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RESEARCH ARTICLE

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Laboratory Evaluation of PE, PP, and SBS Polymers as Bitumen Modifiers

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Article History

Received: 15.01.2023 Revised: 09.02.2023 Accepted: 18.02.2024 Published: 31.03.2024 Communicated by: Assis. Prof. Dr. Abubakar M. Ashir *Email address: <u>mohammed.qadir@tiu.edu.iq</u> *Corresponding Author

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Abstract:

Reactive polymers, such as PE, PP, and SBS, cause a chemical reaction between the polymer and the Bitumen, which can be advantageous for hot-climate asphalt applications. High-density polyethene (HDPE), polypropylene (PP), and styrenebutadiene-styrene (SBS) were used to create elastomeric-modified-bitumen mixes. Modified Bitumen's physical and rheological properties were investigated by containing different polymers percentages (2%, 4%, 6%) into 60/70 Bitumen grade. The results of penetration, flash, fire, softening point, viscosity, elastic recovery and Penetration Index tests revealed an increase in elastic behaviour in modified asphalts, as a result of which material properties suitable for highway construction are obtained. According to the results of physical and rheological tests, the temperature range of asphalt increases by adding polymers and increase the permanent resist deformation as well. Penetration Softening and Elastic recovery tests result in the asphalt modification being elastomeric with good performance at intermediate temperatures stage, especially for the KRG region.

Keywords: Polymer, Modification, Bitumen, Polyethene, Polypropylene, and Styrene Butadiene Styrene, Physical Properties, Rheological Tests.

1. Introduction

Asphalt, also known as Bitumen, is widely used in road construction because of its waterproofing and viscoelastic properties [1]. Bitumen is extracted from crude oil's heaviest fraction. Despite its complex chemical structure, its constituents are typically classified into four general chemical groups: asphaltenes, aromatics, resins, and saturates. It is utilized as a binder component in the production of flexible pavement. Flexible pavement is less susceptible to ravelling than rigid pavement and is simpler to monitor and maintain. As general, the most desirable features in Bitumen are resistance to durability, rutting and cracking, and low-temperature resistance. [2].

Usually, Bitumen from nature does not meet certain specifications, necessitating the addition of other components to enhance its characteristics. This may involve polymers, chemicals, or rubbers. After these substances have been added to Bitumen, It is also known as Modified Bitumen (MB). These additives have showed their capacity to enhance Bitumen's resistance to rutting deformation, low

temperature, fatigue cracking, and binder-aggregate adhesion ([2]). However, they have raised the price of the binder.

In general, polymers are used as binder modifiers, and they are classified into three types: plastomers, elastomers, and reactive polymers. Therefore, they were utilized in Bitumen mixtures and blends to enhance the performance of Bitumen [3]. These PMs can improve the temperature susceptibility of asphalt binders and each category affects their properties. Thermoplastic elastomers deal with the elastic properties of the binder, which increases fatigue resistance, and plastomers or reactive polymers deal with stiffness resistance and deformation corresponding to the applied load [4-5]. The most effective mixture happened when the polymer content was used sufficiently with Asphalt which did not cause phase separation during the storage and application. Moreover, it increased the rutting resistance so that it did not reach too viscosity range in high-temperature and vice versa [6] Besides their advantages, polymers increase the asphalt mixture by 30-40% of their cost [4].

Previous studies used the highest content of polymers (5-10%) by bitumen weight. However, in the last few years, researchers preferred a lower polymer content, between (2-4%) of waste product materials because of their high content proportions [7]. The modification process depends on polymers content and the chemical composition of the polymer structure. PMs have no significant effect on mixture stiffness at low temperatures, while at high temperatures, the effect on stiffness increases [7-8].

2. Polymer Modified Bitumen

MB with polymer provides number in terms of performance advantages; Bitumen's physical characteristics are enhanced without affecting its chemical composition. According to reports, At low service temperatures, these modified bituminous binders provide softer mixes, minimizing non-load-associated thermal cracking. Furthermore, It will improve bituminous mix fatigue resistance, performance in adverse climatic conditions, and performance under high traffic circumstances, while also lowering pavement life cycle costs.

At higher temperatures, Polymer Modified Bitumen enhances the flexibility and viscosity of the mix [9]. The viscosity contributes in limiting deflection, whereas elastic recovery lowers residual deformation. The elastic top layer bridges fractures from unmodified layers underneath, keeping the Asphalt watertight and safeguarding the underlying structure.

Bitumen viscosity is defined as its internal resistance to flow or resistance to deformation by shear or tensile stress. It is an important characteristic in defining the bitumen rheology and technical features of asphalt concrete; it affects workability and mixing resistance [10]. Bitumen viscosity responds complexly under diverse situations, influencing the mixing, laying, and compaction of asphalt mixes as well as pavement performance[11]. Since of these factors, viscosity measurements have been used to categorize Bitumen; however, because Bitumen is viscoelastic, the findings lack consistency as the bitumen source and molecular composition may change[12].

Collins et al. (1991) said that "choosing right Asphalt is critical in order to create a mix with optimum qualities". Improved compatibility has several benefits. Furthermore, the additional polymer's efficiency in elastic recovery decreased from a soft to a hard binder [13].

Lenoble and Nahas (1994) demonstrated that the use of polymer extends the application temperature range of asphaltic binders and improves traffic resistance. Furthermore, the temperature at which the binder achieves a modulus near to its glassy modulus controls the thermal cracking resistance of pavement[14].

Airey and Brown (1998) studied the chemical and rheological changes of polymer-modified Bitumen under short- and long-term ageing conditions. Age-related rheological changes included a reduction in penetration and an increase in viscosity and softening point. It was also established that any change in the ageing behavior of the PMB is related to the presence of the polymer [15].

Edwards studied the rheological characteristics of commercial waxes and polyphosphoric acid in Bitumen at high and medium temperatures. The researchers discovered that adding polyethene wax to Bitumen changes its rheological characteristics at medium and high temperatures. The effect of styrene-butadiene-styrene (SBS) polymer modification on bitumen viscosity was also investigated. The study's findings revealed that SBS-modified Bitumen had a higher degree of non-Newtonian behaviors [16].

Socal et al. (2004) investigated the qualities of polymer-modified binders and discovered that Styrene Butadiene Styrene (SBS) polymer-modified binder has greater elastic recovery than Linear Polyethylene (PE) binder [17].

The most common polymers used by the researchers as PMs are styrene-butadiene-styrene (SBS), polyethene (PE) and polypropylene (PP) [18–21]. SBS is an excellent example of an elastomeric polymer with a high level of elastic recovery, and PP has a high stiffness and deformation resistance in the plastomeric polymer categories[22]. According to a report in 2016-Europe, polypropylene took second place in polymer production, equal to 19.1% of the European Union's global production[23]. The most widely used polymer in binder modifiers is styrene-butadiene-styrene (SBS), and its key benefit is that it has two phase transitions which are composed of glassy microdomains, which are polymerized by polystyrene and connected with rubber polybutadiene segments, showing crosslinked elastomer properties coefficients[24].

3. Materials And Methods

3.1 Asphalt Binder

One local Bitumen supply was investigated, with grades (60/70) from the Lanaz refinery in Erbil/Kurdistan Region, Iraq. Table 1 shows the tests result of the asphalt binder properties.

3.2 Polyethene (PE)

Polypropylene (PE) Hi0500 is a natural high-density polyethene (HDPE) injection modeling which combines with good physical properties [5]. Table 2 shows the Physical properties of the PE material used as a polymer modifier.

Test Method	Controlled Bitumen (60-70)	Standards
Penetration, at 25°C	68	ASTMD5
Flash point	286°C	ASTMD92
Fire point	337°C	ASTMD92
Ductility at 25°C	150CM	ASTMD113
Sp. Gr. at 25°C	1.04	ASTMD70
Softening Point	48	ASTMD36

Table 1: Properties of	f the Controlled	Bitumen (60–70) grades
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Test Method	Controlled Bitumen (60-70)	Standards
Elastic recovery at 25°C	17.77%	ASTMD6084
Viscosity at 135°C	647.7	ASTMD4402

3.3 Polypropylene (PP)

Polypropylene (PP) SABIK PP-575P is a thermoplastic polymer made from oil refineries and also known as polypropene, which is partly crystalline and non-polar [5]. The properties of polypropylene polymer material are shown in Table 3.

3.4 Styrene Butadiene Styrene (SBS)

SBS D1192 is a linear porous pellets copolymer consisting of styrene and butadiene with a bound styrene mass of 30%. [5]. Table 4 also covers the physical features of SBS-D1192 polymer.

Test Method	PE	Standards
Mass Density at 230°C	0.963-0.967g/cm ³	ASTM D1505
Melting point	130°C	ASTM D2117
Vicat Softening Point	124°C	ASTM D1525
Melt Flow Rate at 1900C/2.16 kg	2-6g/10min	ASTM D1238

Table 2: Physical properties of PE (Adapted from [25])

Table 3: Physical properties of PP (Adapted from [26])

Test Method	PP	Standards
Melting flow rate, 230°C/2.16 kg	10.5 g/10min	ASTM D1238
Vicat softening point	153°C	ASTM D1525
Density	0.905 g/cm3	ASTM D792
Melting point	160–170°C	ASTM D7138-16
Melt Flow Rate	4-8g/10min	ASTM D1238

Table 4: Physical properties of SBS (Adapted from [27])

Test Method	SBS-D1192	Standards
Specific Gravity	0.94	ISO 2781
Bulk Density	0.4 kg/dm3	ASTM D1895-B
Melting point	170-180°C	ASTM D7138-16
Melting flow rate, 200°C/5 kg	<1 g/10 min	ISO 1133

3.5 Sample Preparations

The melt blending technique was used in preparing the samples. The first step started with heating the controlled Bitumen of about 600gm in the oven for 30 minutes at 170°C. The second step started with mixing different polymer contents (2%, 4%, 6%) replaced by the weight of Bitumen, then added slowly into the Bitumen and blended with a shear mixer for an hour in order to get the homogenous mixtures. Then the modified bitumen was kept in different container labeled and covered with aluminum foil and stored for further tests.

3.6 Experiment Work

The experimental work was carried out in Erbil Polytechnic University, college of engineering, Iraq, construction material and asphalt laboratory. The experimental work investigating the bitumen grade 60/70 performance a considered as a control when different polymers (PE, PP, and SBS) were added to the Asphalt with different percentages (2%, 4%, and 6%).

3.6.1 Flash and Fire Test

ASTM D92-2018 stated that "flashpoint is the lowest temperature of sample for which application of an ignition source that causes the vapours of the test specimen to ignite under specified circumstances of the test.". This test addresses the safety issues of fires happening in Asphalt during the mixing, spreading, and compaction phases.

3.6.2 Penetration Test

According to ASTM D5-2013, of penetration test method determine the penetration value by using 0.1 mm of standard needle vertically dropped on the asphalt sample surface for 5 seconds and having 100 gm of total weight at 25 °C. This test demonstrates the binder temperature resistance and checks the consistency of Asphalt. A higher penetration value of specimens shows softer asphalts, while a lower value shows stiffer asphalts. Figure 3.10 A and B illustrate the applied penetration and device tests.

3.6.3 Ring and Ball (Softening Point)Test

This test's result was based on ASTM D36-2014 to reflect the binder's temperature susceptibility. The softening point means that the standard ball on the binder, at which temperature starts to fall at 25.4 mm, passes through a ring which is immersed in 5 °C water, as shown in figure 3.11 A and B.

3.6.4 Penetration Index

Penetration index (PI) is a quantitative measurement of the behaviour changing of binder due to variation in temperature, which shows its temperature susceptibility and lower temperature susceptibility of Asphalt protected by a higher value of penetration index. PI can be calculated according to ASTM D3381-2013, and PI value must be ranged between (-2 to +2) as mentioned by[28] . Where P stand for penetration test value at 25°C and RB is Ring and Ball test value, by °C as shown in Eq. (1).

(1)
$$PI = \frac{1952 - 500\log(p) - 20RB}{50\log(p) - RB - 120}$$

3.6.5 Ductility Test

The tensile behaviour of binders was measured using the ductility test by ASTM D113-2017. The test condition elongated in a centimeter measure until the asphalt specimen broke at 25°C and was pulled by a standard motor with a 5 cm/min rate. Figure 3.12 shows a ductility test of binder specimens.

3.6.6 Elastic Recovery Test

The elastic recovery test evaluates the recoverability of materials after applied strain because of the elongation of the binder under the test condition of ASTM D6084-2018. The test was done on the ductility test device and using elastic recovery models at 25°C with a pulling rate of 5 cm/min until elongated the specimens for 10 cm, cut in half with a scissor, and left in a water bath for 60 minutes at 25°C. The result took place after moving back the travelling carriage of the device to a position where the ends of the samples just touched. Eq. (2) was used to calculate the elastic recovery test. Where E used as original elongated of the sample, 10 cm and X used as elongated the sample at the end of the test by cm.

(2) E. R. (%) =
$$\frac{E-X}{E} \times 100$$

4. Findings and Discussion

4.1 Flash and Fire Test

The flash and fire point tests were performed for asphalt binders' samples with PE, PP, and SBS contents of 2%, 4%, and 6%. The higher polymer contents led to a higher temperature requirement to create bubbles on the surface of the samples and avoid the Asphalt from flashing, while all the test results are according to ASTM D946-2015 (>230 °C) and Iraqi standards, SORB-2007 (>232 °C), as shown in Figures 1 and 2.



Figure 1: Flashpoint test results for different polymers content.



Figure 2: Firepoint test results for different polymers content.

4.2 Penetration Test

According to the results shown in Table I and Figure 3 for controlled Bitumen and modified Bitumen, the sharp decrease in the penetration value of 68 dmm for controlled Bitumen to 37,30, and 35 dmm for PE, PP, and SBS at 6% polymer concentration indicates that the increase in the hardness of the PMB was caused by the use of high molecular weight polymers with different melt flow index, but this effect could be reduced by minimizing the percentage polymers content. As result, deciding based on melt flow index is not the final answer since these values are dependent on the force and pressure used during melt flow index calculation for a certain kind of polymer. It is clear from the observations that thermoplastics have a greater impact on penetration as the viscosity of the Bitumen increases [29].



Figure 3: Penetration test results for different polymers content.



Figure 4: Softening Point test results for different polymers content.

Because the melting temperatures of PE and PP are 160°C and 170°C, respectively, it absorbs some oil and releases a low molecular weight fraction into the Bitumen, increasing the viscosity of the PMB [30]. As a result, the viscosity has increased at the conclusion of the mixing procedure, and a hardened mixture has formed. Bitumen hardening may be useful since it enhances the material's stiffness and hence the structure's load-spreading capacities but it can be also contribute to fretting or cracking[29]. Performance tests could be used to assess this at a later stage of the research. The findings of the penetration are very disputed, despite the fact that they are commonly utilized in the paving sector since the penetration needle displaces a relatively small amount of fluid [31].

4.3 Ring and Ball (Softening Point) Test

The results from the ring ball shown in Table I and Figure 4 show a significant difference in the softening temperature for PMBs from 2% polymer concentration in Bitumen to controlled Bitumen as thermoplastics modification significantly affects the softening point as compared to the penetration[29]. One of the reasons is that the internal structure generated by the polymer appears to be thermodynamically stable, which substantially impacts the melting points of the PMB. This is a bad indicator that should be avoided. The performance test at the latter stage of this research will determine if it improves the pavement performance characteristics in terms of rutting, fatigue, and temperature susceptibility. The use of polymers enhances the rigidity of modified Asphalt, according to test data.

4.4 Specific Gravity

Figure 5 shows the effect of different content of polymers on the specific gravity property. The specific gravity decreases by increasing the polymers' content but with different ranges. The specific gravity of the polymer types does not have the same trend and behavior by increasing the PP content and it has a more significant impact on decreasing the Specific gravity than PE, and SBS polymers compared to controlled Bitumen.

4.5 Penetration Index

According to Pefiffer and Doorma, the Penetration Index is another approach to assess the temperature susceptibility of Bitumen. It evaluates the change in bitumen behavior from Newtonian to non-Newtonian. Bitumen is typically rated between +1 and -1 for road construction, with values less than -2 demonstrating Newtonian behavior with brittleness at a lower value, and values larger than +2 being less brittle and exhibiting strong elastic characteristics under higher stresses[29], [32]. According to the data presented in Figure 6, all of the PI values in this research are less than -0.6, with the exception

of SBS polymer and higher polymer concentrations in the blend, indicating that PMB blends are temperature sensitive. The increased amount of SBS and PE demonstrates that Bitumen's behavior has shifted from brittle to less brittle.



Figure 5: Specific Gravity results for different polymers content.



Figure 6: Penetration Index results for different polymers content.

4.6 Ductility Test

Figure 7 illustrates the ductility property losses of different content of polymers, which can be seen that both polymers of PE and PP have sharply decreased in ductility value, while SBS shows a distinct trend of behavior results. Therefore, adding 6%, SBS will cause more decrease in ductility values, which reduces compared to the controlled Bitumen. These decrements were caused by the PE and PP polymers absorbing more oily Materials in the asphalts than the SBS particles.

4.7 Elastic Recovery Test

The elastic recovers "E.R." at 25 oC, influences the degree to which a material resumes its original form after application and release of stress. To prevent persistent deformation, the pavement should have a high degree of elastic recovery. Figure 8 shows the values of E.R. (%) of controlled Bitumen compared to the different content of polymers. Elastomeric polymers used in Asphalt blending increase elastic characteristics, possibly decreasing asphalt binders' permanent deformation. The PE and PP Modified Polymers, show an increase in elastic properties from 18% of controlled Bitumen to 26%, and 20% recovered, respectively. While the small amount of SBS polymer content increase in the elasticity Modified Bitumen. Sample SBS had the highest E.R value, with an 89% recovery rate.

Depending on the composition and source of the crude, the flow behavior of the material represented in terms of viscosity shows Newtonian and non-Newtonian properties. Temperature and stress also have an impact on the behavior characterizing the viscoelastic characteristics of the material. Therefore, the internal structure of the base bitumen is also important [33]. A viscosity of Bitumen with 648 at 135°C increases in viscosity as the polymer content increases. Figure 9 shows the viscosities of PE, PP, and SBS modified asphalt binders. These blends contain (2%, 4%, 6%) polymers. However, failure to meet the viscosity limit should not be regarded as a definitive disqualification. It also implies that such asphalt binders need more significant mixing and compaction temperatures. polymers cause an increase in viscosity. This can be seen in Fig. 9, with the first and second grouped bars of each group with 2% polymer content yielding a further rise in viscosity in most situations. In addition to the PE, or PP present in the asphalt binder phase, introducing suitably stiff SBS into the existing PE, or PP microstructure helps form some similarly strong SBS connections. This increases interlayer friction and as a result, the viscosity of the SBS-modified asphalt binders. The majority of the modified asphalt binders fall within the normal suggested viscosity limits. This phenomenon may be seen in both PP and SBS treated Asphalt with viscosities greater than 1500 cP. The discovered behaviors may be used to overcome one of the issues of polymer binders which is the high viscosity associated with the polymer content, but natural behavior will be acquired only when fatigue and rutting tests are performed at a later stage of this work.



Figure 7: Ductility results for different polymers content.



Figure 8: Elastic Recovery results for different polymers content.



Figure 9: Viscosity results for different polymers content.

5. Conclusion

The experimental investigation demonstrates that polymer modified Bitumen improves ordinary Bitumen (60/70), and the following conclusion can be made from this study: The findings proved the effectiveness of incorporating polymers into asphalt binder. When compared to controlled Bitumen or unmodified binder, polymer addition improves the physical and rheological characteristics of the modified binder. Penetration, flash, fire, softening point, Penetration index, ductility, elastic recovery, specific gravity, and viscosity were all measured in this research. The empirical tests were carried out at 25 oC and revealed an increase in the percentage of elastic recovery of the modified binder when polymers were added to Asphalt. The softening point raised with polymer addition, indicating that persistent deformation improved. The inclusion of polymer led to an increase in PG, which improved the behavior of the asphalt binder across a more comprehensive temperature range. Polymers increased the complex modulus, improving modified binder's permanent deformation. Finally, the application of SBS polymer improved the elastic behavior of Bitumen. The addition of SBS increased both stiffness and elastic responsiveness. These polymers may provide a modified asphalt mixture with excellent operating properties at high and moderate temperatures.

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