

Properties evaluation of natural bitumen-filler mastic mixture

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Abstract

Natural bitumen (NB) is a highly precious material and has drawn increasing attention due to its unique properties, especially since it is available in large quantities and has been used in limited fields. In this research, the exploitation of NB from sulfur springs as an alternative energy resource in the production of asphalt pavement is evaluated. It can be concluded from the experimental results that the chemical composition and surface morphology of NB samples are different from those of base asphalt. Besides, the rheological properties of virgin NB are not sufficient for paving work. To overcome this obstacle, NB from five different springs is modified with limestone filler (LSF) to enhance its properties. LSF is a natural material and is available locally at a low price, usually used as filler material in control asphalt mixtures. The study outcomes reveal that LSF is an effective material and plays a fundamental role in improving the properties of NB since it enhances the resistance against temperature susceptibility and improves the ability of NB to disperse in asphalt mixture. Furthermore, treated NB with LSF boosts the mechanical characteristics, increases the stiffness, and strengthens the resistance against water damage for NB mixtures. Particularly, this research clarified that high Marshall stability is achieved with a treated Al-Mamora sulfur spring-NB mixture, which is 30.4% higher than that of the control mixture. At the same time, the treated Al-Askaree sulfur spring-NB mixture has a stiffness index and tensile strength ratio more than the control mixture by 45% and 3.36%, respectively. In conclusion, adding LSF to NB that is extracted from sulfur springs can produce a new type of asphalt binder more suitable for use in road pavement.

Keywords: Natural bitumen, Sulfur spring, Limestone, Natural bitumen-filler mastic, Water damage, Mechanical characteristics

1. Introduction

Extending the lifespan of road highways is the major concern of pavement researchers. Various factors, such as moisture effect, temperature fluctuations, oxidation, and heavy traffic loads, shorten the lifespan of traditional pavement, and they are the main reasons for rutting, cracks, and other types of failures [1]. To enhance the resistance of pavement against these factors, the researchers recommended using a high modulus mixture [2] and modifying asphalt mixtures with different types of additives and materials like fibers, polymers, and nanomaterials [3]. From an environmental perspective, the exploitation of waste materials has become popular in asphalt pavement production and is necessary to reduce pollution [4]. Researchers must focus on exploring new technologies to produce sustainable and eco-friendly environmental pavement as well as to preserve raw materials. Utilization of natural materials is the key component to promoting sustainable practices in the production of road pavement, and these reasons are the motivation for conducting this research. In recent years, natural bitumen (NB) has been utilized in many industrial uses, especially in pavement sectors. For example, NB can be used as an asphalt modifier, filler material, roofing mastic, waterproofing, and asphalt binder. NB occurs in different forms, such as rock asphalt, Gilsonite, lake asphalt, and other forms [5].

Sulfur spring is one of the most abundant sources of NB in Iraq. NB is categorized into three types based on its rheological properties: viscous flow asphalt (sol-asphalt), strongly elastic asphalt (gel-asphalt), and slightly elastic asphalt (sol-gel-asphalt) [6]. In addition, NB has three types of asphalt, each with different properties. The first type, called Sialy, comes from water, sulfur, and natural gas, as well as it is used to spray streets. The second type, called a local Qaasd, is denser and impurity-free, used for polishing palm-made pots. The third type, the Zdade, is thick and dense, mixed with materials, and used for wiping streets or roofs. These NB are melted and used in various applications [7].

NB is abundant in various regions of Iraq. Iraq is among the five countries that have the largest proven oil reserves in the world, with Iraq (8.4%). Many governorates of Iraq are full of bitumen springs, including Basra, Nasiriyah, Dhi-Qar, Karbala, Anbar, Nineveh, Dohuk, Erbil, and Sulaymaniyah. The northern regions contain a greater amount of NB deposits [8, 9]. These deposits are crucial for the production of liquid asphalt and other petrochemical products, extracted from the ground through mining, thermal distillation, or chemical techniques. The naming of some cities was linked to the presence of NB found in them, for example, the name of the city (Heet) is derived from the Babylonian-Assyrian word (Hetu), which means Qir (indicates Bitumen). The largest NB deposit in Iraq is found in Heet City, located in Al-Anbar province. There are more than twenty-six sulfur springs in that region. The percentage of water in bitumen springs varies based on natural deposits. NB extraction in Iraq involves specialized methods such as mining, drilling, thermal cracking, distillation, and modern techniques [10].

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Asphalt tests are essential for ensuring the quality and performance of NB. They are conducted during extraction, refining, and manufacturing processes to assess their physical and chemical properties. Alkhafaji et al. [11] applied a detailed biomarker analysis using gas chromatography-mass spectrometry to determine the source and the extent of NB extracted from the Mishraq sulfur spring located in the north of Iraq. The chemical composition of NB in western Iraq in terms of saturate fraction is evaluated by Farhan et al. [12]. The experimental tests indicated that the aromatic, asphaltene, polar compounds, and saturated NB are 55.67%, 26.56%, 9.93%, and 8.24%, respectively. Based on saturate fraction results, it can be concluded that the origin NB (liquid-viscous form) is petroleum. Meanwhile, Nejres et al. [13] analyzed the saturate fraction components of NB in northern Iraq by conducting an alluvial system using column chromatography and an infrared spectrophotometer. Based on the findings of this research, the aromatic, asphaltene, polar compounds, and saturated NB are 29.34%, 20.04%, 28.58%, and 19.88%, respectively. Consequently, this NB has a colloidal system of SOL type. It is obvious that the saturated fraction components for NB deposits in the western region are completely different than those in the northern region. This difference in element components and chemical properties of NBs depends primarily on their depositional sources [14].

Recently, limited modern studies have appeared that have shown many methods for treating NB and improving its physical and chemical properties. One of the ways to improve the properties of NB is to mix it with synthetic asphalt, such as in the study of Mohammed et al., [15]. In their study, NB from Abu-Jeer spring was mixed with traditional asphalt (Type 40-50) at various percentages of 0, 20, 40, 60, and 80%. The results indicated that Marshall stability for asphalt mixture with 80% NB was improved by 23.5%, and the resistance against moisture damage of the mixture was slightly enhanced by 0.57%, as compared with the control mixture prepared only with asphalt type (40-50).

Heat treatment is another method to enhance NB properties. After NB extraction, it is heated to 150°C in an oven for 30-150 minutes to dry and separate it from water. The second stage of treatment included: the NB heat treatment process involves reheating the asphalt to a temperature of about 163°C. Heating to 163°C leads to improving the physical and chemical properties of NB and making it identical to refinery asphalt. According to the research of Abdul-Jaleel and Najres [16], the heat treatment increased the stiffness and the viscosity of NB. Furthermore, the chemical properties were affected in terms of increasing the ratio of aromatic and asphaltene parts as well as decreasing the ratio of resins and saturated parts. This is attributed to the fact that NB was oxidized after being exposed to heat for different periods as follows: 5, 10, 25, 35, and 55 hours. Also, Ahmed et al. [17] applied heat treatment to modify NB from Abu-Jeer Spring in the for following periods: 5, 10, 15, 20, and 25 hours. The outcomes of this investigation revealed that NB properties are improved after 20 hours of heat treatment to meet the specification requirements of traditional asphalt. Remarkably, the treated NB was less sensitive to temperature, resulting in higher resistance to deformation. Besides, Marshall stability, rigidity index, and moisture damage resistance for asphalt mixtures with treated NB are enhanced by 17.6%, 2.42%, and 0.37%, respectively, as compared with those of conventional mixtures.

Nejres et al. [13] used eggshell waste and low-density polyethylene polymer (LDP) to treat the NB obtained from the Al-Qayyarah region located in the north of Iraq. Eggshells were collected from local restaurants as food waste products. Meanwhile, LDP is a synthetic additive and is commercially available. Eggshell powder (ESP) was added and mixed with NB in proportions of 3, 5, 7, 10, 15, 20, and 25%. LDP is mixed with NB at three ratios as follows: 4, 8, and 12 %. The outcomes determined that the best enhancement in the physical properties was achieved with the addition of 15% ESP and 8% LDP. Engine oil is another waste material that was used to modify NB from sulfur springs in western Iraq, as studied by Mahmood [18]. Various percentages of waste engine oil, 0%, 2%, 4%, 6%, and 8%, are added to NB. It is worth mentioning that no chemical reaction occurred between NB and waste engine oil during the process of modification, with only different density.

Base asphalt is the residue from the distillation of crude oil and has been used widely to construct flexible pavements. Nevertheless, the depletion of oil resources and the concerns of about approaching the oil peak demonstrates the need to explore new sources of asphalt, and NB is the best option. This experimental study has two main objectives. The first objective is to examine the properties of virgin NB from five sulfur springs in Al-Anbar province. The second objective is enhancing the NB properties by treating it with LSF using a physical mixing method to avoid the harmful emissions, huge cost, and aging impact that result from applying heat treatment on NB. Finally, the test results of the virgin and modified NB are compared with those of base asphalt produced from refinery oil.

2. Study area

The study area encompassed several bitumen springs situated in Anbar-Heet Governorate, approximately 190 km west of Baghdad, Iraq, as depicted in Figure 1a. Heet City is located within the longitudinal range of 15°42' to 15°43' east and the latitudinal range of 33°38' to 34°34' north. Out of all the springs, five specific springs were chosen for this study, as depicted in Figure 1b. The selected springs were named as follows: Al-Mamora, Al-Jabal, Al-Attat, Al-Atffa, and Al-Askaree.

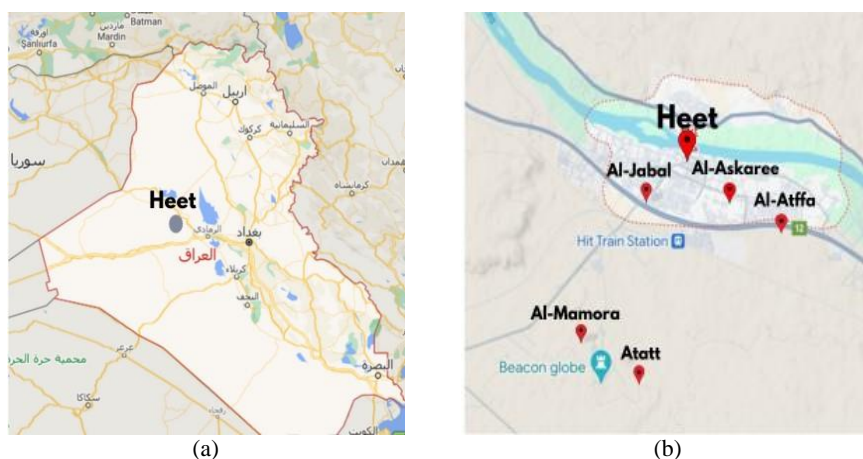


Figure 1 Sulfur spring sites in Heet city-Anbar governorate: (a) Anbar– Iraq; (b) Heet city-Anbar

3. Materials

3.1 Fine and coarse aggregates

The fine and coarse aggregate employed in this study was sourced from Al-Nibaie quarry, following the Iraqi specific requirements [19]. The aggregate was classified as coarse aggregate when it remained between sieves No. 3/4 and No. 4. Conversely, when the aggregate remained between sieve No. 4 and sieve No. 200, it is classified as fine aggregate. In road laboratory, fine and coarse aggregates were sieved and then blended to generate the required aggregate within local criteria as illustrated in Figure 2. Tests were conducted to assess the physical characteristics of both fine and coarse aggregates as showed in Table 1.

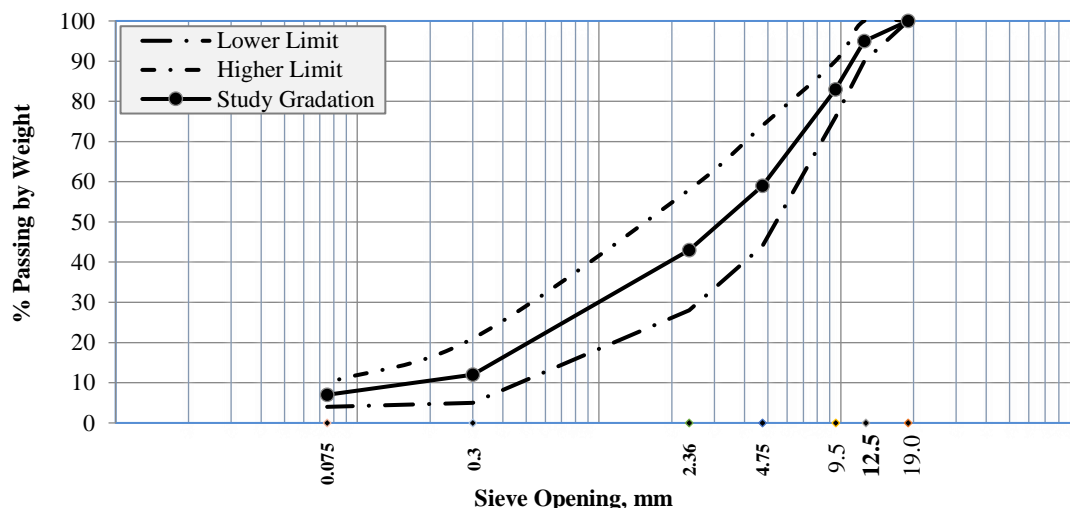


Figure 2 Aggregate gradation for wearing layer

Table 1 Physical properties of coarse and fine aggregates

Aggregate Type	Coarse Aggregate				Fine Aggregate		
Property	Flat and Elongated Particles	Water Absorption	Bulk Specific Gravity	Fractured Pieces	Wearing	Water Absorption	Bulk Specific Gravity
Unit	%	%	Unit less	%	%	%	Unit Less
Limit [19]	10 Maximum	-	-	90 Minimum	30 Maximum	-	-
Method	ASTM D-4791	ASTM C-127	ASTM C-127	ASTM D-5821	ASTM C-131	ASTM C-128	ASTM C-128
Results	4.13	0.22	2.61	97.2	12.7	1.00	2.655

3.2 Asphalt

Two varieties of asphalt were employed in this study.

3.2.1 Base asphalt

The asphalt used in this investigation had a penetration grade of 40–50 according to the specifications of the Department of Roads and Bridges [19]. It is obtained from the distillation process of crude oil at the Dora refinery.

3.2.2 Natural bitumen

Natural Bitumen (NB) was extracted from five different sulfur springs located in the Heet region of Anbar Governorate, Iraq, as depicted in Figure 3. This bitumen contains large quantities of water and impurities. Therefore, it must be removed with water from the sulfur spring. In addition, it is impossible to guarantee obtaining completely pure NB because of the presence of impurities and deleterious materials. However, there is a need to extract NB with a high level of purity to achieve the best performance of NB. For this purpose, it is necessary to avoid taking NB samples from the edges of sulfur springs because NB is under unclean conditions, especially on these edges, and contains high contents of impurities such as clay balls, sand, salts, wood bits, and other forms of contamination.

3.3 Filler

Local Karbala factories in southern Iraq supplied the filler material for this study. LSF is a non-plastic material that passes through a 0.075 mm sieve opening as depicted in Figure 4. The physical properties of LSF are shown in Table 2.



Figure 3 Sulfur Springs in Heet City-Iraq: (a) Al-Mamora Spring; (b) Al-Jabal Spring; (c) Al-Atffa Spring; (d) Atatt Spring, and (e) Al-Askaree Spring



Figure 4 LSF material

Table 2 LSF properties

Property	Unit	Results	Limit [19]
Specific Gravity	Unit Less	2.73	-
Percentage Passing Sieve No. 200 by Weight	%	94	70-100

4. Experimental work

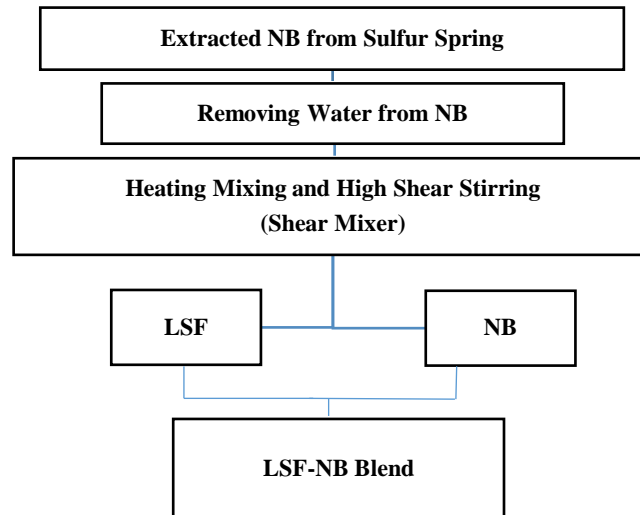
4.1 Modification of natural bitumen with limestone

NB processing includes two stages: the first is drying the NB and isolating it from water by subjecting it to a convection oven at a temperature of 110 °C for different periods ranging from 1.5 to 2.5 hours. The specific drying time depends on the water content in NB. The drying process aims to make NB suitable for the intended application while ensuring that it is completely free of water. The second step is to add LSF, and it must be free of any organic impurities.

The combination of asphalt binder and filler material makes up one mixed so-called bitumen-mastic or asphalt mastic [20]. LSF was used in asphalt mastic by conducting a wet mixing process. By filling the voids and strengthening the bond between the asphalt and the aggregate, LSF enhances the asphalt properties and strengthens the mixture. It is heated to a temperature of 140–145 °C and mixed with NB in proportions of 25, 30, 35, and 40 % at a temperature of 90 °C to achieve the same properties of base asphalt, which is commonly used in paving activities according to local specifications [19]. Moreover, the mixing temperature should not exceed 145 °C, and the process of mixing continues until a homogeneous mixture is obtained. The sulfur springs and their designation, drying time, and mixing time of LSF and NB are listed in Table 3. Also, the treatment process of NB with LSF is illustrated in Figure 5.

Table 3 Drying and mixing time of NB treatment

Sulfur Spring	Designation	Drying Time, hour	Mixing Time, hour
Al-Mamora	MS	2.0	1.00
Al-Jabal	JS	2.5	1.50
Atatt	TS	2.0	1.25
Al-Atffa	AS	1.5	1.00
Al-Askaree	SS	2.5	1.50

**Figure 5** NB treatment process

4.2 Asphalt tests

Multiple tests were performed on base asphalt and NB taken from five springs. These tests are carried out on NB samples before and after being mixed with LSF in varying amounts. Table 4 presents the details of these tests.

Table 4 Physical tests of asphalt binder

Test	Units	ASTM Specification
Penetration, (25 °C, 100 gm, 5 sec)	1/10 mm	ASTM D-5
Ductility, (25 °C, 5 cm/min)	cm	ASTM D-113
Softening Point (Ring and Ball)	°C	ASTM D-36
Flash Point (Cleveland Open Cup)	°C	ASTM D-92
Specific Gravity, at 25 °C	Unit less	ASTM D-70
Solubility in Trichloroethylene	%	ASTM D-2042

4.3 Scanning electron microscopy–energy dispersive X-Ray analysis (SEM–EDX) test

Chemical examination of compounds is crucial as it aids in determining the chemical composition of each material, understanding its basic components to determine their suitability for various uses, and assessing the quality of these materials. It also aids in detecting the presence of impurities in materials that cause damage to the compounds over time. In this study, the energy dispersive X-ray analysis technique (EDX) was used. It is an analysis technique that is used to analyze the elements to know the chemical properties of each element. It is one of the types of X-ray spectroscopy. EDX produces data in the form of spectra, displaying peaks that represent the elements present in the sample under examination. It also plays a crucial role in identifying environmental pollution through the detection of heavy metal contamination. Scanning electron microscope (SEM) comes integrated with EDX spectroscopy. It is considered the most famous and most widely used surface analysis technique [21].

4.4 Marshall test

The asphalt mixture was formulated in accordance with ASTM D 6927-15 by incorporating asphalt heated to 150 °C with an aggregate mixture heated to 135 °C, ensuring that the asphalt was not overheated, since this would alter its properties and adversely impact the total mixture's characteristics. Marshall samples were prepared by incorporating asphalt content varying from 4% to 6%, with an increment of 0.5% for each sample. Three Marshall mixtures were prepared for each asphalt percentage, followed by a period of mixing the hot mixture to achieve homogeneity and assure complete asphalt coverage of all aggregates. Marshall molds are heated to 130 °C, filled with the hot mixture, and then positioned in the Marshall pressure device to compress the samples by 75 strokes per side, following the design parameters for heavy surfaces and basic paving cylinders. The samples are allowed to cool under ambient room conditions for 24 hours following the compression procedure. After that, they are opened, and the samples are prepared for examination by immersing them in a water bath at a temperature of (60 ± 1) for 30 minutes. Immediately after that, the samples are examined with a Marshall device under a constant loading rate of (50 ± 1) mm/min until failure is reached. The force is documented at

the failure point to ascertain the Marshall stability value for each mixture, whereas the total deformation during the stability test is referred to as Marshall flow.

4.5 Indirect tensile strength and tensile strength ratio

The tensile strength ratio (TSR) of compacted mixes was assessed using ASTM D 4867M-96 in order to evaluate the tensile qualities of the mixtures and determine their vulnerability to water damage. This test is employed to ascertain the impact of water on the adhesion and cohesiveness between asphalt particles and aggregate. In order to accomplish this, for base asphalt and NB, after being treated by mixing with LSF in specific proportions for each spring, a set of Marshall specimens was generated using optimum binder content (OBC). The set was divided into two groups: unconditioned and conditioned. The first set of three specimens was subjected to an indirect tensile strength (ITS) test after being immersed in a water bath at 25 °C for 30 min. The second set (three specimens) was then immersed in distilled water at 25°C under vacuum to remove air and then subjected to one cycle of freezing and thawing (16 h at -18°C and then 24 h at 60°C). After that, they were taken out and placed in a water bath for 1 hour while maintaining a temperature of 25 °C, where they were subjected to the same test as the first batch of unconditioned specimens. The rate of 50.8 mm per minute was used to test each specimen. Once the force reached its maximum value, the specimen experienced a total fracture, and this load was recorded. TSR for hot mixture asphalt (HMA) must be more than 80%, which represents the minimum requirement. To compute TSR, the equation below is used:

$$\text{TSR}\% = \frac{\text{ITS con.}}{\text{ITS uncon.}} * 100 \quad (1)$$

$$\text{ITS} = \frac{2000 P_{\max}}{\pi t D} \quad (2)$$

Where:

TSR = Tensile Strength Ratio, %

ITS con. = Average ITS of conditioned groups, kPa

ITS uncon. = Average ITS of unconditioned groups, kPa

P_{\max} = Failure Load, N

t = Thickness of the specimen, mm

D = Diameter of the specimen, mm

5. Results and discussion

5.1 Physical properties of asphalt

To detect the impurities and the contamination in NB samples, a solubility test was conducted since these impurities are not active components and may cause a detrimental effect on the asphalt binder performance. The results of the solubility test, as illustrated in Figure 6, showed that the difference between base asphalt and NB was slight, as NB contains a small percentage of impurities. This is considered normal due to the nature of its formation and composition, as well as its locations, as samples were taken from a medium area of the spring to reduce the amount of impurities. According to the standards specified in the Iraqi specification, which requires the percentage of impurities in asphalt to be greater than 99%, the results revealed that the highest amount of impurities for NB was in AS-NB at a rate of 98.24. Meanwhile, TS-NB and MS-NB are considered almost free of impurities and were the best purity percentage for base asphalt that conforms to the specification.

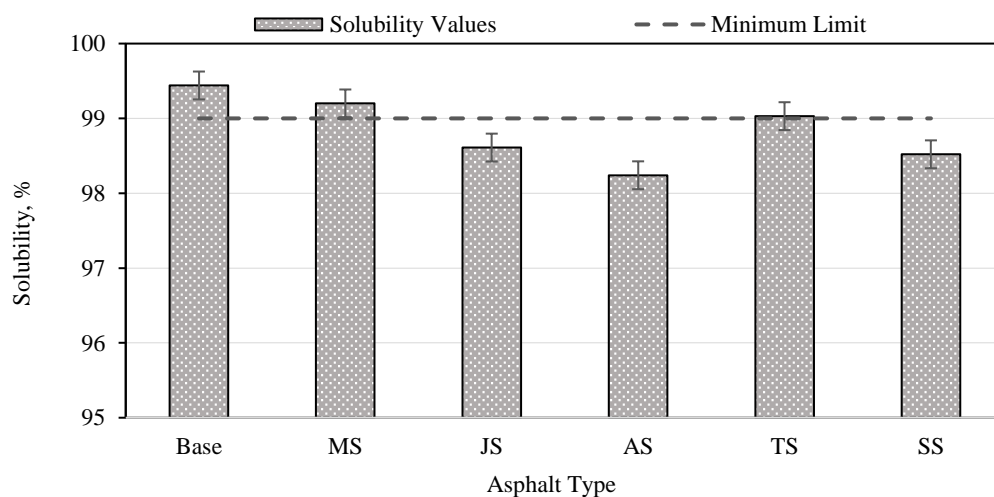


Figure 6 Solubility results for base asphalt and virgin NB samples

The physical characteristics of virgin NB obtained from springs and base asphalt are displayed in Figure 7. Physical testing confirms that base asphalt meets all specified parameters outlined in the local specifications [19], encompassing penetration, flash point, ductility, and other relevant tests. When comparing NB to base asphalt, the test results indicate that NB has a significantly higher penetration value and fails to achieve the specified standards. NB must be treated to enhance its characteristics and meet the requirements.

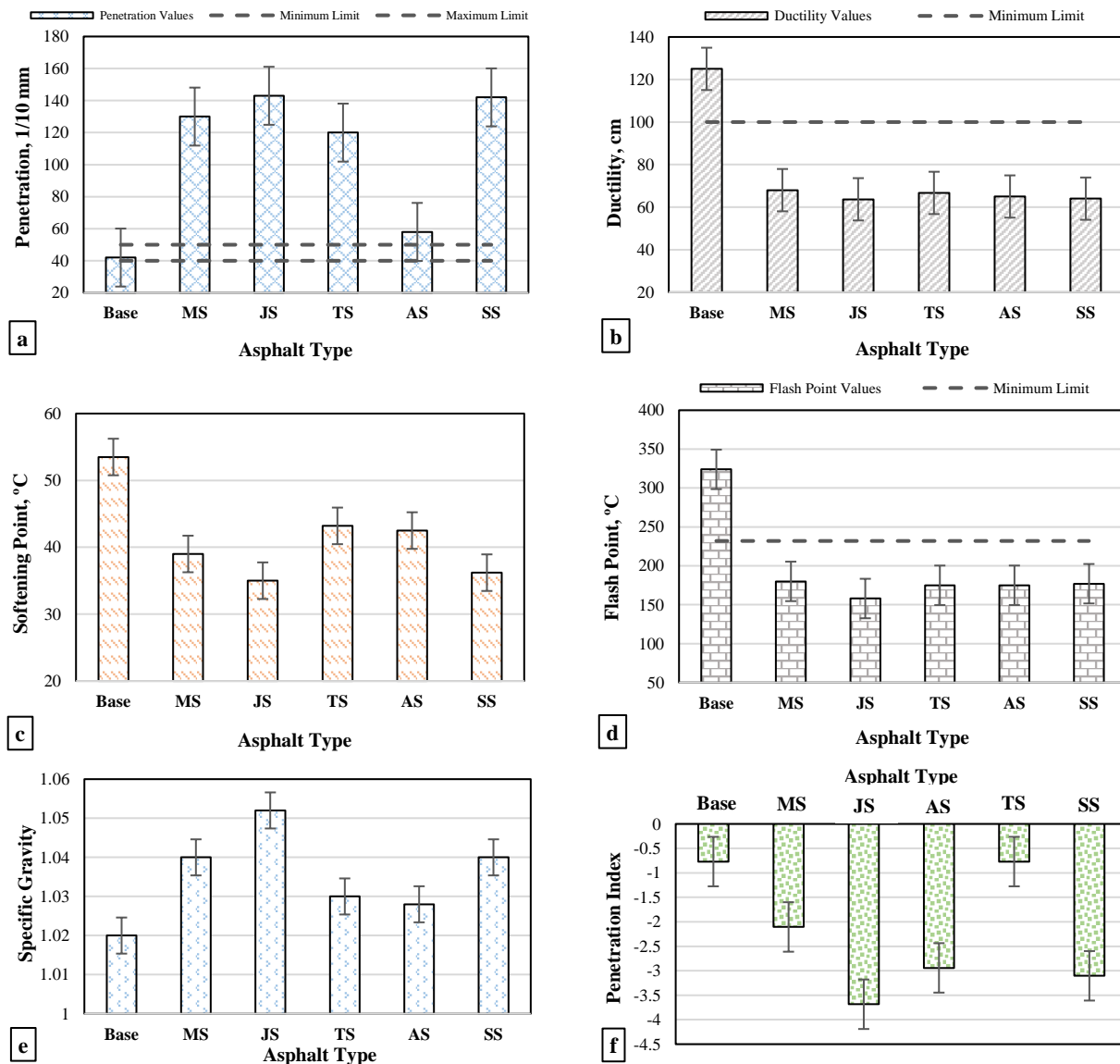


Figure 7 The physical properties of base asphalt and virgin NB: a) penetration; b) ductility; c) softening point; d) flash point, e) specific gravity, and f) penetration index

The physical properties of treated NB with LSF are listed in Table 5. Virgin NB samples from MS, JS, TS, and SS are categorized as soft based on their penetration values, which exceed 120. Whereas, NB from AS can be classified as a hard type since it has a low penetration degree. Hence, it is necessary to treat the NB by blending it with LSF in proportions of 25, 30, 35, and 40 %.

The results of NB-MS demonstrate a decrease in penetration by 65.4% when the LSF ratio reaches 40%. At this ratio, the penetration value reaches 45, which is within the specification limits. This drop in NB penetrability can be attributed to the rise in hardness due to the addition of LSF. During mixing LSF and NB, LSF plays an essential role in toughening and stiffening NB. Additionally, NB-JS demonstrates a significant level of penetration when exposed to the same LSF ratio of 40%, and the penetration of NB-JS decreased to 43. This also meets the local criteria and is close to the penetration of base asphalt. The LSF enhances the asphalt's rheological qualities, resulting in increased hardness. This can be attributed to the fact that LSF can strengthen the uniformity of NB and fill the voids in the asphalt binder, leading to an increase in its strength. This outcome is in full agreement with the previous study [22].

NB-AS is characterized by its hard texture. To address this, it was subjected to a one-hour treatment with 30% LSF. As a result, the penetration reduced to 43, and the softening point climbed to 52, indicating a reduction in softness. This percentage represents the smallest amount of LSF added compared to other samples. For example, the NB-SS sample had a penetration of 142 and was treated with 40% of LSF. As a result, the penetration decreased to 44, and the softening point increased from 36.2 to 52. These changes align with the specifications set by the local standards. Mixing NB with LSF has a significant impact on enhancing the asphalt properties. Specifically, the addition of LSF strengthens the connection between the aggregate and NB, leading to a more cohesive and less permeable mixture. The augmented hardness of the asphalt allows it to endure elevated temperatures without undergoing softening. Consequently, the softening point rises in proportion to the increasing amount of LSF applied. This increase in LSF content improves the thermal sensitivity of asphalt [23].

Treated NB with LSF augments its heat resistance by elevating the flash point, the temperature at which the asphalt commences emitting flammable vapors. It is important to ensure safe processing and application. The results in Table 5 showed that the flash point has a positive correlation with the proportion of LSF incorporated. Notably, the flash point of NB from MS, JS, TS, AS, and SS increased by 36.6%, 47.5%, 37.1%, 34.3%, and 34.5%, respectively. This phenomenon can be attributed to the fact that when LSF

particles are well blended, they can produce an asphalt mixture that is more uniform and constant. Ultimately, this leads to a higher flash point and enhanced molecular bonding within NB. Additionally, LSF helps in the dissipation of heat inside the asphalt structure [24].

Penetration index (PI) is a measure used to describe the susceptibility of asphalt binder to temperature. It provides a theory of how asphalt texture changes with temperature. This is important for understanding asphalt performance, ensuring asphalt longevity, and the quality of asphalt pavements under different temperature conditions. It is calculated based on the penetration degrees of asphalt in the penetration test and the softening point of asphalt in the ring and ball test. High PI values indicate that the asphalt binder is less sensitive to temperature, i.e., it has a more stable texture at different temperatures. Low PI values indicate that the asphalt binder is more sensitive to temperature, i.e., its texture changes significantly with temperature changes. LSF plays a role in improving the sensitivity of asphalt to heat because it increases the hardness of asphalt and improves the softening point due to its high content of oxygen and carbon elements. The results in Table 5 showed that the PI values of treated-NB from MS, JS, and TS were better than base asphalt due to their high PI, meaning that they have less temperature sensitivity and are more suitable for hot regions in Iraq.

Table 5 Physical properties of NB after treated with LSF

Property	LSF	NB					Specification Limits [19]
	Addition, %	MS	JS	TS	AS	SS	
Penetration, 0.1mm	0	130	143	120	58	142	40-50
	25	74	70	74	50	71	
	30	63	62	63	43	60	
	35	56	50	53	-	54	
	40	45	43	46	-	44	
Ductility, cm	0	68	63.7	66.7	65	64	≥ 100
	25	75	70	73	81	80	
	30	83	88	86	>100	99	
	35	96	93	98	-	>100	
	40	>100	>100	>100	-	>100	
Softening Point, °C	0	39	35	43.2	42.5	36.2	-
	25	42	40	45	48	40.5	
	30	47	45.3	48.4	52	43	
	35	50	52	50	-	46	
	40	54	55	53	-	52	
Flash Point, °C	0	180	158	175	175	177	≥ 232
	25	210	178	192	202	189	
	30	221	195	210	235	212	
	35	235	218	228	-	220	
	40	246	233	240	-	238	
Specific Gravity	0	1.0400	1.0520	1.0300	1.0280	1.0400	-
	25	1.0430	1.0524	1.0320	1.0360	1.0410	
	30	1.0438	1.0540	1.0325	1.0363	1.0413	
	35	1.0440	1.0544	1.0332	-	1.0420	
	40	1.0446	1.0548	1.0340	-	1.0421	
Penetration Index	0	-2,105	-3,684	-0,769	-2,941	-3,103	-
	25	-2,592	-3,407	-1,667	-1,667	-3,182	
	30	-1,428	-2,000	-1,045	-1,045	-2,683	
	35	-0,909	-0,769	-1,045	-	-2,105	
	40	-0,476	-0,323	-0,625	-	-1,045	

5.2 Morphology and element analysis properties

Understanding the chemical composition of asphalt is very important because it affects the structural and mechanical properties of the paving mixture, such as strength and hardness. The chemical composition depends on the source of raw asphalt. Asphalt consists of complex hydrocarbons, which are the main components of asphalt and are classified into saturated, aromatic, resinous, and asphaltenes [25]. In addition, it also contains heterogeneous atoms of carbon, sulfur, and oxygen, in addition to small amounts of minerals that have an effect on the physical properties of asphalt. Based on EDX analysis, for base asphalt and NB samples, the spectrum graphs and the percentage of chemical components are displayed in Figure 8 and presented in Table 6, respectively.

The quality of asphalt binder is greatly influenced by its microstructure. The SEM images of base asphalt and NB extracted from the five sulfur springs are shown in Figure 9. Based on the SEM images, base asphalt consists of a uniform mixture of hydrocarbons. The surface of this asphalt appears primarily plain and clear of any noticeable features in the surface morphology. The morphological structure of all NB samples shows a remarkable difference from the base asphalt. This difference may be because NB samples have high component diversity, which contributes to their irregularity in surface morphology. The presence of trace elements with different concentrates, such as sodium, magnesium, silicon, and other contaminants, in NB samples demonstrates their impurity. In addition, the majority of NB in sulfur springs consists of organic compounds, including carbon, sulfur, and oxygen, which have a tendency to promote interactions in materials. These results reflect the diversity of chemical components in NB, which affects its properties and uses.

High carbon content in asphalt has a beneficial impact on the asphalt mixture by enhancing its pliability, hence augmenting the resistance of asphalt to cracking under low temperatures. Additionally, it enhances the asphalt's resistance to deformation at high temperatures, hence boosting its durability [26]. It is clear that NB from the five springs lacks nitrogen and nickel elements in its structure compared to base asphalt, except for NB-MS, which contains nickel with a value of 0.5%. Moreover, the presence of calcium and magnesium in NB has a positive effect on its properties, as it enhances the overall hardness of the material and improves its

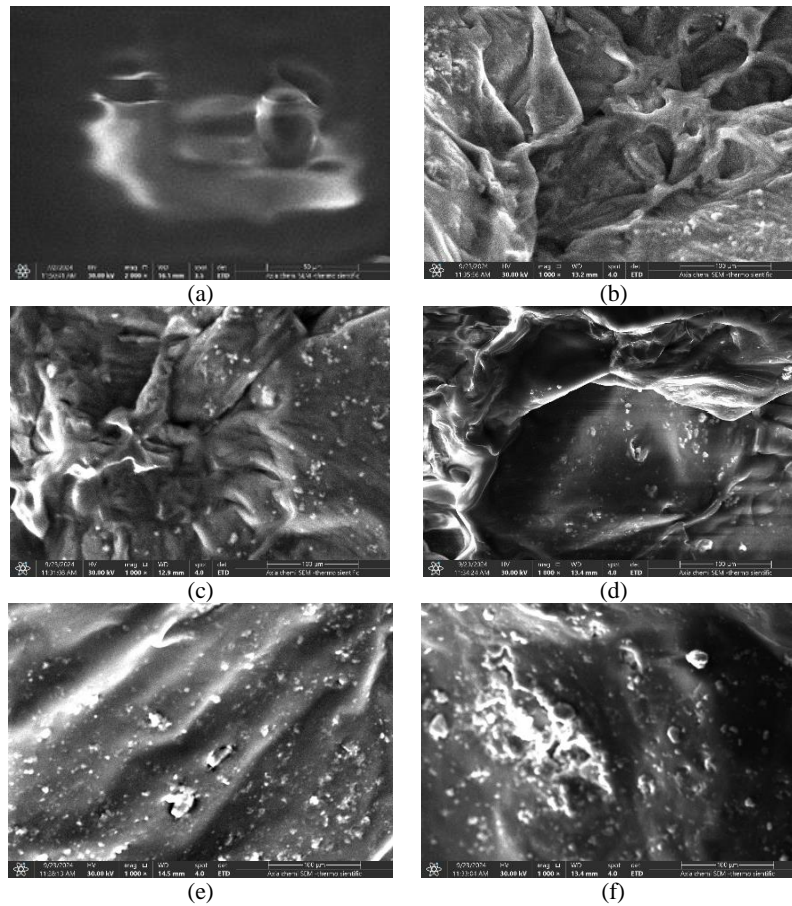
stability, making it more suitable for use in engineering applications [27]. Furthermore, NB is devoid of hazardous substances like lead, mercury, and fluoride, making it exceptionally beneficial for environmental conservation.



Figure 8 EDX spectrum for base asphalt and virgin NB: (a) base asphalt; (b) NB-MS; (c) NB-JS; (d) NB-TS; (e) NB-AS; and (f) NB-SS

Table 6 Elemental composition for base asphalt and virgin NB

Element, %	Asphalt Type					
	Base Asphalt	NB				
		MS	JS	TS	AS	SS
Carbon (C)	83.4	73.2	84.6	81.6	76.9	62.2
Oxygen (O)	1.2	10.7	-	-	-	22.4
Sulfur (S)	11.7	12.1	13.3	14.6	17.8	7.7
Silicon (Si)	0.1	0.6	0.4	-	0.9	1.0
Iron (Fe)	-	0.3	-	-	-	-
Calcium (Ca)	-	1.9	1.4	1.5	3.9	3.2
Magnesium (Mg)	-	0.5	0.3	0.4	0.5	1.6
Sodium (Na)	-	0.2	-	1.0	-	0.8
Nitrogen (N)	2.6	-	-	-	-	-
Nickel (Ni)	1.0	0.5	-	-	-	-
Chlorine (Cl)	-	-	-	0.9	-	1.1

**Figure 9** SEM images for base asphalt and virgin NB: (a) base asphalt; (b) NB-MS; (c) NB-JS; (d) NB-TS; (e) NB-AS; and (f) NB-SS

SEM analysis, shown in Figure 10, indicates that LSF particles have an irregular shape, which enhances their ability to fill gaps in asphalt and maintain homogeneity with it. The rough surface increases friction and thus strengthens asphalt adhesion, and the small size of the particles increases the surface area of interaction with asphalt, which further enhances the properties [28, 29]. In other words, the rough surface of LSF leads to an increase in the interlocking and the internal friction in asphalt mastic between LSF particles and asphalt binder, resulting in a better performance of asphalt mastic than that of base asphalt.

A chemical test of LSF is important to ensure a comprehensive understanding of the performance of LSF and its potential to interact with the asphalt binder. This affects the properties of asphalt and also the properties and bonding of the asphalt mixture components. Figure 11 shows the EDX spectrum and chemical composition of LSF, which consists primarily of oxygen and calcium in high proportions (54.7, 25.8), respectively, and contains small amounts of carbon, sulfur, and silicon. Therefore, LSF is considered an alkaline mineral since the content of Si was the lowest (1.6%) compared to the content of oxygen and calcium elements. The presence of calcium in LSF is important and has a positive effect because it forms strong bonds when in contact with hydrophobic compounds such as asphalt, increasing the tensile strength values [30]. Oxygen participates in reactions with other compounds, which leads to the formation of new compounds that help improve the asphalt and mechanical properties. The most important of these compounds is calcium oxide (CaO), which contributes to hardness by enhancing the adhesion to asphalt, thus increasing the durability and hardness of the asphalt mixture, reducing the appearance of deformations, and also improving moisture damage. Because of its role in enhancing the properties of asphalt and the mixture, it makes it an excellent choice for use as an improving material [31].

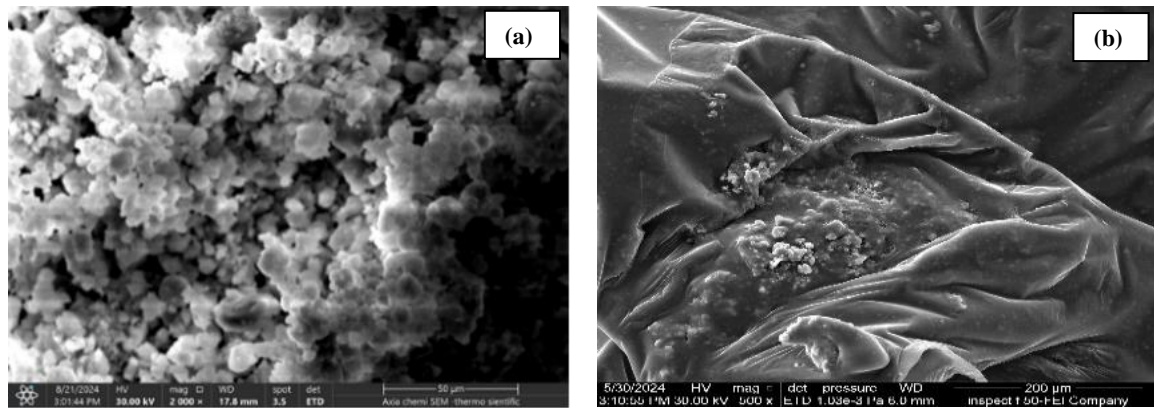


Figure 10 SEM images for LSF material and treated NB-TS: (a) LSF and (b) LSF+NB-TS

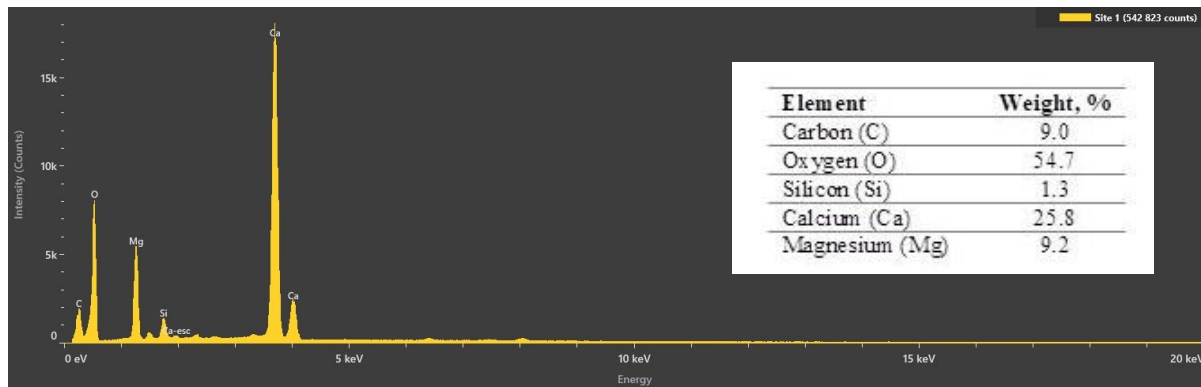


Figure 11 EDX spectrum and elemental composition for LSF material

5.3 Effect of LSF-modified NB on the properties of hot mixture asphalt

5.3.1 Marshall properties

Table 7 shows the volumetric characteristics of control and NB mixtures. According to local specifications [19], the maximum allowable amount of air voids is between three and five percent. The presence of voids in roads contributes to road erosion, as well as accelerated oxidation of asphalt and heightened permeability, resulting in issues that reduce the road's lifespan. Reduced voids also contribute to the gradual deformation and eruption of asphalt. Air void content affects the durability of asphalt pavement mixture [32]. Air void values in both control and NB mixtures are close, and both fall within the acceptable range according to local specifications. The control mixture has the most voids (3.8%), while the NB-SS mixture has the lowest voids (3%). The presence of LSF significantly impacts the voids by effectively filling and reducing them within the mixture, as a result of the decrease in voids, the density of the mixture increases to its maximum and then decreases slightly. This means that adding LSF increases the mixture density and decreases voids, hence enhancing the stiffness [33].

Voids in mineral aggregate (VMA) are the air spaces between the aggregate particles in the compacted mixture, as well as the spaces filled with asphalt. Its purpose is to ensure that the asphalt content is reasonably high to cover the aggregate particles in the mixture. The table shows the VMA results for both NB and control mixtures. There was a slight difference between them. This is attributed to the effect of the added LSF, which reduces the VMA of the mixture. However, the amount of added LSF must be carefully considered, as an increase in LSF leads to an increase in the VMA. This is due to the relationship between the diameter of the particles and the surface area. Increasing the VMA results in an increase in the thin asphalt layer surrounding the aggregate particles, which can have a negative impact and potentially be harmful.

Voids filled with binder (VFB) are the voids filled with bitumen in the spaces between the aggregates in the compacted mixture. They are a basic criterion in the design of the asphalt mix and control the durability of the mix. If the VFB values of the mix are too low, this means that the amount of asphalt required to achieve durability and density under the influence of traffic and bleeding is insufficient and does not achieve the purpose. It can be concluded from the results in the table that VFB values of the control and treated NB mixtures are similar or somewhat lower when compared, except for the VFB value of SS-NB, which increases by 3.8% when compared with the control mixture.

Table 7 Volumetric properties for control and treated NB mixtures

Property	Unit	Mixture Type						Specification Requirements [19]
		Control	NB-MS	NB-JS	NB-AS	NB-TS	NB-SS	
Bulk Density	gm/cm ³	2.330	2.343	2.337	2.331	2.345	2.353	-
Air Voids	%	3.8	3.5	3.2	3.3	3.1	3.0	3-5
VMA	%	16.40	15.95	15.57	16.40	15.6	15.8	14 minimum
VFB	%	76.8	78.1	79.5	79.9	80.1	81.0	-

The Marshall stability and flow significantly influence the performance of HMA pavements by demonstrating the pavement's resistance to deformation from applied stresses. For the control mixture prepared using base asphalt, the OBC percentage is 5%. When the NB samples are mixed with LSF, the percentages of OBC are 5.2%, 4.96%, 5.1%, 4.96%, and 5.1% for the mixture made with NB from MS, JS, AS, TS, and SS, respectively. Figures 12 and 13 display the mechanical properties (Marshall stability and flow) for both control and treated NB mixtures.

In Figure 12, the results demonstrate that the stability of NB mixes surpasses that of control mixtures. The stability values for mixtures including NB from MS, JS, AS, TS, and SS were recorded as 12, 9.8, 11.6, 11, and 11.8 kN, respectively. In contrast, the stability of the control mixture was measured at 9.2 kN. When compared to control mixtures, the stability of NB mixtures increased by 30.4%, 6.5%, 26.1%, 19.6%, and 28.3% for MS, JS, AS, TS, and SS, respectively. The stability values of all mixtures meet the specification's limit (8 kN minimum value for the wearing layer) [19]. The increased stability of asphalt can be linked to the impact of LSF, which is added to improve its characteristics and raise its hardness. The use of LSF in asphalt results in the formation of a very rigid asphalt paste, which enhances the cohesion and bonding strength between the aggregates and NB. Additionally, LSF fills the gaps between the blocks, improving the stability of the asphalt mixtures. This leads to a more stable blend that is able to withstand deformation caused by traffic loads and enhances the ease of handling of the asphalt mixture. However, it is essential to take into account the amount of LSF used, since it directly affects the durability of the asphalt. Indeed, a rise in the quantity of LSF could result in a decrease in durability [34].

According to the Marshall test result, Figure 13 displays the Marshall flow of both the control and the NB mixtures. The flow values are in close proximity. The flow range, as defined by the specification limitations [19], falls within the range of 2 to 4 mm, and all mixtures meet this requirement.

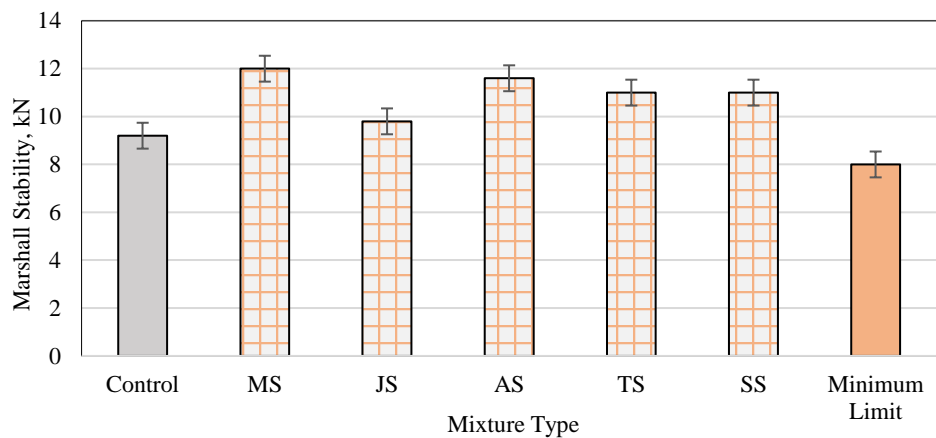


Figure 12 Marshall stability results

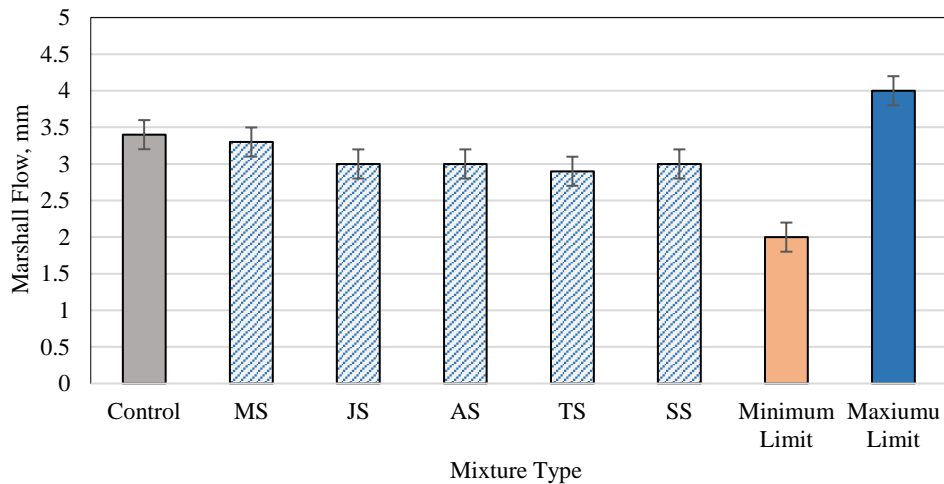


Figure 13 Marshall flow results

The Marshall quotient (MQ), often known as the stiffness index, for both control and NB mixtures, is displayed in Figure 14. MQ is a measure of the ratio between Marshall stability and Marshall flow. It serves as an indicator of the asphalt mixture's capacity to resist rutting. Greater values of MQ imply enhanced stability and increased resistance to deformation in the combination. MQ is influenced by factors like aggregate characteristics, asphalt binder content, and compaction effort. Thus, an increase in MQ indicates an enhancement in the flow behavior stability of the asphalt mixture. This is advantageous for durable, high-quality asphalt pavements [35]. According to the results, MQ was higher for NB mixtures. Among these mixtures, SS-NB had the greatest ratio, with a significant increase of 45% compared to the MQ of the control mixture. Based on the outcomes of the research of Chen et al. [36], increasing the stiffness (rigidity) of asphalt mastic can significantly enhance the strength of asphalt pavement.

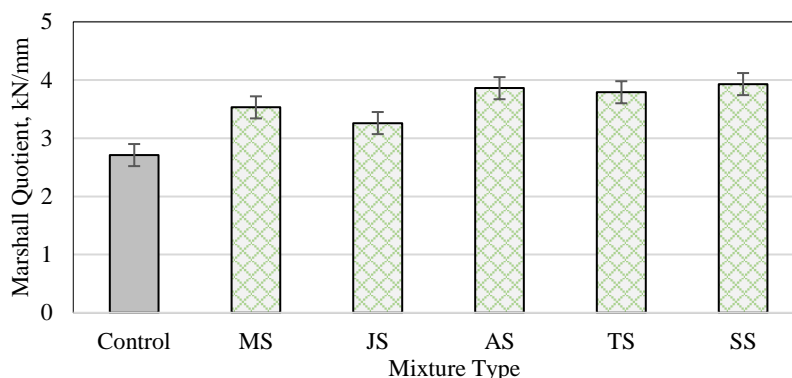


Figure 14 Marshall quotient results

5.3.2 Water damage evaluation

TSR, or the moisture durability index of the mixture, is required to have a minimum value of 80% as specified. The performance of TSR relies on the properties of adhesion and cohesion. Figures 15 and 16 display ITS and TSR values of both control and NB mixtures. The test findings indicate that the NB mixtures exhibited the highest resistance values to moisture and water conditions. The NB-SS mixture has a 3.36% higher resistance to moisture damage compared to the control mixture. In addition, adding LSF to NB has a positive effect on the asphalt mixture as it improves the water resistance of the mixture due to its effect on the cohesion of the asphalt because LSF material is a water-repellent material and thus enhances the bond between the aggregate and the asphalt. Also, the hardness of the mixture increases because LSF has a finer particle, the more it increases the potential hardness of the asphalt and reduces air voids. Furthermore, the abundant presence of CaO in the LSF has a chemical impact on the moisture resistance of the mixture. Besides, when the asphalt binder is exposed to water, the chemical bond between asphalt and LSF is not easily destroyed because the acidic polar group of LSF forms a stable double bond structure with asphalt binder [37]. This results in the formation of a strongly alkaline substance that improves the bonding between the aggregate and the asphalt, hence decreasing the chances of separation due to moisture [38, 39].

ITS test may reflect the combined tensile-compressive properties of the asphalt mixture [40, 41]. Therefore, it is recommended to conduct more tests, such as the uniaxial compressive tests, and four-point bending modulus tests, to assess the strength of control and NB-mixtures for more accurate prediction of asphalt pavement responses.

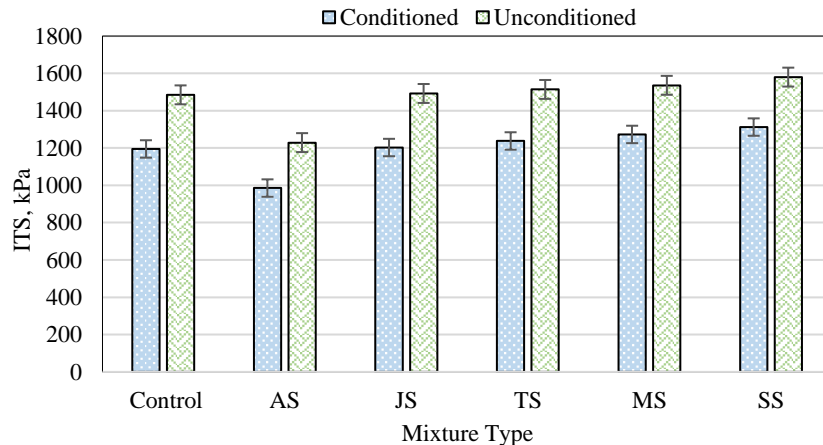


Figure 15 ITS for unconditioned and conditioned mixtures

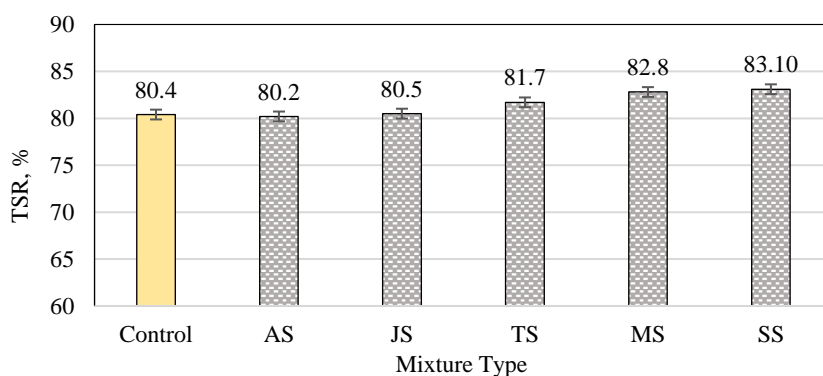


Figure 16 TSR for control and treated NB mixtures

6. Conclusions

LSF is one of the main components of the binder-filler-aggregate system. It has a significant effect on the asphalt properties and paving performance of asphalt mixtures. This paper studied the physical and chemical properties of NB samples, base asphalt, and LSF. Also, the effects of LSF on the physical and mechanical properties of NB after mixing are analyzed. Furthermore, the paving performance of treated NB and base asphalt mixtures was evaluated and compared. The following results were obtained:

- Based on the results of the asphalt tests, unfortunately, virgin NB from all sulfur springs cannot be used in local paving works since the physical properties of NB do not meet the specification limits.
- LSF is a natural filler that is widely available at reasonable prices and has many benefits in improving the physical properties of asphalt and the mechanical properties of the asphalt mixture. This is evident from its chemical composition and the shape of its atoms in the SEM - EDX test.
- LSF has a direct impact on the physical properties of asphalt. The post-curing physical and chemical tests revealed a noteworthy enhancement in the characteristics of NB. These improvements include a reduction in penetration, an increase in ductility, a rise in softening point, an improvement in the sensitivity of asphalt to heat by decreasing PI, and an improvement in the flash point. As a result, the qualities of NB became close to the properties of base asphalt.
- The presence of LSF greatly influences the mechanical properties of HMA. According to the experimental results, the treated NB with LSF mixture had better stability than the base asphalt mixture. This is explained by the fact that LSF had a strong hardening effect on the NB mixture because it filled the voids and increased the bond between the asphalt and the aggregate, which increased the stability and hardness of the mixture.
- LSF enhances the moisture resistance of NB mixtures by augmenting the cohesiveness between the aggregate and the asphalt, thereby diminishing the separation caused by moisture. Upon analyzing the TSR test results, it was found that the NB-SS mixture exhibited the greatest TSR value of 83.1%, surpassing the TSR of the base asphalt mixture by 3.36%. Furthermore, the water sensitivity of all mixtures was sufficient since TSR values for all asphalt mixtures (from 80.4% to 83.1%) exceeded 80%, the minimum requirement. A slight difference in TSR between base asphalt and NB mixtures was noted.
- It is important to mention that increasing the addition of LSF to NB may negatively influence the NB fatigue performance due to the stiffening effect of LSF. Therefore, it is recommended for future studies to evaluate the cracking resistance of NB mixtures at low temperatures. In addition, further experimental evaluation to measure the viscoelastic properties of NB is recommended to advance the understanding of the rheological behavior of NB.

In summary, LSF is a very promising and economical material for enhancing the properties of asphalt mixtures in terms of mechanical properties, stiffness index, volumetric properties, and resistance to moisture damage. NB is also a valuable and economically cheap material because it is widely available throughout our nation, from north to south, and can be invested in and used in paving operations. Thus, NB is a national resource that will require intensive economic research in the future, and more efforts are still needed to discover other ways to treat NB and make it suitable to use as an asphalt binder in the production of flexible pavement.

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