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Abstract

In the present study, the effects of styrene-butadiene-styrene (SBS) and American gilsonite (AG) on the stiffness and thermal sensitivity of bituminous binders were investigated in bitumen modification. Şanlıurfa province was chosen as the application field. It was determined that 18% AG and 5% SBS additives should be used separately to obtain the adequate binder for Şanlıurfa province. It was also determined that 13% AG with 2% SBS, 10% AG with 3% SBS and 6% AG with 4% SBS were required to obtain binders with the same performance level. Penetration, softening point and rotational viscometer tests were conducted on neat and modified bitumen to determine the consistency of binders. Furthermore, thermal sensitivities of binders were determined from penetration index and penetration viscosity number values. It was determined that all additives increased the consistency of the binders, the most effective additive type and ratio was 18% AG on penetration values, and all additives had similar effects on the softening point and viscosity values. In addition, it was determined that the temperature susceptibility was reduced with additive use and the most effective contribution came from 5% SBS.

Keywords: Bitumen, Styrene-butadiene-styrene, American Gilsonite, Consistency, Temperature susceptibility.

Bitüm Modifikasyonunda Stiren-Butadien-Stiren ve Gilsonit'in Birlikte Kullanımının Bağlayıcıların Sertliği ve Isı Hassasiyetine Etkileri

Özet

Bu çalışmada, bitüm modifikasyonunda stiren-butadien-stiren (SBS) ve Amerikan Gilsoniti'nin (AG) bitümlü bağlayıcıların sertliği ve ısı hassasiyeti üzerindeki etkileri araştırıldı. Uygulama alanı olarak Şanlıurfa ili seçildi. Şanlıurfa ili için uygun bağlayıcı elde etmek amacıyla % 18 AG ve % 5 SBS katkı maddelerinin ayrı ayrı kullanılması gerektiği tespit edildi. Ayrıca aynı performans seviyesine sahip bağlayıcılar elde etmek için % 2 SBS ile % 13 AG, % 3 SBS ile % 10 AG ve % 4 SBS ile % 6 AG kullanılması gerektiği tespit edildi. Bağlayıcıların kıvamını belirlemek için saf ve modifiye bitümler üzerinde penetrasyon, yumuşama noktası ve dönel viskozimetre deneyleri yapıldı. Ayrıca bağlayıcıların ısı hassasiyetleri, penetrasyon indeksi ve penetrasyon viskozite sayısı değerleri yardımıyla belirlendi. Tüm katkı maddelerinin bağlayıcıların kıvamını arttırdığı, penetrasyon değerleri üzerinde en etkin katkı türü ve oranının % 18 AG olduğu ve tüm katkı maddelerinin yumuşama noktası ve viskozite değerleri üzerinde benzer etkilere sahip olduğu tespit edildi. Buna ek olarak, ısı hassasiyetinin katkı kullanımı ile azaldığı ve en etkin katkının % 5 SBS olduğu saptandı.

Anahtar Kelimeler: Bitüm, Stiren-butadien-stiren, Amerikan gilsonit, Kıvam, Isı hassasiyeti.

1. Introduction

Bitumen obtained by distillation from crude oil is produced from the residues of this process. Bituminous binders are a complex mixture of organic particles containing aliphatic, aromatic and naphthenic hydrocarbons and could be divided into two basic groups, namely maltenes and asphaltenes [1]. The asphaltenes are dispersed in the maltene phase with a continuous phase and the structure that forms the bitumen structure includes saturates (S), aromatics (A), resins (R) and asphaltenes (AS). The complexity, aromaticity and molecular weight of the fractions are ranked as follows: S < A < R < As [2, 3].

The bitumen exhibits binding properties that binds aggregates due to their cohesive properties

in road applications. The bituminous binder also prevents the aggregate particles from falling apart under traffic loads, improves the driving comfort with the smooth surfaces they create, increases mixture stability with its cohesion, and provides impermeability by filling the voids in the mixture. Bituminous binders have a great effect on mixture performance, even though they are used at a low rate of 5 to 7% by weight in hot mix asphalts [4].

Various additives (modifiers) have been used to improve the properties of binders for a long time, thereby increasing the service life of the pavement by preserving its performance for a longer period of time. The additives used for this purpose vary depending usage. To increase the resistance of bitumens and bituminous hot mixes to heat and traffic loads, generally additive materials of polymer origin are added. Mostly Styrene-Butadiene-Styrene (SBS) Block Copolymers are used in these additives. Several previous studies determined that SBS improves the fracture resistance at low temperatures, and the generation of rutting and fatigue resistance at high temperatures [5-9].

Gilsonite is a solid hydrocarbon mineral available in nature with a potential to improve physical and chemical properties of bitumen [10]. Gilsonite can rapidly dissolve in bitumen since it is a kind of natural asphalt binder [11]. Gilsonite offers different advantages over other modifiers such as easy mixing and compatibility with asphalt mixtures [12]. Economically significant Gilsonite mineral is found in the United States and Iran.

When Gilsonite is added to the bitumen, the penetration of the bitumen decreases, its viscosity increases and as a result, a harder modified bitumen is obtained. Mixtures prepared with Gilsonite modified bitumens have higher stability, lower permanent deformation and temperature susceptibility, as well as higher resistance to stripping caused by water when compared to mixtures prepared with unmodified bitumen [13-16].

Several previous studies reported that storage stability of SBS modified bitumen is low, even though the rheological properties of SBS modified bitumen are superior to that of the neat binder [17, 18]. Furthermore, SBS is also a more expensive additive compared to Gilsonite. Using SBS and Gilsonite together in bitumen mixture instead of using only SBS-modified bitumen, so that the resulting performance would be the same, provides economic benefits [19]. The combined use of the two additives has been common during recent years and aimed to remove the negative aspects of the additives and to increase the rheological properties of the bituminous binders, and thus, increasing the performance of the BHMs in different aspects [20-23].

In the present study, the effect of the combined use of SBS and AG in bitumen modification on consistency and temperature susceptibility of the bituminous binders were examined. For this purpose, penetration, softening point and rotational viscometer experiments were conducted with neat and five different modified bitumen samples. Furthermore, using penetration and softening point values, penetration index value, which is the indicator of temperature susceptibility, is determined. Thus, the effect of two different additives on the consistency and thermal sensitivity of the bituminous binders was determined.

2. Material and Method

The binder design was conducted with the Superpave method in the study. Sanlıurfa, the warmest province in Turkey, was selected as the application field. The information on the highest temperature values for 7 consecutive days and the coldest day during the last 21 years was obtained from Regional Meteorology Directorate and it was assumed that the design traffic estimate was 3-30 million standard axle load equivalent and there was low-speed traffic (<50 km / Hour). It was determined that the binder class that should be used in the design criteria determined for Sanlıurfa was PG 76-10.

In the present study, Styrene-Butadiene-Styrene (SBS) Block Copolymer produced by Shell Bitumen Company and American Gilsonite (AG) obtained from American Gilsonite Company were used as additives to B 160/220 grade bitumen that was procured from TÜPRAŞ refinery in Turkey. The mix was obtained by mixing the neat bitumen and additive material for 60 minutes with 1000 rpm. mixer at a temperature of 180° C (Figure 1). In previous studies, it was determined that when 5% SBS (MB5S), 18% American Gilsonite (MB18G), 2% SBS + 13% AG (MB2S+13G), 3% SBS + 10% AG (MB3S+10G), and 4% SBS + %6 AG (MB4S+6G) were used, the performance level (PG 76-16) was suitable for Şanlıurfa province and the test results for these mixtures were similar. For this reason, 5 different additive ratios determined in the previous study were used.



Fig 1. Modified bitumen mixer and mixing apparatus

2.1. Peneteration Test

The penetration test was conducted to determine the stiffness or consistency of the bituminous binder. Penetration is the amount of vertical penetration by a standard needle at a given temperature under a given load, and over a period of time (Figure 2). The unit of penetration is 0.01 cm. In the penetration test, a load of 100 g was applied to the bituminous binder sample at 25°C for 5 seconds.

In the penetration test, the bituminous binder sample is taken according to EN 58 standard and transferred to the sample container after heating. The samples are allowed to cool for 60-90 minutes at a temperature of 5-30°C. The sample containers are then placed in a constant temperature water bath in the transfer chamber for 1-1.5 hours.



Fig 2. Penetration test apparatus

The transfer container that contains the sample container is placed on the penetration device plate. The needle loaded with the desired weight is adjusted to have contact with the sample surface. At least 3 tests should be conducted using points that are not closer to each other and the side of the container for more than 1 cm. The arithmetic mean of accepted measurements is rounded to the nearest whole number, which is taken as the penetration value.

2.2. Softening Point Test

The softening point test aims to measure the resistance of bituminous binders to high temperatures. Softening point is the temperature at the moment when the softened material touches the base as a result of heating the bituminous material in the standard ring at a certain speed, which was placed in a water bath, with a ball on it. The test setup is shown in Figure 3.

Before the experiment, the bitumen sample is heated and poured into the standard ring up to the top of the ring and cooled for 1 hour. The bitumen overflowing the ring is cut off using a heated spatula.



Fig 3. Softening point test apparatus

Pure water at a temperature of 5°C is placed in the beaker and the ring that contains

the sample is submerged into the water. Then the ball is placed in the beaker filled with water. The temperature of the water bath is kept constant at 5° C for 15 minutes. The ball is then placed in the middle of the sample in the ring using a pair of suitable tongs. The water is heated at a rate of 5° C per minute. The temperature read from the thermometer is recorded as the softening point when the softening material touches the base of the bath as a result of the increase in temperature.

2.3. Rotational Viscosimeter (RV) Test (ASTM D 4402)

The Rotational Viscometer (RV) test is conducted to determine the viscosity properties of the bituminous binders at high temperatures. The high temperature viscosity values of binders are identified to determine whether the binders are sufficiently fluid during pumping and mixing procedures. In the experiment, viscosity values are obtained by measuring the resistance of a shaft rotating at 20 rpm within the binder (Figure 4). In the RV test conducted with the original binders, it is best when the viscosity at 135°C does not exceed 3 Pa.s (3000 cP) [24, 25].

Approximately 30 gr. sample is taken and it is heated in a drying oven with a temperature of less than 150°C. Approximately 11 gr. of this material is placed in the sample compartment, and the sample compartment is placed in a temperature-controlled container with a constant temperature. After the sample is kept at a constant temperature for 15 minutes, the test is conducted. After almost equal viscosity values are obtained, three readings are conducted, and the viscosity of the binder is accepted as the average of these three measurements.

Viscosity values are used to determine the mixing and compaction temperatures of hot mix asphalts (HMAs). For this purpose, the RV test is carried out at 135°C and 165°C temperatures. A temperature-viscosity graph is plotted and the viscosity values are marked on the graph and these values are connected by a straight line. The viscosity values of the bituminous binder of 0.170 ± 0.20 Pa.s and a compaction value of 0.280 ± 0.30 Pa.s are desired when mixing HMAs [24]. The temperature values

corresponding to these viscosity values are accepted as the mixing and compaction temperatures.



Fig 4. Brookfield Viscosimeter

2.4.Temperature susceptibility of bituminous binders

2.4.1. Peneteration Index Method

Penetration Index (PI) is used to determine the temperature susceptibility of bituminous binders. The Penetration Index is determined with the standard penetration and softening point test results (Formulas 1, 2). P25 in the formula depicts the penetration value of the bitumen at 25°C and TYN indicates the softening point. The PI values decrease as the temperature susceptibility of bitumen binders increases. If the Penetration Index is less than -2, the bitumen is very sensitive to heat, whereas when it is greater than +2, it indicates that the bitumen is less sensitive to heat [26].

$$A = \frac{\log 800 - \log P_{25}}{T_{YN} - 25}$$
(1)

$$PI = \frac{20 - 500A}{1 + 50A}$$
(2)

2.4.2.Peneteration Viscosity Number Method

Penetration-Viscosity Number (PVN), also called Pen-Vis Number, is an empirical correlation between asphalt cement factors and low temperature pavement cracking experiences. Asphalt cement factors considered in the original correlation are penetrations at 25°C, viscosity at 135°C and equation is proposed for selecting asphalt cements to prevent low temperature cracking of asphalt concrete pavements. The PVN method is used to quantify temperature susceptibility of an asphalt cement and estimate its ability to prevent lowtemperature cracking. Lower values of PVN indicate higher temperature susceptibility, and asphalt mixtures containing binders with lower temperature susceptibility should be more resistant to cracking. The PVN number of a paving asphalt can be calculated precisely from the equations as follows [27];

$$PVN = -1.5 \frac{4.258 - 0.7967 \log P_{25} - \log V}{0.795 - 0.1858 \log P_{25}} \quad (3)$$

In the formula, P25 indicates the bitumen penetration at 25°C and V indicates the viscosity value at 135°C.

2.5. Experimental study

In the present study, neat and modified binders were subjected to penetration, softening point and rotational viscometer tests to determine their consistency. Furthermore, the penetration index values, which indicate the temperature susceptibility of the bituminous binders, are determined using softening point and penetration values.

3. Results

3.1. Peneteration results

The results obtained from the penetration tests applied to binders are presented in Figure 5.



Fig 5. Penetration test results

Figure 5 demonstrates that penetration values decreased with the use of additives. It was determined that the use of 18% AG and 5% SBS were the least effective methods for decreasing the penetration value when compared to the neat

bitumen. The penetration values increased as the SBS content increased when dual additives were used. The penetration values of the modified bitumen that contained 5% SBS, 18% AG, 2% SBS + 13% AG, 3% SBS + 10% AG and 4% SBS + 6% AG were 3.14, 7.50, 6.32, 5.34 and 4.56 times lower than neat binder, respectively.

3.2. Softening Point Test results

The results obtained in the softening point tests applied to the binders are presented in Figure 6. Figure 6 demonstrates that the softening point values increased with the use of additives. This indicated that all utilized additives would increase the high temperature resistance of the bituminous binders. When the impact of the additives on the softening point values is considered, it was found that all the additives had similar effects on the softening point and increased the softening point values by about 45% when compared to the neat binder.

80 80 00 00 00 00 00 00 00 00 00 00 00 0						
	PG 52-28	MB5S	MB18G I	MB2S+13G	MB3S+10G	MB4S+6G

Fig 6. Softening point test results

3.3. Specific gravity of bituminous binders

Specific gravity values of neat and modified bitumen are determined based on the EN 15326 standard. The results obtained in specific gravity tests are presented in Figure 7.

1,04 1,038 1,036 1,034 1,032 1,034 1,032 1,034 1,032 1,034 1,032 1,034 1,034 1,034 1,034 1,036 1,034 1,036 1,034 1,036 1,034 1,036 1,036 1,036 1,034 1,036 1,034 1,036 1,034 1,036 1,034 1,036 1,034 1,036 1,034 1,036 1,034 1,036 1,034 1,036 1,034 1,036 1,034 1,036 1,034 1,026 1,0	MB18G ME2S+13GME3S+10GME4S+6G
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Fig 7. Specific gravity values of neat and modified binders

As can be seen in the figure above, the specific gravity values of bituminous binders

were between the specification limits 1.0-1.1. When the changes in specific gravity with the use of additives were examined, it was observed that the specific gravity of binders increased with the use of American Gilsonite, whereas the specific gravity values decreased with the use of SBS. With the combined use of AG and SBS, specific gravity values increased regularly when compared to the use of 5% SBS, but decreased regularly when compared to 18% AG use.

3.4. Penetration Index values

The Penetration Index values for neat and modified bituminous binders determined using formulas (1) and (2) are given in Figure 8.



Fig 8. Variation in binder penetration index values with additive use

The Penetration Index results that demonstrate the temperature susceptibility of bituminous binders showed that the PI value increased as the SBS ratio in modified binders increased (Figure 8). Since the temperature susceptibility of bituminous binders decrease as the PI value increased, it could be argued that the decreased the temperature use of SBS susceptibility of bituminous binders. It was determined that as AG content in modified bitumen increases, the PI values decreases and thus, the temperature susceptibility increases. When SBS and AG are used together in the modified bitumen, it was determined that AG had a negative effect on the temperature susceptibility, and that this problem was eliminated by using SBS in conjunction with AG.

3.5. Penetration Viscosity Number values

The Penetration Viscosity Index values for the neat and modified bituminous binders determined using the formula (3) are given in Figure 9.



Fig 9. Variations in binder penetration viscosity number values with additive use

When the penetration viscosity number results that reflect the susceptibility of bituminous binders to low temperatures were evaluated, it was determined that the use of AG had a negative effect on temperature susceptibility in PVN values, similar to the PI values, when compared to that of the SBS. It was also determined that SBS can be used to avoid the negative impact of AG on temperature susceptibility. It was determined that the temperature susceptibility decreased with the use of SBS instead of AG in dual modified bitumen (Figure 9).

3.6. Rotational Viscometer Test results

Neat and modified binders were subjected to rotational viscometer tests at 135° C and 165° C temperatures. In the experiment, 20 rpm speed was used. Modification index values were obtained from the ratio of the viscosity value of the modified binder to the viscosity value of the neat binder (nmodified / nneat). The viscosity results obtained from the tests are presented in Figure 10. The viscosity and modification index values obtained at high temperatures (135° C and 165° C) demonstrated that SBS increases the stiffness of the binders and reduces the workability at high temperatures.



Fig 10. Variations in binder viscosity values with temperature and additive use

Figure 10 demonstrates that the viscosity values of all binders met the Superpave specification limit maximum value of 3000 cP at a temperature of 135oC. With additive use, the viscosity values increased at all temperatures. It was determined that the viscosity values of 5% SBS modified binders were the highest and the bituminous viscosity values with 18% AG were the lowest, despite their similar performance levels. It was determined that as the SBS content in the modified bitumen increased, the viscosity values increased at both temperatures.

The modification index (nmodified / nneat) values determined by the ratio of the viscosity value of the modified bitumen to the viscosity value of the neat binder are given in Figure 11. When the modification index values were examined, it was determined that the additive that maximizes the viscosity value at all temperatures was SBS and the least contributing additive was AG. It was determined that the viscosity values of SBS and AG were higher than those of the sole use of AG and the viscosity values increased regularly as the SBS ratio in the mixture increased.

When the bituminous binders are mixed with the aggregates at the plant and the hot mix asphalts are compacted in the field, they must possess adequate workability. It is desirable that the bituminous binder has a viscosity value of 0.170 \pm 0.20 Pa.s for compaction and 0.280 \pm 0.30 Pa.s for the mixing [24]. In the plotted temperature-viscosity graphs, the viscosity values were marked and these values were joined with a straight line. The temperature values that correspond to these viscosity values are accepted as the mixing and compaction temperatures. As an example, the viscosity-temperature graph that shows the mixing and compaction temperature ranges for the modifier binder that contains 18% AG is presented in Figure 12.



Fig 11. Variations in binder modification indexes with temperature and additive use



Fig 12. Viscosity-temperature chart for the modified binder including 18% AG

The variation in the mixing and compaction temperatures with the type of additive is presented in Figure 13.



Fig 12. Mixing and compaction temperatures of binders

As shown in Figure 13, higher mixing and compaction temperatures were required with the use of additives. This demonstrates that although higher binder performance is achieved with additive use, higher temperatures will be required during mixing it with the aggregate and compaction of the HMA, thus requiring more energy during the preparation at the plant. When the effects of the additive types and combined use of additives were evaluated, it was determined that the highest mixing and

compaction temperatures were found with 5% SBS modified bitumen and the lowest values were found with 18% AG modified bitumen. As the SBS content in the modified bitumen increased, higher mixing and compaction temperatures were needed.

4. Conclusion

In the study, consistency and specific gravities of neat and 5 different modified bitumen were determined and their temperature susceptibility was evaluated with two different methods. Initially, 18% AG and 5% SBS were used as additives separately, then 2% SBS and 13% AG, 3% SBS and 10% AG, and 6% AG and 4% SBS were used in conjunction to obtain bituminous binders with the same performance level.

• Penetration tests demonstrated that all additives increased the stiffness significantly. It was determined that the most effective additive was 18% AG and the least effective additive was 5% SBS.

• Softening point tests demonstrated that all additives increased the high temperature resistance of the bituminous binders significantly, and all modified bitumen had similar softening point values.

• It was determined that the specific gravity values of AG modified bitumen were higher since both the usage ratio and density of AG was higher than those of SBS as demonstrated in specific gravity tests.

• Penetration index values demonstrated that 5% SBS modified bitumen had the lowest thermal sensitivity and 18% AG modified had the highest. This negative aspect of AG modified binders was reduced with the addition of SBS.

• Penetration viscosity number values were consistent with PI values. With the use of AG additive, the low temperature susceptibility of bituminous binders have increased, and the use of SBS has reduced the low temperature susceptibility.

• It was determined that additive use increased binder stiffness values as observed in viscosity values. Consequently, the mixing and compaction temperatures increased with the modification values. The most effective additive on viscosity values was 5% SBS modification.

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