EFFECTS OF NATURAL ASPHALTS ADDING ON THE RHEOLOGICAL CHARACTERISTICS OF PAVING GRADE BINDERS

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ABSTRACT

This paper presents a laboratory-scale evaluation of the conventional and the fundamental rheological characteristics of modified binders containing natural asphalts such as gilsonite, asphaltite and Syrian natural asphalt. Bitumen samples that are modified with natural asphalt were produced by mixing base bitumen with gilsonite, asphaltite and Syrian natural asphalt at five different additive contents. The conventional properties of the modified binders were evaluated in terms of their properties using penetration, softening point and dynamic viscosity tests. The fundamental viscoelastic properties of the modified binders were determined using dynamic (oscillatory) mechanical analysis and were presented in the form of temperature- and frequency-dependent rheological parameters. It was determined by the results obtained from the conventional tests that the softening point and the viscosity increased and the penetration and the thermal sensitivity decreased with increased natural asphalt content. Dynamic shear rheometer test results indicated that the temperature and the frequency had a significant effect on the complex modulus of natural asphalt modified binders. Investigation of the types of different additives yielded that the most effective additive was gilsonite in all experiments. The least effective additive in terms of penetration, softening point and viscosity was the Syrian NA and it was asphaltite in terms of PI, whereas the Syrian NA and asphaltite were both less effective in the evaluation of G. It was determined that binders possessing the same additive content showed different performance characteristics in different experiments.*

Keywords: Natural Asphalt, Additives, Modified Binders, Rheology, Temperature susceptibility.

1. INTRODUCTION

Flexible pavement surfaces are composed of two major components; bitumen and aggregates. Cohesion in flexible pavement mixtures is provided by the bituminous binders whereas the aggregates provide the internal shear strength and the stability of the mixture. Bitumen binders also bind the aggregate particles together preventing their breakdown under the traffic load as well as providing driving comfort by providing smooth surfaces. Additionally, they increase the stability of the mixture through cohesion and enabling its impermeability by filling in the voids in the mixture. Although bituminous binders are used in pavement mixtures in as low amounts as 5-7% by weight, they have a significant effect on the mixture performance [1].

Flexible pavement surfaces could be prepared in different forms although the highest strength among these methods is shown by the hot mix asphalts. In the hot mix asphalts (HMAs) used in the flexible pavements of highways, various deformations such as permanent deformation, moisture damage and cracks due to low temperature and fatigue regularly occur. In this context, various modifier materials are employed to enhance the resistance of HMAs against deformation and thus improve the pavement's performance and extend its service life. The modifiers can be added into the bituminous binder as well as into the mixture directly. Styrene-butadiene-styrene (SBS) [2,3], ethylene-vinyl-acetate (EVA) [4,5] and rubber [6] were used in the bitumen modification whereas lime [7] and carbon black [8] were used in mixture modification. The positive characteristics that they possess in order to prevent the variety of deformations that would be encountered in HMAs would thus be observed.

Another type of modifier, which is used in the HMAs, is natural hydrocarbons. Natural bitumens varieties are comprised of two subclasses, namely the subclass of soluble natural bitumens and the subclass of pyrobitumens. Soluble natural bitumens are then divided into three subclasses of mineral wax, natural asphalt (Athabasca, Trinidad Lake, tabbyite) and asphaltite (gilsonite, grahamite, glance pitch) [9]. Trinidad Lake asphalt and gilsonite [10] are the most commonly utilized natural hydrocarbons in the modification of HMA. In Turkey, pylon type natural hydrocarbon deposits are located in the provinces of Şırnak and Silopi in the Southeastern Anatolian Region at an economically feasible thickness. Although the natural hydrocarbons mined from this region are classified to be in between asphaltite and pyrobitumens in terms of their solubility in carbon disulfide, they are collectively named as asphaltite. In the Southeastern Anatolian Region of Turkey, the asphaltite reserves are estimated to total 82 million tons, of which 44.5 million are proved reserves [11].

Large bituminous sand deposits are found in the Al-Beshery and Kefria region of Syria. Al-Beshery material contains a natural mix of sand and bitumen and is more promising than the material in Kefria because of its higher bitumen content and lower sand content. The estimated available Al-Beshery asphalt reserve is about 100 million tons [12]. The use of this natural asphalt is currently limited to low-volume roads, in which the raw asphalt mix is crushed and laid down in layers, to act as a low-quality surface dressing [13].

In this study, three different types of natural hydrocarbons; asphaltite, gilsonite and Syrian Al-Beshery region natural asphalt, were added into the bituminous binder as an additive in 5 different ratios. The effect of different hydrocarbons on the rheological properties of bituminous binders would thus be aimed to be determined in a wide range of experimental conditions.

2. MATERIALS AND SAMPLE PREPARATION

Modified binder specimens were prepared using B 160–220 base asphalt cement obtained from the Batman Petroleum Refinery in Turkey. The physical and the rheological properties of the base bitumen are given in Table 1. The asphaltite was supplied from an asphaltite mine in the Silopi region in Turkey. The asphaltite was ground and the particles smaller than 0.075 mm were collected to be used in bitumen modification. Gilsonite was obtained from the American Gilsonite Company in a natural and resinous hydrocarbon form found in the Uintah Basin in Northeastern Utah. The physical and the chemical properties of the gilsonite and the asphaltite that were used are given in Table 2.

Table 1: Fundamental properties of the binder prior to and following short term aging

Properties	Standard	Results	Specification limits
Penetration (0.1 mm), 100 g, 5 s	ASTM D5	190	160-220
Softening point (°C)	ASTM D36	40.9	35 – 43
Viscosity (cP, 135°C)	ASTM D4402	237.5	max. 3000
Viscosity (cP, 165°C)	ASTM D4402	87.5	-
G*/sinδ (kPa), 58°C	AASHTO T5	1.08	Min. 1.0 kPa
Mixing temperature range (°C)	-	142-149	-
Compaction temperature range (°C)	-	127-133	-
Penetration index (PI)	-	0.12	-
After RTFOT			
Mass Loss (%)	ASTM D2872	0.935	max. 1.0
Penetration (0.1 mm), 100 g, 5 s	ASTM D5	97	-
Retained Penetration, (%)	-	51	min. 37
Softening point (°C)	ASTM D36	50.3	min. 37
Increase in Softening Point (°C)	-	9.4	max. 11
G*/sinδ (kPa), 58°C	AASHTO T5	5.33	min. 2.2 kPa
Penetration index (PI)	-	0.67	-

Table 2: Physical and chemical properties of gilsonite and asphaltite [11]

Properties	Gilsonite results	Asphaltite results
Penetration (0.1 mm), 100 g, 5 s	0	0
Softening point (°C)	160 - 193	-
Ash content (%)	< 1	32.53
Moisture content (%)	< 0.5	6.14
Specific gravity	1.06	1.48
Carbon (weight %)	84.9	54.22
Hydrogen (weight %)	10	5.07
Nitrogen + Oxygen (weight %)	4.7	0.25

Syrian natural asphalt (NA) was obtained from the Al-Beshery region in Syria and NA consisted of two types of materials, including asphalt and mineral aggregate. The results of the extraction test indicated that the mixture was composed of a 17% fraction of bitumen and an 83% fraction of mineral. The results of the NA and the base asphalt analyses are provided in Table 3. While the asphaltane and the resin contents of NA were higher, the saturates and the aromatics contents were lower than those of B 160/220. The modified bitumen samples were prepared using the bitumen extracted from the Syrian natural asphalt. Natural asphalts are shown in Figure 1.

Table 3: Elemental analysis of the Syrian natural asphalt

	Saturates	Asphaltane	Resins	Aromatics
Natural asphalt	2.96	39.20	21.13	36.71
B 160/220	7.55	32.70	16.15	43.60

Figure 1: Appearance of the **Syrian NA, gilsonite and asphaltite**

Different blends of asphalt binders were produced with the selected modifiers using a laboratory scale mixing device equipped with a four-blade impeller as shown in Figure 2. The blends were prepared at a temperature of 180ºC and a rotational speed of 1000 rpm during a period of 1 h. Gilsonite and asphaltite were used in 5 different concentrations in the range of 0-10% by weight of bitumen increasing in 2% increments whereas the Syrian natural asphalt was used in 5 different concentrations in the range of 0-25% by weight of bitumen increasing in 5% increments.

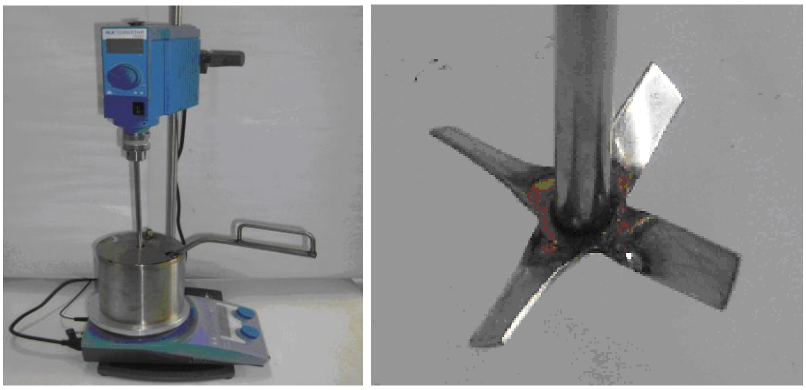

Figure 2: Apparatus and the setup for the preparation of the modified binders

3. TEST METHODS

Conventional binder tests such as the penetration test at 25°C and the softening point test were performed on the neat and the modified binders as stated in ASTM D5 and ASTM D36, respectively. Penetration index (PI) [14] was calculated from the following relationship,

$$\frac{20 - PI}{10 + PI} = 50\frac{\log 800 - pen}{T_{SP} - 25} \qquad (1)$$

where T_{SP} is the softening point and *pen* is the penetration at 25°C. Temperature susceptibility is defined as the change in the consistency parameter as a function of temperature. Lower values of PI indicate higher temperature susceptibility [14, 15].

A Brookfield viscometer (DV-III) was used for the rotational viscosity tests on the neat and the modified bitumens. A viscosity-temperature relationship was obtained in order to determine the mixing and the compaction temperature. The rotational viscosity was determined by measuring the torque required to maintain a constant rotational speed (20 rpm) of a cylindrical spindle while submerged in bitumen maintained at a constant temperature. The mixing and the compaction temperatures were determined for each binder using both 170±20 and 280±30 cP viscosity values.

At present the most commonly used method for fundamental rheological testing of bitumen is by means of dynamic mechanical methods such as oscillatory-type testing. These oscillatory tests are carried out using dynamic shear rheometers (DSRs). The principal viscoelastic parameters obtained from the DSR are the complex shear modulus (G^*), and the phase angle (δ). G^* is defined as the ratio of maximum stress to maximum strain and provides a measure of the total resistance to deformation when the bitumen is subjected to shear loading. G^* has elastic and viscous components, which are designated as the storage modulus (G') and the loss modulus (G''). These two components are related to the complex shear modulus and to each other through the phase (or loss) angle (δ), which is the phase, or the time, or the lag between the applied shear stress and shear strain responses during a test. The phase angle, defined above as the phase difference between stress and strain in an oscillatory test, is a measure of the viscoelastic balance of the material behavior. If δ equals 90° then the bituminous material can be considered to be purely viscous in nature, whereas δ of 0° corresponds to purely elastic behavior. Between these two extremes the material behavior can be considered to be viscoelastic in nature with a combination of viscous and elastic responses [4].

The dynamic shear rheometer (DSR) test was performed on all bitumen samples using a Bohlin DSRII air bearing rheometer manufactured by Bohlin Instruments. The test was performed under controlled-stress loading conditions using frequency sweeps between 0.01 and 10 Hz at temperatures between 40 and 80°C. The tests were carried out with a test plate having the geometry of 25 mm diameter, 1 mm gap and parallel plates.

4. RESULTS AND DISCUSSIONS

It was aimed to determine the rheological characteristics of the neat (B 160/220) and of the 15 different modified binders in this study. Gilsonite and asphaltite were evaluated at 5 different concentrations in the range of 0-10% by weight of bitumen increasing in 2% increments whereas the Syrian natural asphalt was evaluated at 5 different concentrations in the range of 0-25% by weight of bitumen increasing in 5% increments. Initially it was considered to evaluate Syrian NA also in the same concentration range as that for gilsonite and asphaltite (5 different concentrations in the range of 0-10% by weight of bitumen increasing in 2% increments), however, the amount of additive was increased since no significant difference could be observed between the neat and the modified binders when these pre-determined concentrations were used. The variation in the penetration values with respect to the changes in the type and the content of the additive are given in Figure 3, the variations in the softening point are shown in Figure 4 and the variations in the penetration index are displayed in Figure 5. The letter A denotes the modified binders with 2% asphaltite or gilsonite, or 5% Syrian NA; the letter B denotes the modified binders with 4% asphaltite or gilsonite, or 10% Syrian NA; the letter C denotes the modified binders with 6% asphaltite or gilsonite, or 15% Syrian NA; the letter D denotes the modified binders with 8% asphaltite or gilsonite, or 20% Syrian NA; the letter E denotes the modified binders with 10% asphaltite or gilsonite, or 25% Syrian NA in the Figures.

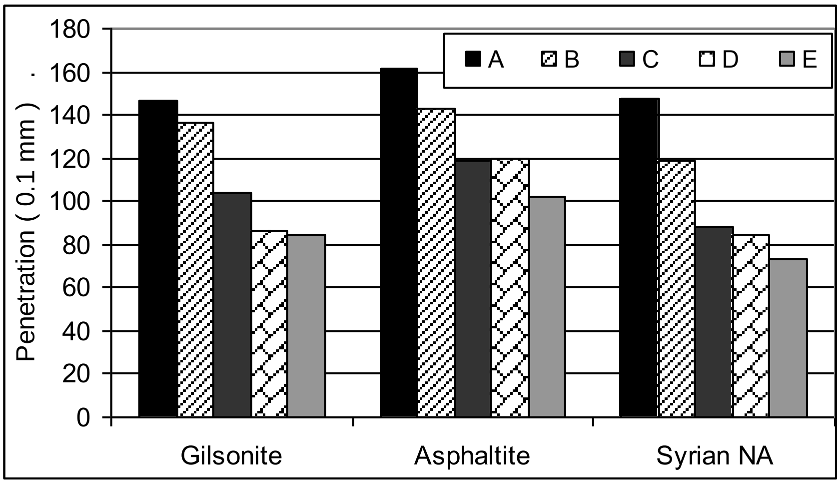

Figure 3: Variation in the penetration values with respect to the type and the amount of the additive

It can be seen in Fig 3 that the penetration was decreasing with the increase in the content of additives. The penetration decreased by 56% when 10% wt gilsonite was used and it decreased by 46% or by 61% when 10% wt asphaltite or 25% Syrian NA were used, respectively. Comparison of the penetration of the binders with 10% additive content indicated that the most effective additive was gilsonite while the least effective one was the Syrian NA (37% decrease).

As it can be observed in Figure 4, the softening point of the binders increased with an increase in the amount of natural additives. This increase in the softening point would indicate increased resistance to permanent deformation through the use of natural asphalt. When 10% wt gilsonite was used, the softening point increased by 36% in comparison to its value for the neat binder whereas it increased by 24% when 10% wt asphaltite was used and by 38% when 25% wt Syrian NA was used. Comparison of the softening point of the binders with 10% additive content indicated that the most effective additive was gilsonite while the least effective one was the Syrian NA (20% increase).

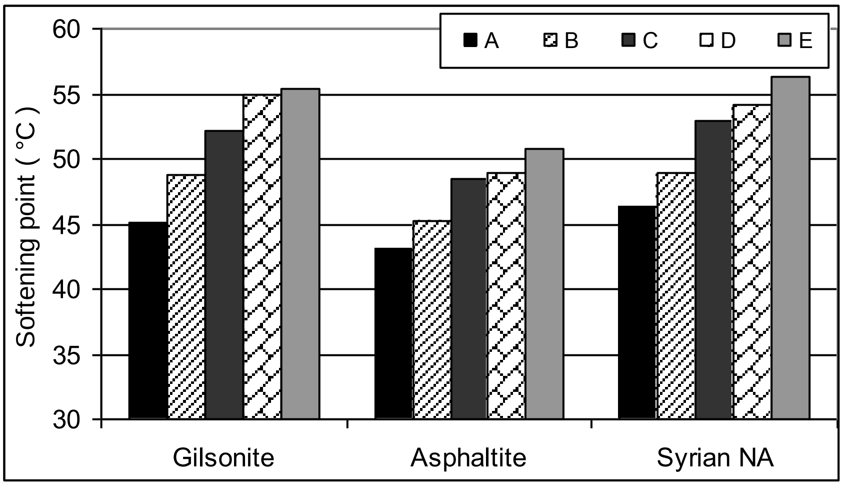

Figure 4: Relationship between the softening point, and the type and the amount of the additive

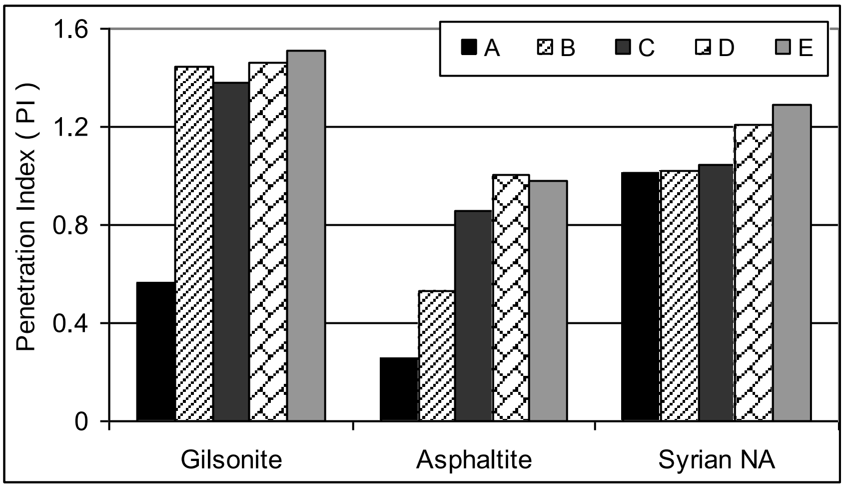

Figure 5: Variation in the penetration index (PI) with respect to the type and the amount of the additive

Investigation of the changes in PI, observed in Figure 5, as a response to variations in the type and the amount of additive yielded that as the amount of natural additives was increased, PI increased and the temperature sensitivity of the binders were observed to decrease in return. Investigation of the type of additives on PI yielded that the highest PI value at 10% additive content was obtained through the addition of gilsonite into the binders and the lowest PI value was obtained through the addition of asphaltite into the binders. It was determined that the high PI bitumen samples would not only have high resistance against permanent deformation but also against low temperature fractures [16]. Taking PI into consideration, it could be concluded that the use of natural asphalt additives would increase the resistance of bituminous binders against both permanent deformation and low temperature fractures.

The variations in viscosity with respect to the type and the amount of additive content at temperatures of 135^{o}C and 165^{o}C are listed in Table 4. The modification index (MI) was used to determine the effect of the type and the amount of the additive content. The modification index was determined by taking the ratio of the viscosity values of the modified binders to the viscosity values of the neat binders.

In accordance with the results of the penetration and the softening point experiments, the viscosity of all types of binders was determined to increase with increasing additive content and their workability would thus decrease. However, since the viscosity of all modified bitumen samples were below 3000cP, the required threshold viscosity for contracts, all binders would be suitable in terms of workability. The results of the viscosity experiments indicated the need for higher mixing and compaction temperatures in the case of using natural asphalt modified binders. The investigation of the modification index comparatively between the values collected at 135^{o}C and 165^{o}C showed that the index values were lower at 165^{o}C indicating that the effect of additives on viscosity would decrease with increasing temperature. Among the binders having 10% wt additive, the highest viscosity was measured for the gilsonite containing binders at 135°C and the viscosity values for all binders having any type of additive were determined to be in close vicinity at 165°C.

Table 4: Viscosity test results for the neat and the modified binders

Additive type and content		Viscosity (cP, 135°C)	Modification index (η_{mb}/η_{neat})	Viscosity (cP, 165°C)	Modification index (η_{mb}/η_{neat})	Mixing temperature range (°C)	Compaction temperature range (°C)
Neat		237.5	-	87.5	-	142-149	127-133
Gilsonite content (%)	2	475.0	2.00	137.5	1.57	160-164	150-155
	4	500.0	2.11	150.0	1.71	162-165	151-156
	6	562.5	2.37	162.5	1.86	163-166	154-158
	8	687.5	2.89	200.0	2.29	166-168	158-162
	10	725.0	3.05	225.0	2.57	167-170	160-164
Asphaltite content (%)	2	475.0	2.00	150.0	1.71	161-165	150-156
	4	500.0	2.11	162.5	1.86	163-166	152-157
	6	537.5	2.26	175.0	2.00	164-167	154-159
	8	550.0	2.32	200.0	2.29	166-169	156-161
	10	575.0	2.42	212.5	2.43	167-170	157-162
Syrian NA content (%)	5	437.5	1.84	125.0	1.43	159-163	147-153
	10	475.0	2.00	137.5	1.57	160-164	150-155
	15	637.5	2.68	175.0	2.00	164-167	156-160
	20	687.5	2.89	200.0	2.29	166-168	158-162
	25	725.0	3.05	212.5	2.43	166-169	159-163

The frequency dependence of the complex modulus for the gilsonite, asphaltite and Syrian NA modified bitumen samples has been displayed in Figures 6, 7 and 8 as obtained by producing the rheological master curves at a reference temperature of 40°C using the time–temperature superposition principle (TTSP) [3].

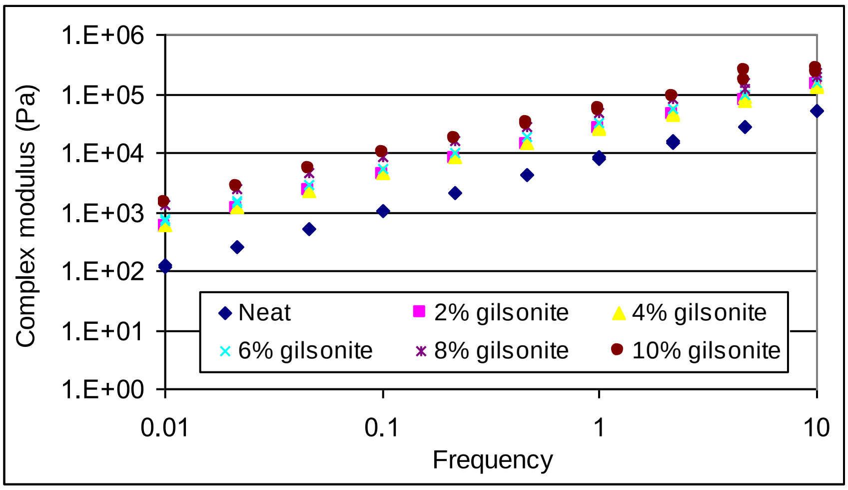

Figure 6: Complex modulus master curves for the gilsonite modified bitumen samples

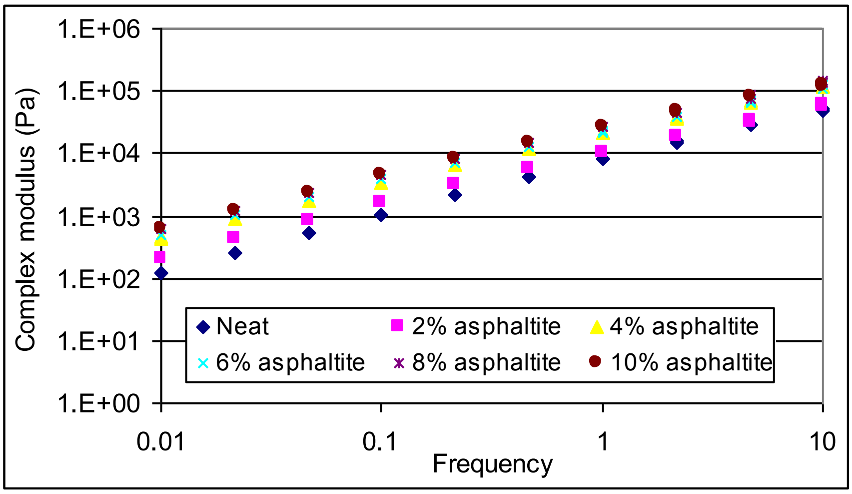

Figure 7: Complex modulus master curves for the asphaltite modified bitumen samples

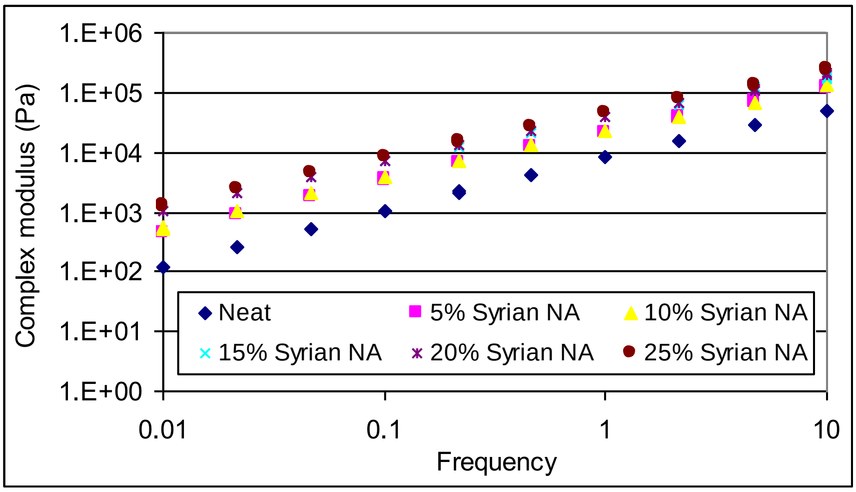

Figure 8: Complex modulus master curves for the Syrian NA modified bitumen samples

As it is observed in Figures 6, 7 and 8, the G* values of all binders increased at all frequencies with the use of additives. Investigation of the complex modulus master curves for the neat binders and the gilsonite added bituminous binders (Figure 6) indicated major differences. The master curves for the bituminous binders with 2%, 4% and 6% gilsonite content were similar. Although the highest complex modulus was obtained for the binder with 10% gilsonite content, the values for the master curves were very similar for the binders with 8% and 10% gilsonite content. The complex modulus values increased as the asphaltite content increased as shown in Figure 7. However, asphaltite was determined not to be as effective on complex modulus values as gilsonite. Investigation of Figure 8 indicated that there was a significant difference between the master curves of the neat binders and the Syrian NA modified binders as it is for the other modified binders. Similar master curves were observed for the binders containing 5% or 10% Syrian NA and for the binders containing 15%, 20% or 25% Syrian NA. The superposed values in order to evaluate the effect of the type and the amount of the additive in the binder yielded complex moduli with four different frequencies, phase angles and modification indices and the values were then compared (Table 5). The modification index values were obtained by taking the ratio of the complex modulus of the modified binders to the complex modulus of the neat binders.

Investigation of Table 5 showed that G* values increased and δ values decreased with increasing additive content and frequency. The phase angle decreased at frequencies lower than 1Hz and it increased at frequencies higher than this threshold. Investigation of the modification index indicated that MI values increased with increasing additive content and decreased with increasing frequency. The additive that increased G* the highest among the binders with 10% wt additives was gilsonite. The G* and the δ values of the binders having 10% content of asphaltite or Syrian NA were similar.

Table 5: G*, δ and the modification index values obtained from the master curves at 40°C

		G* (Pa)				δ (°)				G* Modificatiıon Index G^*_{mb}/G^*_{neat}			
Frequency (Hz)		0.01	0.1	1	10	0.01	0.1	1	10	0.01	0.1	1	10
Neat		121.3	1072	8062	50030	87.96	82.46	76.1	73.66	-	-	-	-
Gilsonite content (%)	2	557.8	4167	24820	143300	81.98	73.98	68.48	73.23	4.60	3.89	3.08	2.86
	4	599.8	4458	26710	135300	82.31	74.46	68.52	64.8	4.94	4.16	3.31	2.70
	6	729	5519	31780	158700	82.31	74.31	68.55	71.89	6.01	5.15	3.94	3.17
	8	1288	8767	47740	197100	79.87	70.71	67.21	71.1	10.62	8.18	5.92	3.94
	10	1386	9710	54050	263600	79.77	71.7	69.59	77.18	11.43	9.06	6.70	5.27
Asphaltite content (%)	2	204	1635	10570	59280	82.22	74.73	68.82	71.42	1.68	1.53	1.31	1.18
	4	442.1	3371	21440	120000	82.53	75.02	68.68	70.87	3.64	3.14	2.66	2.40
	6	514.2	3791	22510	125100	82.36	76.85	68.84	71.41	4.24	3.54	2.79	2.50
	8	599.8	4458	26710	135300	81.07	74.05	68.79	72.00	4.94	4.16	3.31	2.70
	10	599.9	4495	26620	130400	80.46	73.94	68.93	72.13	4.95	4.19	3.30	2.61
Syrian NA content (%)	5	442.1	3371	21440	120000	83.39	76.28	69.54	69.86	3.64	3.14	2.66	2.40
	10	541.1	3930	22510	130700	82.16	73.43	67.99	72.14	4.46	3.67	2.79	2.61
	15	1061	7072	37730	178200	79.42	70.74	65.8	75.33	8.75	6.60	4.68	3.56
	20	1039	7205	38930	191300	80.03	70.89	66.4	77.08	8.57	6.72	4.83	3.82
	25	1218	8318	45020	222500	79.42	70.65	67.28	81.42	10.04	7.76	5.58	4.45

5. CONCLUSION

The effect of three different natural asphalt additives (the American Gilsonite, the Turkish Asphaltite and the Syrian NA) used at five different ratios on the rheological characteristics of bituminous binders was investigated in this study. Based on the results from this study, the relevant findings and the conclusions can be summarized as follows:

The penetration values were observed to gradually decrease with increasing additive content as indicated by the results of the penetration experiments. Comparison of the modified binders all having the same amount of additives (10%) indicated that the most effective additive on penetration values was gilsonite and the least effective additive was the Syrian NA.

The softening point values were determined to gradually increase with increasing additive content as indicated by the softening point experiments. As it was the case for the penetration experiments, the most effective additive was gilsonite and the least effective additive was the Syrian NA.

Investigation of the penetration indices indicated that the PI values would increase with increasing natural additive content, thus decreasing the temperature sensitivity of the binders. Investigation of the PI values indicated that gilsonite was the most effective additive. Asphaltite was the additive, which was the least effective on the PI values.

The requirement for higher mixing and compaction temperatures would be needed if natural asphalt modified binders were used as indicated by the results of the viscosity experiments that were conducted at 135^{o}C and 165^{o}C. Additionally, the effect of additives on viscosity was determined to decrease with increasing temperature. Gilsonite was determined as the additive that would increase viscosity the most as indicated by the experiments conducted at 135^{o}C using different binders with equal amounts of additives. The viscosity of all binders in the same additive group was determined to be in close vicinity at 165^{o}C.

Investigation of the results of the DSR experiments indicated that the complex modulus values increased significantly as the additive content and the frequency increased. In addition, the effectiveness of the additives was shown to decrease with increasing frequency. Frequency was also shown to be effective on the phase angle. The phase angle values decreased noticeably with increasing frequency values. Investigation of the additive types indicated that the most effective additive was gilsonite whereas asphaltite and the Syrian NA were the additives that were the least effective on G* values.

Through the evaluation of the results of all experiments integratively, it could be concluded that by the utilization of the American Gilsonite, the Turkish Asphaltite and the Syrian NA as additives, the high temperature resistance of bituminous binders would increase and their temperature sensitivity would decrease. The G* and the phase angle values indicated that the modified binders would present more elastic behavior. Investigation of the types of different additives yielded that the most effective additive was gilsonite in all experiments. The least effective additive in terms of penetration, softening point and viscosity was the Syrian NA and it was asphaltite in terms of PI, whereas the Syrian NA and asphaltite were both less effective in the evaluation of G*.

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