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Evaluation of the Effect of Waste Polystyrene on Performance of Asphalt Binder

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ABSTRACT

Expanded polystyrene (EPS) is widely used in industry, as a packaging material, construction material, and in a household appliances. Contrary, their wastes (WPS) have disturber environmental effect. So, in our study, aimed to re-use (WPS) as effectiveness product; (WPS) was used to enhancement the performance of hot mix asphalt. Improvement of the performance of the local asphalt; penetration grade (60/70), was mixed with various percentages of (WPS; M.wt. = 77,000) 2%, 3%, 4%, 5%, and 6%. Physical properties of modified and un-modified asphalt as; Penetration value, Softening point, Absolute viscosity at 60°C, Kinematics viscosity at 135°C, and Durability were evaluated through Marshall's tests. The morphology was examined by an optical microscope (OM). The best results were recorded for polymer modified asphalt (PMAs) containing (5% WPS). This amount was improving workability and enhances the resistance to deformation of the asphalt concrete mixture. The thermal properties for PMAs were investigated by Thermal Gravimetric Analysis (TGA).

Keywords: *Polymer modified binder, Waste polystyrene, Road construction, Marshall Tests***.**

1. INTRODUCTION

For years, people have been trying to keep the environment clean. Scientific studies provide us with information about, how we can maintain the natural balance of life, and recycling has a primary role in these studies. Because of natural disasters or increasing population and urbanization, great amounts of waste materials are produced. These waste materials include iron, wood, glass, ceramics, rubber and WPS. Unmodified WPS foams have a cellular microstructure with closed cell membranes made of expandable polystyrene (PS) and its density is typically less than 50 kg/m^3 [1] Polystyrene is a plastic which has been extensively used in packaging. Various investigations have adopted chemical recycling of waste polystyrene into the corresponding monomers or hydrocarbons [2, 3]. However, the process is not efficient because the cost of hydrocarbons and monomers is low compared to that obtained by recycling. Therefore, it is useful to find an efficient technique to recycle WPS [4].

Asphalt (or bitumen) is an important construction material with many applications; however, asphalt binder is used mainly for the construction of roads and airfields. Nowadays, asphalt pavements have to sustain increasingly large loads. When these loads are combined with adverse environmental conditions, the distress modes in pavements lead to the rapid deterioration of road structures. To improve the engineering properties of asphalt, it is frequently blended with various polymers. The modification of asphalt by polymers produces new material called polymer modified asphalt (PMAs), which sometimes has very different mechanical properties from the original base (conventional) asphalt. Generally, PMAs is a material with strong viscoelastic behavior [5]. The use of PMAs is a key component in providing roads with a long service life. PMAs are proprietary binders formed by introducing virgin polymers into straight run bitumen, resulting in a binder with enhanced performance characteristics. The quantities of virgin polymer used can be significant (up to 5%) and results in a binder cost that is considerably more expensive than the straight run bitumen. A feature of current asphalt practice is that PMAs are used where it is essential that roads are designed to resist deformation and fatigue cracking due to the presence of large volumes of vehicles [6].

Others [7] have reported on the incorporation of several waste streams into asphalt, including glass, steel slag, tires and plastics. Polymer modification was found to be effective in increasing rutting resistance at high temperature and fatigue resistance at intermediate temperatures. This behavior was omitted due to micro-structural changes related to the development of polymer-rich phase and molecular arrangements [8-11]. The disturbed structure of polymer modified asphalt PMAs binders during shear was found to reform with time, and this property imparts the ability of self-healing [12]. Essentially in polymer modification, optimization of blend composition, shear rate, temperature and time produces the best rheological properties of modified asphalt binder [13, 14].

In this study, it's clear need to find not only new areas of application for recycled plastics, but also means of adding value to these materials. We used the most commercial waste product in Egypt (waste polystyrene-WPS) and show its performance effect as asphalt binder.

This approach will enhance the commercial viability of the recycling process as a whole and promote a more sustainable approach towards waste management.

2. MATERIALS AND METHODS

Waste polystyrene (molecular weight = 77,000, and its polydispersity = 1.2). The molecular weight was measured by gel permeation chromatography (GPC) was performed at 30 ^oC using GPC-Water 2410, with a refractive index detector using column styragel HR THF 7.8 x 300 mm, equipped with a water 515 HPLC pump. THF was used as an element at a flow rate of 1 ml/min. WPS used to be packaging consisting of white coffee cups. This waste was collected after verifying the ISI code of polystyrene Fig. 1. It was cleaned and dried in the atmospheric environment.

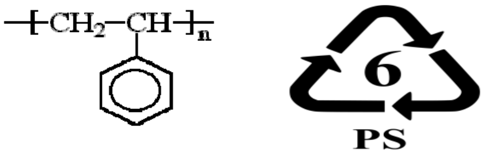

Fig 1: Chemical structure and ISI code of polystyrene

Local asphalt of penetration grade 60/70, produced by El-Nasser Petroleum Company Suez - Egypt was used. Its physical properties and chemical constituents are shown in Table 1.

Table 1: Physical properties and chemical constituents of asphalt cement 60/70.

Properties	Values
Physical properties:	
- Penetration at 25°C 100 g, 5 seconds, 0.1 mm.	61
- Kinematics viscosity at 135°C, C.st.	405
- Absolute viscosity at 60°C, poise..	1916
- Flash point, °C (Cleveland open cup).	+250
- Ductility at 25 °C, 5 cm/min, cm..	+100
- Softening point °C (Ring and Ball).	52
- Solubility in trichloroethylene, %.	99.9
Chemical constituents, wt %:	
- Oils %.	26.0
- Resins %.	49.4
- Asphaltens %.	24.6

- **Aggregates:**

  - Coarse aggregate: The used crushed Limestone aggregate was originally obtained from Ataka Suez-Egypt. The bulk specific gravity and water absorption for coarse aggregate were 2.621g/cm^3 and 1.3% respectively, according to ASTM C127.
  - Sand: Crushed sand was originally obtained from Ataka Suez-Egypt. The bulk specific gravity and water absorption for sand were 2.661 g/cm^3 and 1.8% respectively, according to ASTM C128.
  - Mineral filler: Limestone dust filler of bulk specific gravity and water absorption for mineral filler was 2.621 g/cm^3 and 1.3% respectively, according to ASTM C128. Aggregates, sand and mineral filler used for hot asphalt mixtures were obtained from the station of El-Nile Company for Road Construction. The gradation of fine and coarse aggregates was illustrated in Table 2.

Table 2: Gradation of the applied mixture

Sieve size	Job mix formula	Specification Limits
$1^{//}$	100.0	100
$3/4^{//}$	100.0	100
$1/2^{//}$	96.7	85-100
$3/8^{//}$	90.0	-
No. 4	66.6	65-80
No.8	56.6	50-65
No. 30	37.9	25-40
No. 50	19.8	18-30
No. 100	11.7	10-20
No. 200	9.2	3-10

2.1 Standard Bitumen Tests

The successfully developed binders were subjected to a series of standard bitumen tests for viscosity, softening point and penetration value.

2.1.1 Viscosity

The kinematic viscosity was measured at 135°C and Absolute viscosity at 60°C, according to ASTM D-2170.

2.1.2 Penetration Value

Penetration is a measure of the consistency or hardness of bitumen and is the most common control test for penetration grade. The penetration test was carried out at 25°C according to ASTM D5-97.

2.1.3 Softening Point

The softening point is a measure of the temperature at which bitumen begins to show fluidity. It has a direct influence on the required mixing temperature when incorporating the binder into a bituminous mixture. The softening point of modified bitumen indicates of the improvement in temperature susceptibility of the binder achieved through the addition of the polymer. The softening point (Ring and Ball) was measured according to ASTM D36-95.

In addition, the temperature susceptibility of the modified bitumen samples was calculated in terms of penetration index (PI) using the results obtained from both penetration and softening point temperature tests.

Temperature susceptibility is the change in the consistency parameter as a function of temperature. A classical approach related to PI calculation is reported in the Shell Bitumen Handbook as shown by the following equation:

$$PI = \frac{1952 - 500 * \log(Pen_{25}) - 20 * SP}{50 * \log(Pen_{25}) - SP - 120}$$

Where P_{25} is the penetration at 25 ^{o}C and SP is the softening point temperature of the modified bitumen samples. Lower values of PI show higher temperature susceptibility [15, 16].

2.2 Preparation Of Modified Asphalt Binder

The polymer modified asphalt binder was prepared using a high shear mixer at 180 ^{each} and a speed of 600 RPM. Initially, 600 g of asphalt was heated to fluid condition and poured into a 2000 ml spherical flask. Up on reaching 175 ^{0}C, the waste polystyrenes were added slowly to the asphalt, thus preventing any polymer aggregate formation during the mixing process. Mixing was then continued at the 180 ^{O}C for 2 h to produce polymer modified asphalt (PMAs) [16]. Various blend compositions were prepared according to the following content rates: (2%, 3%, 4%, 5%, and 6% by weight of asphalt), the prepared samples were namely A_1, A_2, A_3, A_4 and A_5 respectively.

2.3 Mix Tests

2.3.1 Marshall Test

Marshall Test method was carried out on all modified and unmodified asphalt mix (asphalt binder with aggregates) according to ASTM D-1559; two principal features of the Marshall method of mix designs are a density-voids analysis and a stability-flow test of the compacted test specimens. The stability of the test specimen is the maximum load resistance that the standard test specimen will develops at 60±1 $^{\circ}$C when tested as outlined. The flow value is the total movement or strain, in units of 0.25 mm occurring in the specimen between no load and maximum load during the stability test. Six different asphalt mixes were prepared for each modified binder beside the sideusing the control binder (4%, 4.5%, 5%, 5.5%, 6% and 6.5% by weight of aggregates). Both faces of the specimen are compacted with 75 blows. Samples rewe extruded from molds and left to cool down before starting test at constant temperature about 25ºC for 24 hr, and then extracted from the mold by using a hydraulic system. The samples were left at 60±1 $^{\circ}$C for 30 minutes. Each read is the average of three specimens. Stability and flow were measured at 60±1 $^{\circ}$C.

2.3.2 Thin Film Oven Test (TFOT)

The procedure according to (ASTM D1754) was developed to simulate the effects of heating in a hot-mix plant operation on asphalt cement. In the standard TFOT procedure, the asphalt cement sample is poured into a flat-bottomed pan to a depth of about 3.2 mm. The pan with the asphalt sample is then placed on a rotating shelf in an oven and kept at a temperature of 163 °C for five hours. The properties of the asphalt before and after the TFOT procedure are measured to determine the change in properties that might be expected after a hot-mix plant operation

3. RESULTS AND DISCUSSION

3.1 FT-IR

The constituents of asphalts occur in a very broad range of molecular weights, shapes and chemical functionalities (including hetero atoms). These characteristics are responsible for different kinds of interactions among molecules: π- π interactions among aromatic rings, van der Waals interactions among aliphatic chains, polar interactions (hydrogen bonding, ionic, etc) involving hetero atoms. These interactions produce intermolecular associations, which are critical to the physical and mechanical properties of the asphalt.

The formation of these products has been verified by infrared spectroscopy. The characterization of the modified and unmodified asphalt was carried out on Nicolet iS10 FT-IR, spectrometer [Thermo Fisher Scientific (USA)]. Fig. 2 shows the infrared spectra of unmodified (a) and (b) after modification.

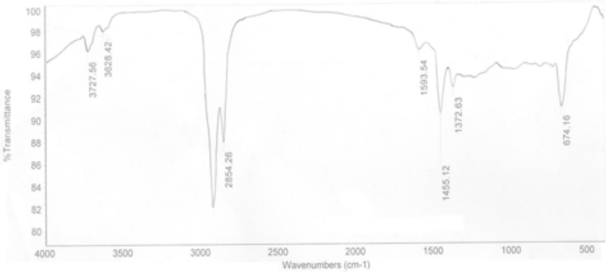

Fig 2. a: FTIR of asphalt without

Bands at 1633 cm^{-1} correspond with carbonyl compounds after modification, which show that it's probably formed in the stage of blending of asphalt with polymer [17], that effected by the heat and the time of blending.

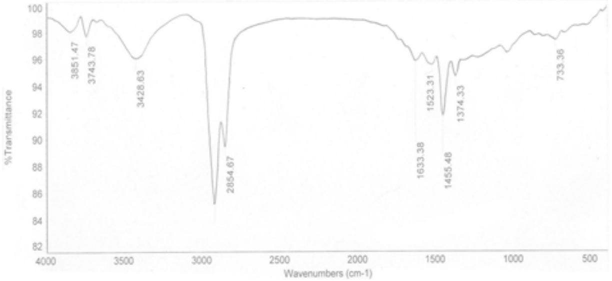

Fig 2. b: FTIR of asphalt with WPS

The broadband at 3428 cm^{-1} corresponding with an O-H group, assigned to the degradation products of the polymer and asphalt [18]. The stretching vibration band and bending vibration band of alkane C–H bonds at 2950–2854 cm^{-1}. It can be concluded that asphalt has no obvious changes in functional groups before and after modification, suggesting that the modification of asphalt with WPS is a physical process [19].

3.2 Standard Bitumen Test Results

The effects of polymer content on physical characteristics of the base asphalt were evaluated and the results are shown in Fig. 3-5 and Table 3. The results indicate that WPS modifier is effective in improving the physical properties of asphalt cement. As presented in Table 3, apparently the addition of WPS to unmodified asphalt decreases the penetration values for all modified binders comparing with the control binder.

The higher decrease on penetration was obtained with 5% and 6% ($A_4$and A_5) modifier contents that equal to 29.5% and 31% respectively referring to the blank sample. Indeed, blending polymers with asphalt often leads to an increase in consistency of the modified asphalts which means high resistance to deformation in the road pavement. This is due probably to diffusion of maltenes (oil fraction of bitumens) in the polymeric phase causing swelling and also to interactions between the polymer modifiers and polar molecules of asphaltenes [20].

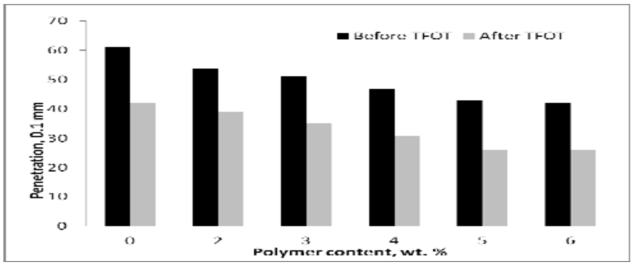

Fig 3: The penetrations of the modified and unmodified asphalt after and before TFOT

However it noted that penetration seems to be less affected by polymer loading rates even though a small decrease in the penetration values of the modified samples is observed within the range 2–6 wt. %., this decrease of penetration indicating higher resistance to deformation.

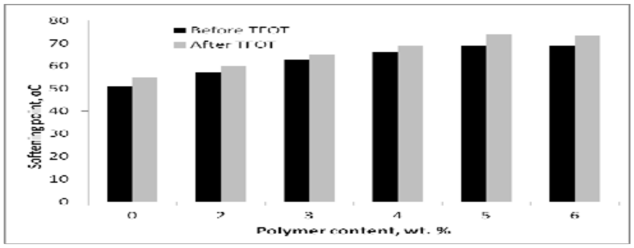

Fig 4: Softening point of the modified and unmodified asphalt after and before TFOT

The softening point values were increased with the addition of WPS modifier. The study showed that the softening point of unmodified asphalt was raised by 11.7% to 35.2% it's equal to 6°C to 18°C with the addition of PMAs. It can be concluded that the WPS enhances the softening temperature of asphalt, thus improving its high temperature stability [19].

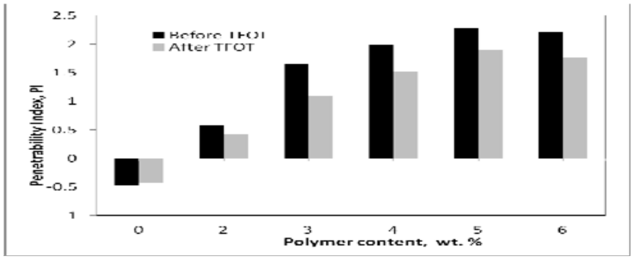

Fig 5: The penetrability index (PI) of the modified and unmodified asphalt after and before TFOT

The softening temperature increased with increasing the modifier percent up to 5%. This may be due to the stiffness of modifiers.

Table 3: Physical properties of modified and unmodified asphalt before TFOT.

properties	**Base asphalt**	**WPS modified asphalt, wt.%**				
		2	**3**	**4**	**5**	**6**
Penetration, 0.1 mm	61	54	51	47	43	42
Softening point, °C	51	57	63	66	69	69
PI	-0.48	0.59	1.65	1.99	2.28	2.22
Kinematic viscosity at 135 °C, C.st.	405	633	726	818	962	991
Absolute viscosity at 60 °C, poise	1906	2015	2205	2409	2712	2906

Temperature susceptibility of modified bitumen samples is evaluated by the determination of the penetration index (PI), which is the change in the consistency parameter as a function of temperature [16]. Higher penetration index (PI) indicates less temperature susceptibility and more rubbery elastic behavior. From Fig. 5 and Table 3, it is observed that the polymer modification by WPS significantly reduces the temperature susceptibility of the base bitumen. The results indicate that the initial PI value of the base asphalt is -0.481and this value increases with increasing the polymer contents reach to 2.28 for A_4. This clearly shows that polymer modification induces a rubbery elastic behavior conferring to the bitumen better resistance to low temperature cracking and permanent deformation [15].

The viscosity of asphalt binders (absolute viscosity at 60°C and Kinematic viscosity at high temperature 135°C is considered to be an important property because it represents the binder's ability to be pumped through an asphalt plant, thoroughly coat aggregate in asphalt concrete mix, and be placed and compacted to form a new pavement surface. The results in Table 3 show that, the increasing of PMAs the increasing in the binder viscosity at both testing temperatures, compared to the control binder. The increase percent in absolute viscosity at 60°C ranged between about 8% to 52.4% for A_1 and A_5 respectively. Also, at 135°C the increase percent in Kinematic viscosity ranged between about 56% and 144.6% for A_1 and A_5 respectively. The maximum increase was found with 6% PMAs compared to control binder. The higher the viscosity index value, the more aged the sample [21]. The increase in the viscosity value at high temperature is a good property for rutting resistance. It is important to record that the difference between 5% and 6% polymer content is not significant.

3.3 Physical Characterization Of The Modified And Unmodified Asphalt After Ageing (Tfot)

The basic properties of the unaged and aged polymer modified asphalts were evaluated. Fig. 3-5 and Table 4; show the summary of the measured properties of unaged and aged polymer modified asphalts. Basic test properties indicated that the addition of polymer content to base binder reduces the penetration and percentage loss of heat values, whereas increases in softening point values were observed with the addition of polymer modifier. The study showed that the increase in softening point values ranged between about 8 ºC and 22 ºC, for 2% and 5% polymer content, compared to base binder. Whereas the penetration values were found to be lower by 64% and 43%, and loss of heat values were found to about 0.22 and 0.196 for 2% and 5% polymer content respectively.

The increasing in softening point and the reduction in penetration values of base binder with the addition of polymer modifier can be attributed to the hard consistency of polymer-asphalt binders and also the completely dissolved in base binder. The reduction in percentage loss due to heat and air (Thin Film Oven Test) related to that polymer occupied a space of total mix and causes reduction in asphalt volume, (i.e. Durability increased slightly with the addition of polymer content in the mix).

Table 4: Physical properties of modified and unmodified asphalt after TFOT.

Properties	Base asphalt	WPS modified asphalt, wt.%				
		2	3	4	5	6
Penetration, 0.1 mm	42	39	35	31	26	26
Softening point, °C	55	60	65	69	74	73
PI	-0.43	0.43	1.11	1.51	1.91	1.76
Loss on heating, %	0.220	0.196	0.19	0.182	0.17	0.196

The penetrability index (PI) after thermo-oxidative aging (TFOT) was shown in Fig. 5 and Table 4. All unmodified and modified samples exhibit a large decrease of penetration, while softening point temperature and PI values have increased. These results clearly indicate a hardening of the material, the increased penetration index (PI) observed for the modified samples after aging indicate a significant reduction in temperature susceptibility with polymer modification, particularly at higher polymer contents.

3.4 Marshal Test

The optimum asphalt content (OAC) for the modified and unmodified mixes that were summarized in Fig. 6, 7 and Table 5. All modified binder mixes have slightly increased in optimum asphalt content by 0.1 watts. %. The increasing of polymer content raises the Marshall stability of control mixes in rang 8%-25% for A_1-A_4, respectively.

Mix No.	Poly. cont. , %	Asph. cont. %	Dens. (T/m$^{3)}$	Stabil. (N)	Flow (mm)	Air void %
Control	0	5.2	2.367	9890	2.9	3.6
Mix 1	2	5.30	2.358	10710	2.70	3.8
Mix 2	3	5.30	2.357	10920	2.60	3.9
Mix 3	4	5.30	2.355	11450	2.58	3.9
Mix 4	5	5.30	2.351	12340	2.55	4.0
Mix 5	6	5.30	2.352	12450	2.51	4.0

Table 5: Effect of modifier type and modifier content on Marshall Test results.

When modifier percent increased from 5% to 6% the stability value was slightly increased about 1% compared with control mix made from the same dense grade aggregate. This may be due to the increases in the viscosity value of the modified binders lead to formation of thicker binder film in the mix. Whereas decreasing in flow value was observed with increasing the polymer content.

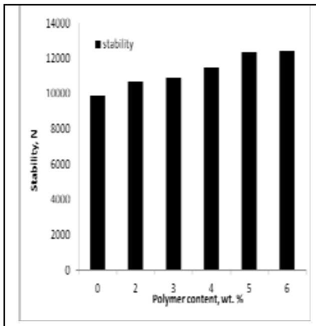

Fig 6: Effect of polymer content on stability.

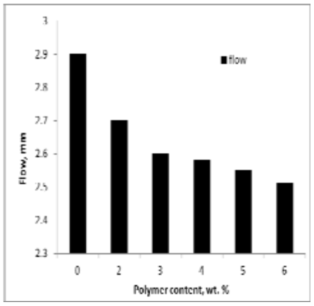

Fig 7: Effect of polymer content on flow.

As indicated in Table 5 all modified asphalt samples satisfy the specified limits of 3–5% air voids [22]. The addition of modifier to asphalt binder slightly reduced the flow value of the mixes in range of 7% -13%. This is attributed to the stiff nature of PMAs.

3.5 Thermal Analysis (TGA and DTA)

The thermal stability of modified asphalt is an important property to be considered in the analysis of the structural characteristics of asphalt binder [23]. The thermal stability of PMAs was studied by thermo gravimetric analysis using TA Instruments SDTQ 600 simultaneous TGA-DSC thermo gravimetric analyzer. The analyses were conducted for a total sample mass of 7.0 ± 0.4 mg. A known amount of the sample was loaded and evenly spread on the alumina micro cube. The samples were heated under nitrogen at a temperature ranging from 50 to 800°C, at 10 °C min^{-1}, the thermodynamic behaviors of all samples are similar, which can be illustrated by that of the 5% by weight. of modified asphalt as shown in Fig. 8 and Table 6.

We can show that all the samples undergo a single mass loss process. The degradation occurred in the temperature range, 250–550 °C, and the mass loss of binder was mainly due to the volatilization of light asphalt components such as saturates and aromatics and the decompositions of asphaltenes and WPS [24].

Table 6: Thermo grave metric characteristics in N_2.

Sample	Onset temp. $^0C^{-1}$	Max. Temp. DTA $^0C^{-1}$	Mass loss% 250-550 ^0C
Control	384	447.8	83.26
3%	381	451.2	85.01
4%	388	452.9	83.93
5%	390	454.3	83.99

The addition of polymer to asphalt improves its thermal stability of the data of Table 6 the onset temperatures of the mass loss process of the samples indicate that the A4 has the greatest thermal stability, followed by A_3 and A_2 the lowest stable but more stable than the control sample.

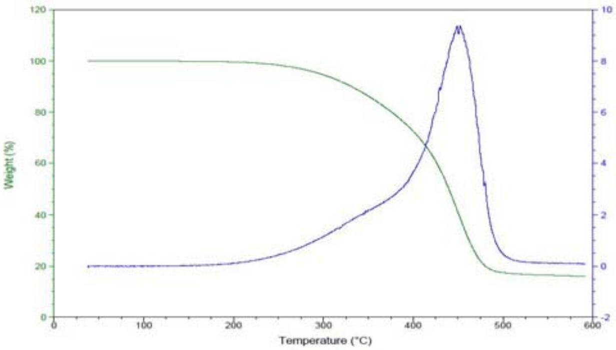

Fig 8: Thermodynamic behavior of 5% wt.WPS modified asphalt.

Because the shape of the multiple peak of DTG curve was very irregular for all binder samples and the differences among peak values in each multiple peak were very small, so it was unreasonable for us to evaluate the thermal stability of the binder by using the maximum decomposition temperature of DTG curve.

3.6 Morphology

The compatibility between polymer and asphalt is critical to the properties of PMAs [25].

Fig.9 a. morphological properties asphalt without Polymer additives

The morphology of PMAs is usually investigated using an optical microscope (Binocular Olympus Microscope with Digital camera 5050-R) by characterizing the distribution and the fineness of polymer in the asphalt matrix. Fig. 9 shows a) the original micrographs of the asphalt without additives and b) WPS-modified asphalt 5 wt% on a hot plate at 160°C.

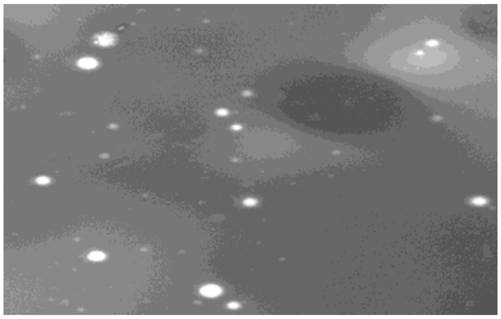

Fig 9. b: morphological properties asphalt with WPS 5% wt. additives

Apparently the white PS particles are a good dispersed in the asphalt matrix (appears black) coarsely and has a uniform dispersion. The size of PS particles becomes smaller. This means that the compatibility of WPS-modified asphalt has been improved significantly. Hence, this blend can be stored at elevated temperature.

4. CONCLUSIONS

The mechanical properties of the base asphalt binder are highly affected by the addition of the polymer WPS. Analysis showed that, the viscosity was significantly increased at the temperatures of 60°C and 135°C (42% and 137% respectively), the softening temperature increase by 18°C than the base asphalt binders, the penetration decreased by 29.5%. All modified binders gave higher stability values, increase in air voids and slightly decrease in flow. The morphological properties of the PMAs evidenced the compatibility between polymer and asphalt matrix. In general, the strategy behind modifying asphalt is to depend on the base asphalt to provide good low temperature properties while depending on the modifier to provide good high temperature properties. On the other hand to resolve the environmental pollution problem caused by the huge production of WPS and use it in industrial application.

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