubes were less than 2.5 °C up to (4) % PET which give indication that modified bitumen has a good storage stability at high temperature but this difference exceed (2.5 °C) when adding (5)% PET.

<table>
<thead>
<tr>
<th>Polymer content %</th>
<th>PET</th>
<th>Top (R&amp;B)</th>
<th>Bottom (R&amp;B)</th>
<th>Difference (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td>55.50</td>
<td>55.50</td>
<td>0.0</td>
</tr>
<tr>
<td>2</td>
<td></td>
<td>57.50</td>
<td>58.0</td>
<td>0.5</td>
</tr>
<tr>
<td>3</td>
<td></td>
<td>58.50</td>
<td>59.50</td>
<td>1.0</td>
</tr>
<tr>
<td>4</td>
<td></td>
<td>61.30</td>
<td>63.20</td>
<td>1.9</td>
</tr>
<tr>
<td>5</td>
<td></td>
<td>63.0</td>
<td>65.60</td>
<td>2.6</td>
</tr>
</tbody>
</table>

A comparison between stored and original binders indicates that samples from the bottom section become harder and those from the top section become softer at 5% PET. The top section samples are also more elastic, while the bottom section samples are more viscous than their original modified binders.
6. Conclusion

1. PET can be used as a modifier for asphalt binder for sustainable management of plastic waste as well as for improved performance of asphalt.
2. Generally, the addition of PET polymer can decrease the penetration, increase softening point, increase the penetration index, and that is better in hot climates.
3. The addition of PET kept the ductility values at a minimum range of ASTM and SCRB specifications of 100+ cm up to 4% PET content.
4. The percent loss of air and heat decreases with the addition of polymers (i.e. the resistance of asphalt to the action of temperature and temperature changes, and the action of heat increases).
5. Compatibility can be better when the polymer is exposed to thermal degradation.
6. Adding 4% of PET give good homogeneity and storage stability for the modified binder.

References