



## Research Article

# Comparison of the conventional-rheological properties of Iraq gilsonite and SBS modified bitumen

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## ABSTRACT

Styrene-butadiene-styrene (SBS) is widely used to improve the properties of the base bitumen and hence bituminous mixtures. As SBS modification induces cost increase, natural asphalts have become an important issue. In this study, the effects of gilsonite supplied from Iraq, a type of natural asphalt (NA) on conventional and rheological properties of bitumen, were investigated. NA was used 20%, 35%, and 50% by weight of bitumen. The properties of NA modified bitumen were compared to 2%, 3%, and 4% SBS modified bitumen. Conventional (penetration, softening point, rotational viscosity) and rheological (dynamic shear rheometer, bending beam rheometer) binder tests were conducted on base and modified bitumen. The index values representing together the workability, high and low temperature properties were defined by dividing the rutting parameter to viscosity and rutting parameter to low temperature stiffness. Overall, the test results show that 20% NA modified B160/220 bitumen exhibited similar performance with B160/220+3%SBS modification produced at 16% higher cost in terms of the high and low temperature properties.

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## INTRODUCTION

Bituminous binders are mostly modified with polymers and other additives such as polypropylene, crumb rubber, and steel, to resist heavy traffic and the adverse effects of the environment. Styrene-butadiene-styrene (SBS), the most used additive, have been found to be effective in preventing temperature and traffic-based deterioration [1–8]. The breaking of the polymer network after aging [9] and the high cost of polymers have brought the use of alternative and natural asphalt to the agenda.

Natural asphalts are solid or semi-solid materials containing hydrocarbons and aromatic molecules. Natural asphalts consist generally of carbon and hydrogen, they also contain nitrogen, sulfur as well as a small number of metals such as iron, nickel, and vanadium [10]. Gilsonite and Trinidad lake asphalt (TLA) are the most common types of natural asphalt used to improve the properties of bitumen and bituminous hot mixtures [11].

TLA increased the softening point while lowering the ductility and penetration value of the binder. In addition,

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TLA has developed a high degree of stability and rutting durability for base binder [12]. Huang and Xu have evaluated the resistance to permanent deformation of Iranian rock asphalt, Buton rock asphalt, Trinidad Lake asphalt, and SBS modified mixtures. It was determined that the most important contribution was that of Iranian rock asphalt and the least contribution was that of Trinidad Lake asphalt [13]. The compound of TLA and SBS exhibits a remarkable combined effect on enhancing the deformation and aging resistance of asphalt [14]. According to the thin film oven (TFO), pressure aging vessel (PAV), and ultraviolet (UV) aging procedures, TLA modified bitumen found to have improved anti-aging properties [15].

It was found that QC (Qingchuan) rock asphalt has shown better results in terms of high temperature resistance. The use of 6% QC rock asphalt and 1% silica appeared to be optimal [16]. In terms of balance between performance and polarity, 30% button rock asphalt has been found to be the optimum value [17]. The rock asphalt additive improves the tensile stress, moisture damage, fatigue performance of mixtures and high temperature performance of the binder. However, low temperature performance was observed to worsen [18]. Buton rock asphalt was found to increase the dynamic viscosity and shear modulus at 60°C. It also showed better rutting resistance [19]. Buton rock asphalt can behave as a stabilizer and improve the performance of stone mastic asphalt [20]. In the study where asphaltite was used as filler in a bituminous hot mixture, it was stated that 3% asphaltite content was the optimum value, and the fatigue life of these mixtures increased significantly [21].

Aflaki and Tabatabaee (2009) evaluated the performance criteria of the gilsonite modified bitumen 2%, 4%, 7%, 10% and 13% by weight. As the gilsonite ratio increased, the viscosity, softening point, and complex modulus of the binders have increased while the phase angle and penetration of the binders have decreased [22]. Gilsonite modified samples provided the best fatigue resistance. However, the mixtures containing the base binder showed higher strength than the gilsonite modified mixtures when the strength of the mixtures after crack formation was examined [23]. Although gilsonite has improved the high temperature performance of bituminous binders, the low temperature performance has worsened with increasing gilsonite content in the modified bitumen [24–27]. It was determined that 2% SBS with 13% AG (American gilsonite), 3% SBS with 10% AG, and 4% SBS with 6% AG were required to obtain the same performance level binders [28]. Gilsonite, a natural asphalt, showed better storage stability than SBS. [29]. With the increase of the gilsonite ratio, the rutting parameter was increased but remained low compared to that of the SBS modified bitumen. Gilsonite has been proposed to be used as an alternative to reduce the cost of the asphalt mixture [30]. Gilsonite-containing mixtures had greater resistance to moisture damage. Mixtures containing 10% gilsonite

gave the highest elastic modulus values and provided an important increase in the rutting resistance of mixtures [31]. The thermomechanical and durability properties of the mixtures enhanced by using gilsonite along with warm mix additive [32]. Gilsonite was found to have a great effect to improve temperature sensibility. 30 wt% gilsonite with 7.5 wt% styrene-butadiene-rubber was recommended for an improved high and low temperature performance [33]. The use of 4%, 8%, and 12% gilsonite made the PG 58-22 neat binder as PG 64-22, PG 70-16, and PG 76-16, respectively. While all gilsonite contents enhanced the high temperature performance grade, the high usage induces a slight decrease in low temperature performance grade [34].

In this study, Iraq gilsonite, which is 20% cheaper than the original refinery bitumen, was used as a modifier. This study aims to determine the effect of Iraq natural asphalt on base binder properties and also whether the use of natural asphalt with the base binder is an alternative material to SBS or not in terms of low and high temperature performance.

## MATERIAL AND METHODS

### Materials

160/220 penetration grade bitumen obtained by Turkish Petroleum Refineries was used as the base binder. The binder was modified by 20%, 35%, 50% natural asphalt (gilsonite) and by 2%, 3%, 4% SBS by weight of bitumen to compare the natural asphalt modification with SBS modification. Natural asphalt (Figure 1) was supplied from Iraq. Elemental analyses of base asphalt and natural asphalt are given in Table 1. Natural asphalt is not so different from base bitumen in terms of elemental content. The used SBS the properties of which are given in Table 2 was Kraton D-1101 and supplied by Shell Chemicals Company. The binders were modified by adding the additive to the base

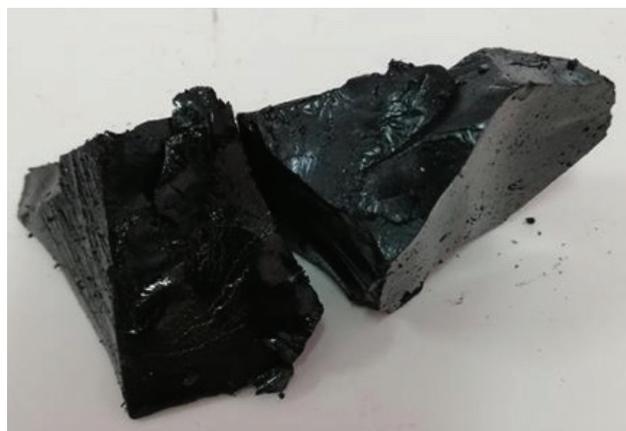
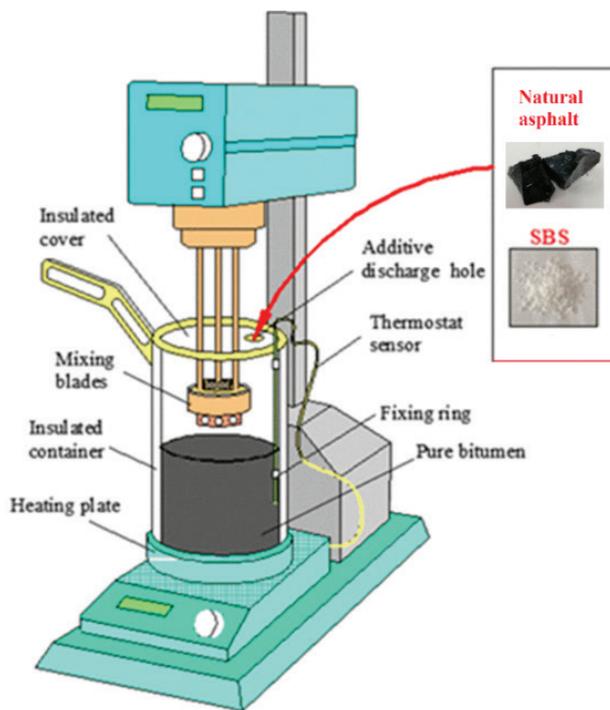


Figure 1. Natural asphalt (Iraq gilsonite).

**Table 1.** Elemental analysis of base and natural asphalt

Binders	C (%)	H (%)	N (%)	S (%)	Solubility in TCE (%)
Base Bitumen	82.83	8.96	0.604	7.227	99.21
Natural asphalt	81.52	10.82	0.455	5.914	97.95

**Figure 2.** Modified bitumen preparation system.

bitumen at different contents and then by mixing at 1000 rpm at 180°C for 1 h in a laboratory-type mixer (Figure 2).

Base and modified bitumen were subjected to penetration, softening point, rotational viscosity, dynamic shear rheometer, and bending beam rheometer tests. Natural asphalt modified bitumen are represented by 20NA, 35NA, and 50NA for the binders modified by 20%, 35%, and 50% natural asphalt by weight of base bitumen, respectively. SBS modified bitumen represented by 2SBS, 3SBS, and 4SBS for the binders modified by 2%, 3%, and 4% SBS by weight of base bitumen, respectively. The experimental program is given in Figure 3. The approximate costs of the binder are given in Table 3. The fact that the price of natural asphalt is lower than that of refinery bitumen has made the price of bitumen with NA added even cheaper than bitumen without additives. 35NA bitumen can be produced 17% cheaper than 3SBS bitumen and 21% cheaper than 4SBS. Addition of NA to bitumen can be done in bitumen tanks with mixing equipment and does not need an additional modified bitumen plant. On the other hand there should be an additional bitumen blending equipment (bitumen modified plant) in the SBS modification, it is clear that the NA modification will be much more economical.

**Table 2.** The physical characteristics of SBS

Styrene Double Block	Tensile Strength (MPa)	Supported Hardness A	Melt Flow Rate (g/10min)	Volatile Matter (%)	Solubility in TCE (%)
14-18	> 4	28-38	8-12	< 0.7	< 0.2

**Table 3.** The cost of the binders

Binder	Base bitumen (kg)	NA (kg)	SBS (kg)	Total cost (€)	Total weight (kg)	Unit cost (€/ton)
20NA	1000	200	-	605	1200	504
35NA	1000	350	-	667	1350	494
50NA	1000	500	-	730	1500	486
2SBS	1000	-	20	585	1020	573
3SBS	1000	-	30	617	1030	599
4SBS	1000	-	40	649	1040	624
Base bitumen (B 160/220)						522
NA						417
SBS						3196

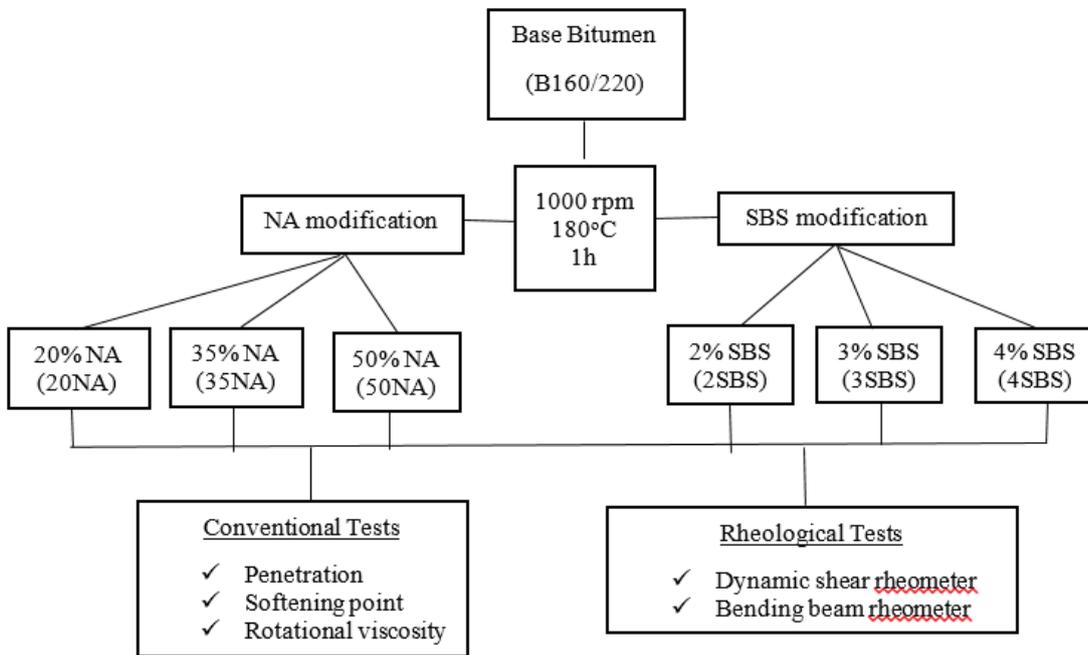


Figure 3. The experimental program.

TEST AND RESULTS

Softening Point and Penetration Test

The softening point test was conducted according to ASTM D36. The softening points of base and NA modified bitumen are given in Figure 4. The softening point values of SBS modified bitumen are also seen as threshold values in the figure. Natural asphalt modification has a great effect on the softening point. 20%, 35% and 50%NA modification increase the softening point of the base binder as 15%, 28%, and 66%, respectively. 2%, 3%, and 4% SBS modification increase the softening point of the base binder as 10%, 22%,

and 29%, respectively. 50%NA modified bitumen gives 15.3°C higher softening point than that of the 4% SBS modification produced at 28% higher cost. The softening point of 2%, 3%, and 4%SBS modification can be obtained by 17%, 29%, and 33% natural asphalt modification, respectively.

Bitumen begins to flow at its softening point temperature. Hence, if the softening point of a particular bitumen is higher than the pavement temperature, it can be said that the bituminous mixtures can well resist permanent deformations. The binder with a high softening point designates more high temperature resistance of bituminous mixtures in which it is used. 50%NA modified bitumen exhibited

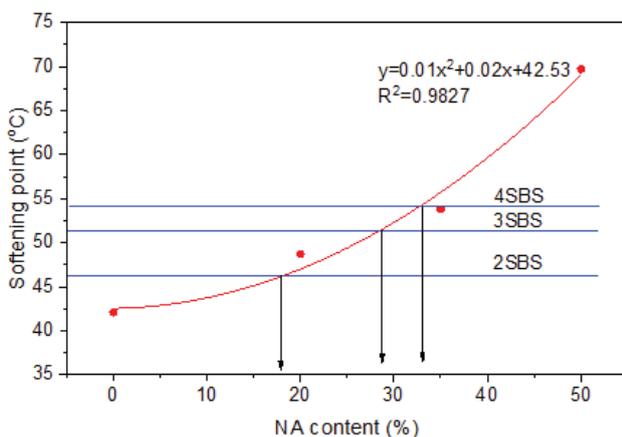


Figure 4. Softening points of binders.

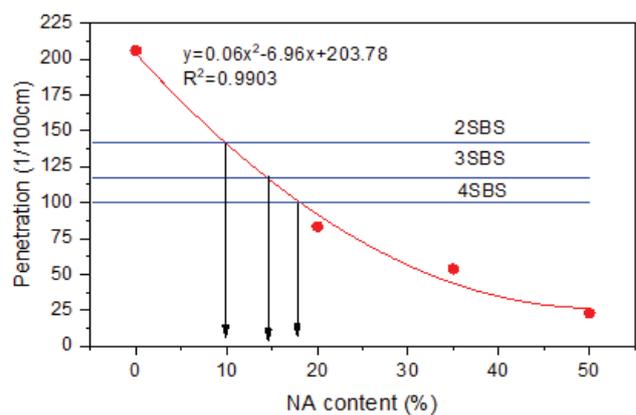


Figure 5. Penetration values of binders.

better performance among the binders indicating a good resistance to permanent deformation.

The penetration test was conducted according to ASTM D5. The results of the penetration values are given in Figure 5. NA induces a significant reduction in penetration values. NA has a stiffening effect on the base binder. SBS modification does not have as stiffening effect as the NA modification. 10%, 15%, and 18% NA modified bitumen have the same penetration values with 2%, 3%, and 4% SBS modified bitumen, respectively.

The penetration index determined by using softening point ( $T_{RB}$ ) and penetration values ( $Pen_{25}$ ) represents the temperature sensitivity of bitumen. The penetration index can be accepted as a measurement of consistency for moderate temperatures (20-40°C). The lower the penetration index the faster the binder changes its consistency as the temperature changes [35]. The penetration index (PI) is determined by the following formula;

$$PI = \frac{1952 - 500 \cdot \log(Pen_{25}) - 20 \cdot T_{RB}}{50 \cdot \log(Pen_{25}) - T_{RB} - 120} \quad (1)$$

The PI values of binders are given in Figure 6. Base binder has the same PI values with the 50% NA modified bitumen. The NA modification is more efficient on softening point than penetration. The softening point continuously increases with the increase of NA content, however, the penetration decrease with the NA content is not pronounced at higher NA content. This difference increases at high NA content on behalf of softening point values which reveals the increasing of PI at 50%NA content. While the PI values of base and NA modified bitumen remain between -1 and +1 indicating a moderate temperature sensitivity, SBS exhibits low temperature sensitivity compared to NA modification with the PI values higher than +1. 3%SBS modification exhibited better temperature sensitivity than the 4%SBS modification.

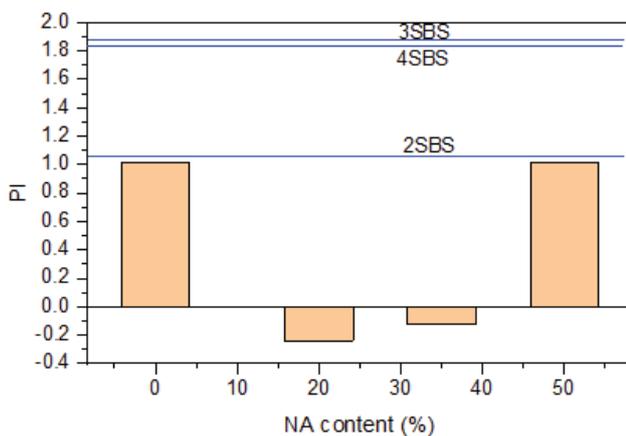


Figure 6. PI values of binders.

### Rotational Viscosity Test

The viscosity of binders at 135°C were determined by a Brookfield DV-III rotational viscometer. The rotation of the spindle is selected as 20 rpm. The test was conducted according to ASTM D4402. The asphalt viscosity should be lower than 3.0 Pa.s at 135°C to provide good workability, in other words, easy handling of hot mix asphalt during manufacturing and construction [36]. The variation of viscosities are given in Figure 7. The viscosity of base bitumen increases significantly with the increase of NA content. Nevertheless, the 3000 cP viscosity value is not exceeded even at the highest NA content. Therefore, there is no workability problem by using 50%NA with base bitumen.

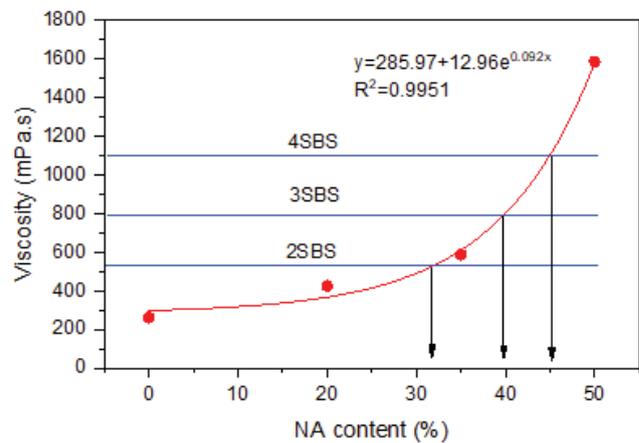


Figure 7. Viscosities of the binders.

The viscosity increase is observed more after 35%NA content. The viscosity values increase 1.62, 2.24, and 6.06 times compared to base binder' viscosity by using 20%, 35%, and 50%NA, respectively. The viscosity values of 2%SBS, 3%SBS, and 4%SBS can be obtained by 32%NA, 40%NA, and 45%NA. It is concluded that the usage of 50%NA with base binder will be able to exhibit better or the same performance compared to 4%SBS modification in terms of resistance to permanent deformation without any workability problem.

### Dynamic Shear Rheometer (DSR) Test

The DSR test was conducted using a Bohlin DSR II rheometer for unaged binders under the controlled-stress conditions at 64°C according to ASTM D7175 standard. The test was performed using a 25 mm diameter plate, and a 1 mm gap opening at 1.59 Hz frequency. The rutting parameters ( $G^*/\sin\delta$ ) and phase angles were determined to evaluate the high-temperature performance and also to compare the elastic components of the SBS and NA modified bitumen. The variation on the rutting parameter by NA content is given in Figure 8. Rutting parameters increase

exponentially with the increase of NA content. 20%, 35%, and 50%NA modified bitumen induce 2.9, 5.7, and 21 times higher rutting parameters compared to the base bitumen, respectively. The increment effect is observed more above 35%NA content. The mixtures produced by 50%NA modified bitumen will have a superior performance in terms of resistance to permanent deformation at high temperature under the influence of heavy traffic load. It is clear that the bituminous mixtures with 35%NA modified bitumen will be better than the mixtures produced by 4%SBS modified bitumen in terms of endurance to plastic deformations. The rutting parameters linearly increase with the additive content in SBS modification. 2%, 3%, and 4% SBS modified bitumen have 2, 3.2, and 4.9 times higher  $G^*/\sin\delta$  value than that of the base bitumen, respectively. 18%, 25%, and 32% NA are required in order to obtain the same  $G^*/\sin\delta$  values with the 2%, 3%, and 4% SBS modification respectively. In order to obtain similar performance with the SBS modification, the NA ratio should be used approximately 8 times higher than the SBS content.

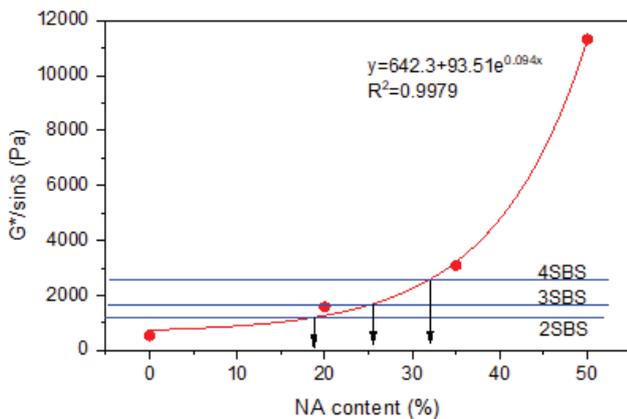


Figure 8.  $G^*/\sin\delta$  values of binders at 64°C.

The variation of phase angles ( $\delta$ ) with the additive content is given in Figure 9. There is a linear relationship between the phase angles and NA content. 20%, 35%, and 50%NA modification give 6%, 10.7%, and 17.9% lower phase angle than that of the base bitumen. 20%NA modification has a lower phase angle than 2%SBS modification, 35%NA modification has a lower phase angle than 3%SBS modification, and 50%NA modification has a lower phase angle than 4%SBS modification. The required NA contents in order to provide the same phase angles with 2%, 3%, and 4%SBS are ranged as 17%, 27.7%, and 38.4%, respectively.

In order to make a rational comparison considering the phase angles with the rutting parameter, the variation on rutting parameter versus phase angles is shown in Figure 10. There is an exponential relation between the  $G^*/\sin\delta$  and phase angles for either modification. The phase angles

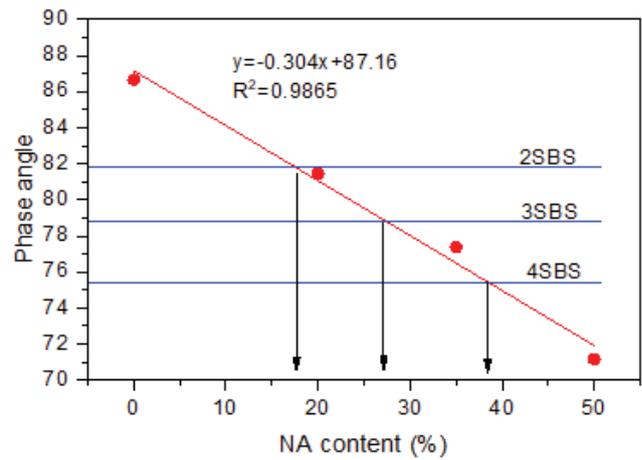


Figure 9. Phase angles of binders at 64°C.

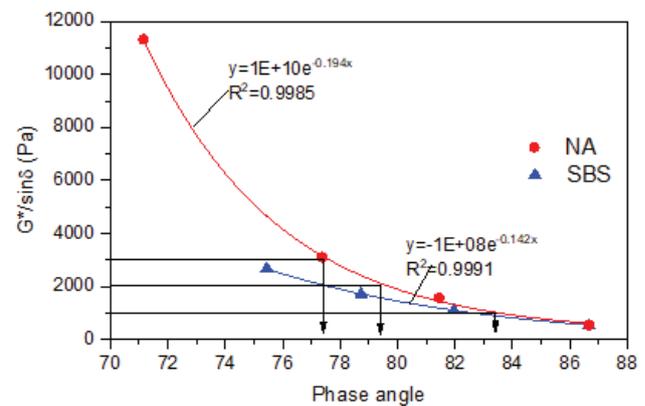


Figure 10. The variation of  $G^*/\sin\delta$  versus phase angles.

at the same  $G^*/\sin\delta$  values such as 1000, 2000, and 3000 Pa are seen in the figure.

Although NA modification has a significant increase in rutting parameters, it is clear that SBS modification has lower phase angles than NA modification at any constant  $G^*/\sin\delta$  value. It was determined that SBS modified bitumen will exhibit a bit more elastic behavior than NA modified bitumen independently from additive content. The differences between the phase angles of NA modified bitumen and SBS modified bitumen rise with the increase of  $G^*/\sin\delta$  value. The high amount of SBS modification which results in a high rutting parameter is potentially more useful in resisting deformation than the mixtures containing a high amount of NA.

### Bending Beam Rheometer (BBR) Test

As a thermoplastic material, bituminous binders tend to contract at low temperatures. The difference between the thermal expansion properties of bituminous binder and aggregate induce internal stresses during the surrounding

temperature drop. A single severe drop above the critical temperature or repeated thermal contraction can cause thermal cracking without the effects of traffic loading [37]. The low temperature stiffness ( $S_t$ ) and relaxation properties ( $m$ -value) of bituminous binders are determined from the BBR test. These parameters represent the ability of bituminous mixtures to resist low temperature cracking. The creep stiffness of an asphalt beam sample at any time of loading ( $t$ ) is determined by;

$$S_t = PL^3 / (4bh^3 \delta_t) \quad (2)$$

Where  $S_t$  is creep stiffness (MPa),  $P$  is applied constant load (N),  $L$  is span length of beam sample (102 mm),  $b$  is beam width (12.7 mm),  $h$  is beam thickness (6.35 mm), and  $\delta_t$  is deflection (mm) at time  $t$ . The test was conducted at  $-10^\circ\text{C}$ ,  $-15^\circ\text{C}$ , and  $-20^\circ\text{C}$  according to ASTM D6648 standard. The variations on creep stiffness are given in Figure 11.

It is seen that the relation between the creep stiffness and additive content is exponential for NA modification and linear for SBS modification. NA modification is more effective in stiffening the bitumen than SBS modification at all temperatures. The increase in hardening by the increase of the additive content is observed more at lower temperatures

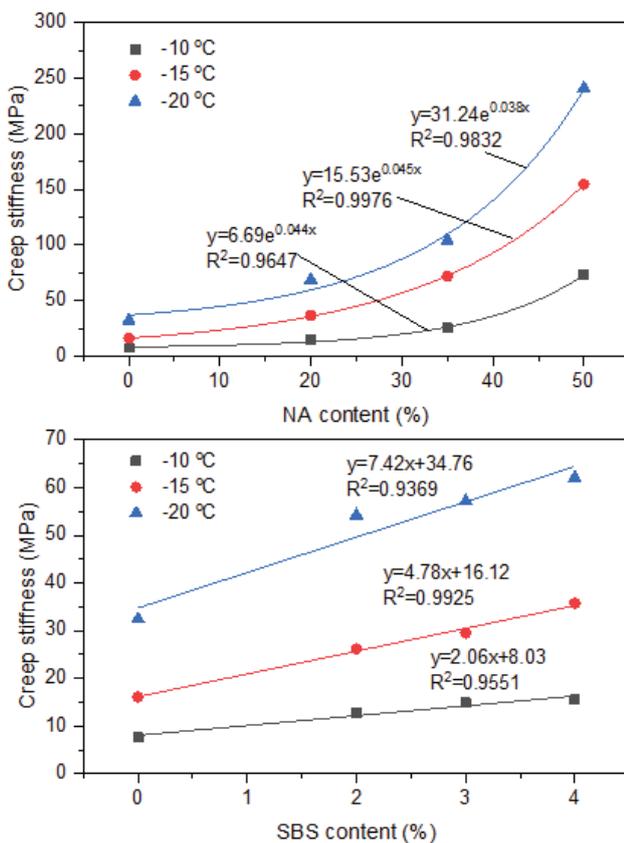


Figure 11. The variation on creep stiffness of binders.

for both types of modification. SBS content is also more effective in increasing the stiffness while the temperature is decreasing. 20%, 35% and 50% NA modification induce 2.1, 3.2, and 7.4 times higher stiffness value compared to that of the base bitumen at  $-20^\circ\text{C}$ . The stiffness values of 2%, 3%, and 4% SBS modified bitumen are 1.6, 1.8, and 1.9 times higher than base bitumen at  $-20^\circ\text{C}$ , respectively. 20% NA modified bitumen gives close stiffness values to the 4% SBS modification at  $-10^\circ\text{C}$ ,  $-15^\circ\text{C}$ , and  $-20^\circ\text{C}$ .

The flexible behavior at low temperature requires low creep stiffness with high creep ratio ( $m$ -value) values. Hence, the “ $\lambda$ ” value is obtained by dividing the creep stiffness to  $m$ -value for a more realistic evaluation [38]. The small value of  $\lambda$  is desirable for elastic behavior at low temperatures. It is obvious that among the binders which are in same stiffness values, the one with high  $m$ -value will be preferred. Increasing the relaxation has a more significant effect on reducing the low temperature cracking than the decreasing the stiffness. Hence,  $\lambda$  values enable to compare the binders in terms of relaxation properties at a given stiffness and at a specific temperature.  $\lambda$  values provide a relationship between stiffness and relaxation which is needed to assess the non-load related cracking. The variations of  $\lambda$  values are given in Figure 12.  $\lambda$  values increase with the increase of additive content at NA and SBS modification. However, the increasing trend is much more at NA modification compared to SBS modification.  $S_t/m$ -value ratio of 20% NA modified bitumen is close to that of the 4% SBS modified bitumen. The stiffness of the NA modified bitumen significantly increase above 20% NA content. 50% NA modification exhibits the worst low temperature behavior by having 5 times higher  $\lambda$  values than 4% SBS modification at all temperatures. It is clear that the bituminous mixtures prepared by the binders modified with much more than 20% NA content will not well resist low temperature cracks compared to any of the SBS modifications.

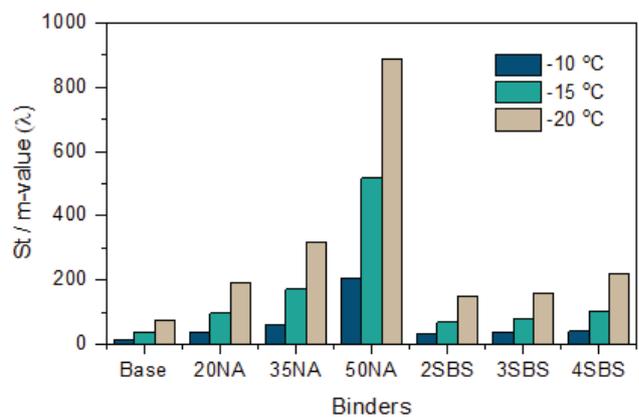


Figure 12. The variation on  $\lambda$  values at different temperatures.

### Combined Evaluation of the Conventional and Rheological Tests

The fluidity properties, resistance to rutting, and resistance to low temperature cracks of binders determined by rotational viscometer, DSR and BBR tests respectively were evaluated together in order to make a comprehensive decision on the performance of binders and to make more a realistic comparison. The viscosity value greatly affects the ability of bitumen to spread, penetrate into the voids of bituminous mixtures and cover the aggregates. High viscosity adversely affects the workability of the mixtures and causes the bituminous mixtures to be produced and laid at high temperatures. Hence, an evaluation considering both rutting parameter and viscosity value together seems rational. While a high rutting parameter is desired for good resistance to permanent deformation at high temperatures, low viscosity is required for a good workability property. Therefore,  $I_1$  index value was defined by dividing the rutting parameter determined at 64°C to viscosity determined at 135°C. Higher  $I_1$  values represent an enhanced high temperature performance with a good workability property. Note that the viscosity values must not be higher than 3.Pa.s at 135°C in order to evaluate the  $I_1$  values in the light of the interpretation denoted above. Any of the viscosities at 135°C do not exceed this limit value in this study. The variations of  $I_1$  values are given in Figure 13.  $I_1$  values increase linearly with the increase of additive content for two types of modification. The binders modified with NA content undertaken in this study has a much more positive effect on  $I_1$  values than %2, 3%, and 4% SBS modification. While rutting parameters increase significantly at NA modification, the viscosities do not increase as it is in the SBS modification with the increase of additive content. The  $I_1$  values of 20NA binder are 84%, 66%, and 52% higher than that of the 2%, 3% and 4% SBS modified bitumen respectively. The mixtures prepared by the lowest content of NA used in this study have more potential ability to carry the heavy loads without any workability problem during its production compared to the highest content of SBS modification.

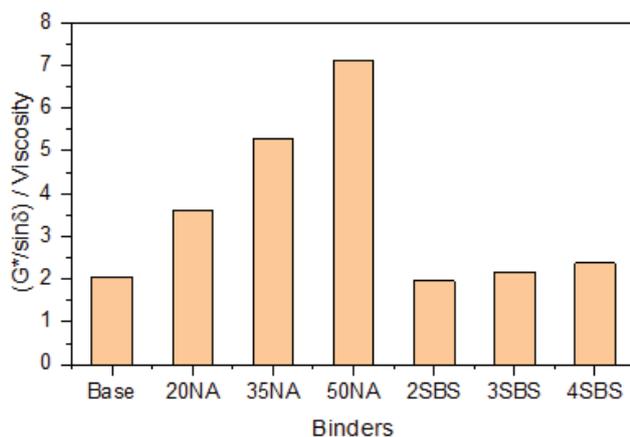


Figure 13. The variation of  $I_1$  values of binders.

Another evaluation can be done by considering the high and low temperature behavior together.  $\lambda$  values determined before by dividing the creep stiffness to m-value is a good indicator of load dissipation ability at low temperatures. The bituminous mixtures are desired to be strong enough at high temperatures and exhibit flexible behavior at low temperatures. Therefore,  $I_2$  index value was defined by dividing the rutting parameter to  $\lambda$  values. As comparing the  $I_2$  values of different types of binders it is clear that higher  $I_2$  values stand for improved resistance to permanent deformations as well as improved thermal cracking resistance. The variation of  $I_2$  values calculated by using  $G^*/\sin\delta$  at 64°C and  $\lambda$  values at -20°C are given in Figure 14.  $I_2$  values increase with the increase of additive content for either modification indicating an improved high and low temperature performance. 20NA binder exhibits better performance than the 2SBS binder. 50NA binder has the highest ( $I_2$ ) value by an increment of 80% compared to base bitumen. 35NA binder gives close values to the 3SBS binder.

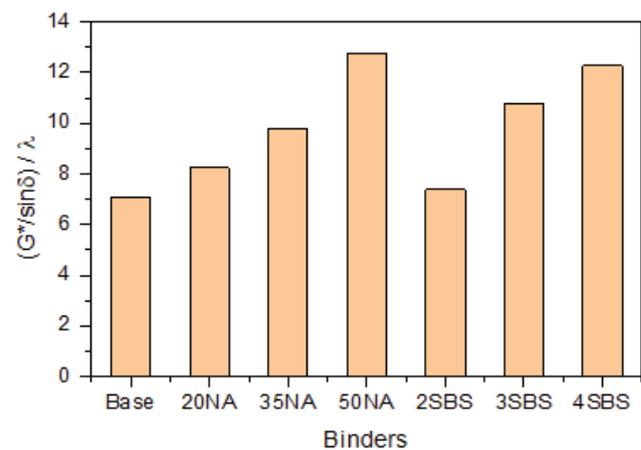


Figure 14. The variation on  $I_2$  values of binders.

### CONCLUSION

In this study, the performance of natural asphalt modified bitumen was investigated and compared to SBS modification in terms of conventional and rheological properties. Based on the experimental study, the following conclusions could be drawn:

Natural asphalt (NA) modification has a great effect on increasing the softening point of base binder. 50% NA modified bitumen has a higher softening point value than 3% SBS modification. While penetration index values of base and NA modified bitumen remain between -1 and +1 indicating a moderate temperature sensitivity, SBS exhibits lower temperature sensitivity compared to NA modification with the PI values higher than +1. According to the viscosity test, it was concluded that the 3000 cP viscosity value is not exceeded even at the 50% NA content. The 20NA and

35NA binders will contribute to the natural environment and human health, as they will provide lower mixing and compaction temperatures in parallel with lower viscosity values than the 3SBS modification, which is frequently used in asphalt road applications.

Rutting parameters increase exponentially in NA modification and linearly in SBS modification with the increase of additive content. 18%, 25%, and 32% NA are required in order to obtain the same  $G^*/\sin\delta$  values with the 2%, 3%, and 4% SBS modification, respectively. It was determined that SBS modified bitumen will exhibit a bit more elastic behavior than NA modified bitumen independently from additive content.

NA modification is more effective in stiffening the bitumen than SBS modification at low temperatures. 20% NA modified bitumen gave close stiffness values to the 4% SBS modification at  $-10^{\circ}\text{C}$ ,  $-15^{\circ}\text{C}$  and  $-20^{\circ}\text{C}$ . According to creep stiffness/m-value ratios which is a good indicator for evaluating the low temperature behavior, it was determined that the bituminous mixtures prepared by the binders modified with much more than 20% NA content will not well resist to low temperature cracks compared to any of the SBS modification.

Two types of index values, one of which is defined by dividing the rutting parameter to viscosity and the other is defined by dividing the rutting parameter to creep stiffness/m-value, were used to evaluate the workability, high and low temperature behavior together. It was determined from the index values that the mixtures prepared by the 20% NA have more potential ability to carry the heavy loads at high temperatures without any workability problem during its production compared to 4% SBS modification. 35% NA modified bitumen seems to exhibit similar performance with 3% SBS modification produced at 21% higher cost in the case of considering together the high and low temperature properties. However, the multi stress creep recovery test and life cycle assessment must be performed to NA and SBS modified bitumen in order to accurately compare the elastic behavior and efficiency of the binders and mixtures in further studies.

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