https://doi.org/10.31436/iiumej.v26i2.3452

NATURAL BITUMEN IN HOT ASPHALT MIXTURE: SUITABILITY OF USING TREATED NATURAL BITUMEN INSTEAD OF PETROLEUM ASPHALT BINDER

HANEEN KAREEM MOHSIN, ROAA HAMED LATIEF*

Department of Civil Engineering, College of Engineering, University of Baghdad, Baghdad, Iraq

**Corresponding author: roaa.hamed@coeng.uobaghdad.edu.iq*

(Received: 23 September 2024; Accepted: 17 February 2025; Published online: 15 May 2025)

ABSTRACT: In recent years, the search for economic and environmentally friendly alternatives has become a global necessity to achieve sustainability and preserve raw materials. From this concept, natural bitumen (NB) derived from sulphur springs is now one of the most promising alternative energy resources for many applications, especially in asphalt pavement construction. Its low price and abundance characterise NB since sulphur springs produce thousands of tonnes of NB annually and are used in very limited fields. Two main objectives were adopted for this work. The first objective is to examine the virgin NB properties from five sulphur springs and compare them with petroleum asphalt. The second objective is to enhance NB properties by applying heat treatment. The experimental results reveal that heat treatment is highly effective in improving the NB properties. This was confirmed by conducting many tests, such as asphalt, Marshall, SEM-EDX, and indirect tensile strength tests. Based on the experimental test outcomes, virgin NB properties do not conform to asphalt specification limits and are unsuitable for flexible roads. Also, MS-NB significantly affected the mechanical properties of the asphalt mixture, as Marshall's stability increased by 41.3% compared to the conventional mixture. In addition, SS-NB was more effective in moisture damage resistance due to increasing the tensile strength ratio by 5.72 % and provided a higher stiffness index than the conventional mixture by 40.36%. In conclusion, the treated NB can successfully be used as a binder material in hot asphalt mixtures.

ABSTRAK: Beberapa tahun kebelakangan, pencarian alternatif baharu ekonomi dan mesra alam telah menjadi satu keperluan global bagi mencapai kemampanan dan memelihara bahan mentah. Melalui konsep ini, bitumen asli (NB) yang diperoleh daripada mata air sulfur kini merupakan satu sumber tenaga alternatif yang berguna bagi digunakan dalam pelbagai aplikasi, terutamanya dalam pembinaan turapan asfalt. NB dicirikan oleh harga yang rendah dan kelimpahannya kerana mata air sulfur menghasilkan beribu-ribu tan NB setiap tahun dan digunakan dalam bidang terhad. Dua objektif utama telah dipakai untuk kajian ini. Objektif pertama adalah memeriksa sifat NB dara daripada lima mata air sulfur dan membandingkannya dengan asfalt petroleum. Objektif kedua adalah meningkatkan sifat NB dengan menggunakan rawatan haba. Dapatan eksperimen mendedahkan bahawa rawatan haba sangat berkesan dalam meningkatkan sifat NB. Ini disahkan dengan menjalankan pelbagai ujian, seperti ujian asfalt, ujian Marshall, ujian SEM-EDX, dan ujian kekuatan tegangan tidak langsung. Berdasarkan dapatan eksperimen, sifat NB dara tidak mematuhi had spesifikasi asfalt dan tidak sesuai untuk jalan fleksibel. Selain itu, MS-NB mempunyai kesan ketara pada sifat mekanikal campuran asfalt kerana kestabilan Marshalls meningkat sebanyak 41.3% berbanding campuran konvensional. Tambahan, SS-NB lebih berkesan dalam rintangan kerosakan lembapan kerana meningkatkan nisbah kekuatan tegangan sebanyak 5.72 % dan memberikan indeks kekukuhan yang lebih tinggi daripada campuran konvensional sebanyak 40.36%. Kesimpulannya, NB yang dirawat boleh berjaya digunakan sebagai bahan pengikat dalam campuran asfalt panas.

KEYWORDS: *Natural bitumen, heat treatment, petroleum asphalt, Marshall, moisture damage.*

1. INTRODUCTION

Road construction is crucial for improving connectivity by creating efficient, safe, and durable routes for traffic movement, while also enhancing the quality of life for communities and promoting economic growth. Therefore, ensuring high performance and quality of paved roadways is essential to maximize progress across the industrial, agricultural, and commercial sectors. More specifically, high-performance roads are necessary for the industrial sector to accommodate larger wagons safely. For the agricultural sector, roads enable easier market access to preserve perishable products and reduce transportation costs. For the commercial sector, roads offer faster routes for transporting goods over short distances, thereby minimizing costs and reducing delays. This requires continuous road development efforts by developing maintenance programs, expanding road networks, and, most importantly, enhancing road pavement quality to prevent failures. The evolving landscape of technologies and materials supports the development of a global social economy, leading to the adoption of innovative approaches and materials in road construction [1-5].

The global issue of high-viscosity oil and natural bitumen (NB) commercial development is influenced by increasing hydrocarbon consumption, oil reserve structure changes, heavy crude dominance, and innovative technologies for bitumen production and processing [6], [7]. Oil remains a significant global resource, representing over 30% of energy consumption. In 2019, global oil stockpiles decreased by 0.1%, leading to decreased quantities of asphalt binder produced from refinery oil, as part of the crude oil is converted to asphalt binder during the distillation process. Furthermore, Venezuela, Saudi Arabia, Canada, Iran, and Iraq have the largest proven reserves [8-9]. This has prompted road engineers to search for alternatives, as asphalt affects the properties of the mixture and is considered 7-8 times more expensive than aggregate. The high cost and lack of resources have prompted the search for alternatives to industrial asphalt. NB exists in bitumen deposits, asphalt lakes, asphalt rocks, etc. It improves pavement performance and reduces hot mixture asphalt (HMA) prices [10].

The first major discovery of NB in the UK was at Alphalt in 1877, which became the first oil shale mine. However, oil extraction and refining became more critical than the mine, and it closed in 1919 as the shale oil industry phased out. Despite its numerous advantages, NB remains a lesser-known material globally because it is found in specific places worldwide and cannot be used in pavement production without treatment [11].

NB is a complex mixture of organic compounds with a chemical composition that includes long hydrocarbon chains and intense, polar aromatic rings. This gives asphalt a wide range of properties, from solid to liquid, encouraging researchers to study and improve its properties for use as an alternative in paving operations. NB is a unique material found in Trinidad and the Dead Sea. It has been used for centuries in various applications, including bath construction, embalming, shipbuilding, and waterproofing [12-13].

Bitumen, a natural asphalt found in Iraq, is formed from decomposing living organisms and solids. It is a petroleum derivative that remains on the Earth's surface after the evaporation of light elements. Bitumen is found in geological areas affected by faults or fissures, causing groundwater to flow from the ground to the surface. It contains salts and sulphur in high percentages. The Heet region is oil-rich, as evidenced by bitumen springs [14].

Bitumen is a petroleum-derived, non-crystalline solid or viscous substance that dissolves in carbon disulfide. Its composition consists of carbon (87%), hydrogen (11%), oxygen (2%), and trace amounts of nitrogen, iron, and nickel [15]. It has a brown or black appearance. It is a suitable option for construction materials, as it is porous, brittle, and prone to oxidation. Although flammable, it is difficult to ignite [16-17]. NB is a black, hard, and brittle material made from a mixture of high molecular weight hydrocarbons, with many carbons, up to 150 atoms. It has a viscous fluid consistency, a special smell, and is easily soluble. There are two forms: solid asphalt rocks and sticky materials from warm sulphur springs [18].

Many physical and chemical processes have been used to treat and develop NB and meet the requirements of industrial use. Shlimon et al. [19] evaluated oil wells in the Kurdistan region (North of Iraq) to determine the origin of NB and its components. Also, the chemical properties of NB from the springs in the Heet region are investigated by Farhan et al. [20] by conducting many chemical tests and identifying the chemical elements of NB. Also, Muttar et al. [21] studied the NB and water of the three springs in Heet: Al-Khader, Al-Khalidiyah, and Al-Shuhada. In this study, the NB was dried, separated from the water, and treated by heating to enhance its properties. The physical and chemical properties were examined, and improvement was shown. Furthermore, the study revealed a chemical similarity between NB and conventional asphalt.

The investigation by Al-Dulaymie et al. [14] studied the components of NB from the Abu Al-Jeer spring. It evaluated the characteristics of the water from this spring and its components. In this study, a ranking and screening technique was developed to assess preferred sulphur springs specified for natural therapy, and the application of this technique demonstrates four graded, consequent preferred sulphur springs for balneotherapeutic investment. In Abdul-Jaleel and Najres [22] investigation, oxidation treatment was applied to enhance the properties of NB produced from the Abu Al-Jeer spring. In this treatment, NB was heated in an oven at 160°C for different periods: 5, 10, 25, 35, and 55 hours. The chemical test results indicated that the aromatic ratio was decreased, whereas the asphaltenes increased due to oxidation. This leads to the transformation of the NB to a gel type. Also, increasing viscosity and ductility, along with reducing penetration, are other effects of oxidation treatment on the physical properties of NB.

The extraction and examination of NB for use as an alternative to refinery asphalt were studied by Abdul-Jaleel et al. [23]. There were two methods to conduct the study: the first, using liquid chromatography, the asphalt was separated, then Sarah N-pentane was used as a solvent to examine the chemical properties, and Fourier Transform Infrared (FT-IR) analysis was performed to analyze the components thoroughly. Improving the rheological properties of NB derived from the Abu-Aljeer sulphur spring was the second strategy by combining NB after removing water in specific proportions (5, 15, 35, and 35) % with natural, readily available, in large quantities in the Al-Anbar governorate, and low-cost minerals (limestone), due to the great effectiveness of calcium oxide CaO (limestone) and its ability to create bonds of adsorption Chemo–Physics to form the connection between asphalt hydrocarbon chains. The outcomes revealed that the softening point, flash point, fire point, and viscosity are increased, and the penetration is decreased with the addition of limestone; moreover, the optimal ratio of limestone addition is 35%.

Mahmood et al. [24] studied and evaluated the NB derived from Heet city and used polyvinyl chloride (PVC) pipes as a modifier to enhance the properties of NB and lower the harmful impact of this waste on the environment. PAV powder was added to NB in three percentages (10, 20, and 30%) by the weight of NB. The findings indicated that adding PVC to NB produced a notable decrease in penetration and a significant increase in viscosity. This enhancement in the physical properties of NB is due to the chemical compatibility between NB and PVC, which provides more adhesion and cohesion between the two, as both PVC and NB are considered hydrocarbon materials. According to local and international specifications, NB modified with 45% to 60% of PVC is suitable for roofing works such as waterproofing and insulation purposes. Meanwhile, Ahmed et al. [25] used heat treatment to enhance the properties of NB from the Abu Al-Jeer spring to use it in asphalt pavement. NB is heated at a temperature of 163°C for various periods (5, 10, 15, 20, and 25 hours) to improve its physical properties. The results showed improved penetration, ductility, softening point, and other properties. The Marshall test for asphalt mixtures containing NB demonstrated an increase of 17.8% in Marshall stability values compared to asphalt mixtures made from petroleum asphalt, as heat treatment increased the stiffness of NB more than that of petroleum asphalt. To avoid the external heat treatment costs, Ahmed et al. [16] studied the process of mixing NB with petroleum asphalt in different proportions (20, 40, 60, and 80) %. The outcomes of the experimental tests revealed that the combination of NB and refinery asphalt leads to the production of a new asphalt that meets the requirements of the local standard for asphalt used in road construction. The mixtures that contain 80% NB have greater Marshall stability values than traditional mixtures by 23.5%.

In the present research, NB was supplied from five sulphur springs: Al-Mamora, Al-Jabal, Atatt, Al-Atffa, and Al-Askaree, which were located in the city of Heet, Anbar Governorate. Previous studies have been conducted to evaluate the NB from these springs and have focused on its physical properties. Therefore, an extensive and deep analysis is needed to understand the nature of NB produced from these springs. This research has two main aims. The first aim is to investigate the suitability of virgin NB in producing HMA within the specification limits. The second aim is to improve the physical properties of NB by applying a heat treatment for different periods depending on the spring type. Also, the chemical composition and microstructure scanning of treated NB and petroleum asphalt are examined. In addition, the current study assesses NB's ability to improve mechanical properties and resistance to water damage for asphalt pavement. Finally, the experimental results for the virgin and treated NB are compared to those of petroleum asphalt from crude oil refineries.

2. STUDY AREA

The sulphur springs are the primary source of NB in Iraq. The research area comprised many bitumen springs in Heet, inside the Anbar Governorate, approximately 190 km west of Baghdad, Iraq, as shown in Figure 1(a). The city of Heet was situated between longitudes 15°42'-15°43' to the east and latitudes 15°33'-38°34' to the north. Five of the numerous springs in the study region were selected for this study, as indicated in Figure 1(b). The selected springs are Al-Mamora, Al-Jabal, Atatt, Al-Atffa, and Al-Askaree.

https://doi.org/10.31436/iiumej.v26i2.3452

(a) Anbar governorate, Iraq (b) Heet city, Anbar governorate

Figure 1. Sulphur spring sites in Heet city, Anbar governorate.

3. MATERIALS

This project used materials locally in Iraq to achieve economic benefit and take advantage of national resources. The materials are listed in the following sections.

3.1. Petroleum Asphalt

It is derived from the distillation process of crude oil in the Doura refinery and asphalt approved for use in central and southern Iraq for paving AC (40-50), following the specifications of the Department of Roads and Bridges [26].

3.2. Natural Bitumen

Natural bitumen (NB) was provided from five springs in different areas of Anbar Governorate in Iraq. As illustrated in Figure 2, NB extracted from these springs differs in physical and chemical properties from the synthetic asphalt resulting from the destructive distillation of crude oil, whose characteristics are largely controlled by the Doura refinery. NB contains a large water concentration and pollutants, so it cannot be used directly. Therefore, NB must be dried and treated before being used in paving operations.

(a) Al-Mamora spring (b) Al-Jabal spring (c) Al-Atffa spring

(d) Atatt spring (e) Al-Askaree spring

Figure 2. Sulphur springs in Heet city.

3.3. Fine and Coarse Aggregate

The fine and coarse aggregates in this work were provided by the Al-Nibaie quarry. Aggregate retained between sieve No. ¾" and sieve No. 4 are categorized as coarse aggregates. Meanwhile, the aggregate retained between sieve No. 4 and sieve No. 200 is categorized as fine aggregates. In the road laboratory, fine and coarse aggregates were sieved and combined to produce the selected gradation within the limits of the local specification [26], as illustrated in Figure 3. The physical characteristics of fine and coarse aggregates were tested and listed in Table 1.

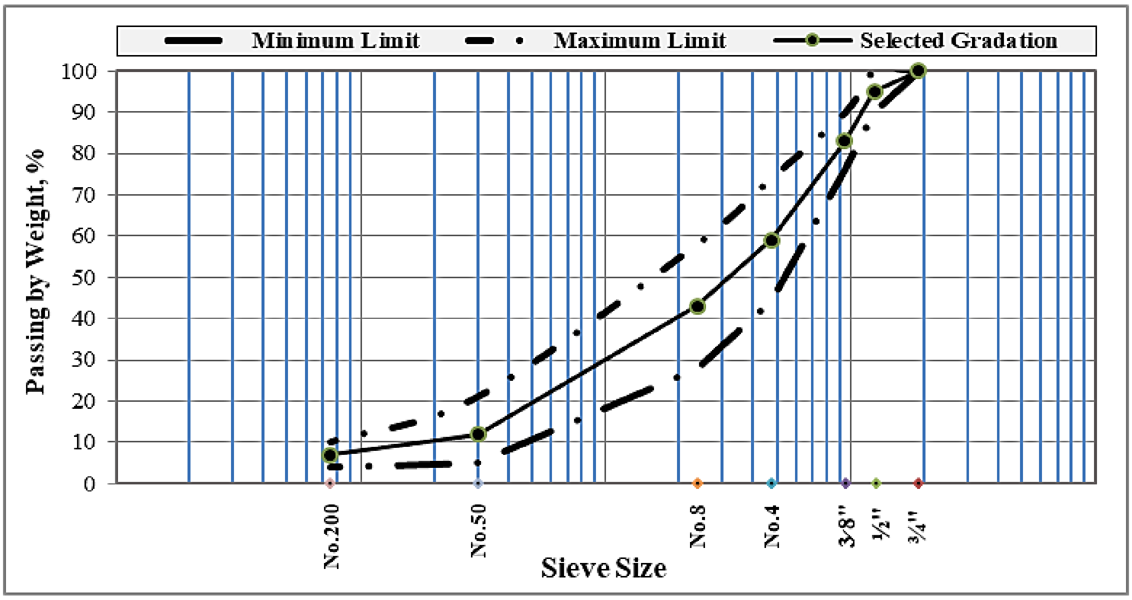


Figure 3. Aggregate gradation curve for wearing layer.

Table 1. Fine and coarse aggregates properties

Property	Test Method	Test Result	Specification Limit [26]
Coarse aggregate			
Wearing (Los Angeles Abrasion), %	ASTM C-131	11.6	30 Max.
Bulk Specific Gravity	ASTM C-127	2.63	-
Water Absorption, %	ASTM C-127	0.19	-
Fractured Pieces, %	ASTM D-5821	96.3	90 Min.
Flat and Elongated Particles, %	ASTM D-4791	3.75	10 Max.
Fine aggregate			
Water Absorption, %	ASTM C-128	0.93	-
Bulk Specific Gravity	ASTM C-128	2.661	-

3.4. Mineral Filler

The limestone filler was prepared from the city of Karbala in southeastern Iraq. The properties of this filler were tested, and the outcomes are presented in Table 2.

Table 2. Limestone properties

Property	Result
Specific Gravity	2.73
Percentage Passing Sieve No. 200 by Weight, %	94

4. WORK PROGRAM

NB's properties were investigated through experimental testing both before and after heat treatment. In addition, many tests were applied to investigate the suitability of using the treated NB as a binder material in preparing HMA instead of petroleum asphalt.

4.1. Natural Bitumen Processing

Drying and isolating the NB extracted from the sulphur springs is the initial action in the treatment process. This is accomplished by heating the NB in a convection oven to a temperature of 110 °C for a variable amount of time, depending on the spring's type and water content, while keeping in mind that asphalt does not burn [27]. The second stage of treatment involves heating the NB in a convection oven to a temperature of 163 °C, with the required time varying for each sample, depending on the confirmation that the bitumen has been separated from the water and making it conform to the local specifications [26]. NB is obtained from five springs and treated as follows:

4.1.1. Al-Mamora Spring (MS)

Al-Mamora Spring is located on the Heet-Kabisa road. Because of its liquid nature and high water content, the NB recovered from it requires two hours to dry in an oven at 110 °C. Next, to align with the properties of asphalt AC (40-50) used in paving operations, following the local specification [26], the NB is heated in a convection oven at 163 °C for 17 hours, divided into periods of 5, 10, 15, and 17 hours.

4.1.2. Al-Jabal Spring (JS)

Al-Jabal Spring is in the Jabal neighborhood of Heet-Anbar, also known as Ein Al-Khader. The liquid NB contains a significant amount of water, which is removed by drying in the oven at 110 °C for approximately two and a half hours. Then, it is equipped with heating in the oven at 163 °C for 25 hours and separated into periods ranging from 5 to 25 hours to match the asphalt used for pavement operations according to the local specification [26].

4.1.3. Atatt Spring (TS)

Atatt Spring is located in Heet-Anbar, close to the Al-Mamora Spring. The drying process for the NB and removing water takes two hours in an oven at 110 °C. The heat treatment lasts 21 hours at 163°C, with the time divided into periods 5, 10, 15, 20, and 21 hours, ensuring compatibility with the local specification [26].

4.1.4. Al-Atffa Spring (AS)

Al-Atffa Spring is located in Heet-Anbar, in the fire pit neighborhood. Drying the NB from the water took an hour and a half in an oven at 110 °C because the bitumen extracted from this spring is solid and has a low degree of penetration compared to other sulphur springs. The heat treatment takes two hours to meet the limits of the local specification [26].

4.1.5. Al-Askaree Spring (SS)

Al-Askaree Spring is located in the Askaree neighborhood of Heet-Anbar. It is also called the Siali Spring, as the NB found here is of the Siali type, which is very liquid and has a high degree of penetration. The drying process took two and a half hours in an oven at 110 °C to get rid of the water and separate the bitumen. The NB was then treated by heating for 26 hours, divided into periods of 5, 10, 15, 20, 25, and 26 hours, to conform to the local specification [26].

4.2. Asphalt Cement Tests

As shown in Table 3, many tests were conducted on petroleum asphalt, untreated NB, and treated NB from the five springs, heated for varying durations according to each sample.

Table 3. Asphalt binder tests

Test	Methods
Penetration	ASTM D-5
Ductility	ASTM D-113
Softening Point	ASTM D-36
Flash Point	ASTM D-92
Specific Gravity	ASTM D-70

4.3. Scanning Electron Microscopy-Energy Dispersive X-Ray Analysis (SEM-EDX)

The chemical composition of bitumen's molecular components significantly impacts the material's internal structure. Bitumen is a complex blend composed mainly of hydrocarbon compounds, with some structurally similar heterocyclic species, as well as functional groups comprising oxygen, nitrogen, and sulphur atoms. In addition, bitumen contains trace amounts of metals such as vanadium, nickel, iron, aluminium, and calcium, which exist as inorganic salts, oxides, or porphyrin complexes.

Energy dispersive X-ray analysis (EDX) in Figure 4 is an analytical technique used to analyze a sample's elements or chemical characterization based on the interaction of specific sources. The device is manufactured in the Netherlands. The EDX device's operation principle analyzes the X-rays emitted from the sample when the electron beam collides and interacts with the sample atoms. Each element has a distinct X-ray energy, and the EDX detector records the emitted X-rays, distinguishing the elements present and their concentrations. This technique is used to study environmental pollution, where EDX microanalysis can greatly detect heavy metal pollution [28].

In addition to these detectors, modern SEM tools are equipped with an inbuilt EDX sensor, which allows for compositional analysis of materials, called scanning electron microscopy (SEM) with EDX spectroscopy (SEM-EDX). EDX testing can assist road engineers in understanding the physical-chemical behavior of asphalt pavements through chemical element analysis. On the other hand, SEM images can provide information about elemental analysis in conjunction with EDX, as they help analyze the surface and interactions and understand the surface properties of compounds. Also, SEM-EDX testing helps to examine changes occurring at the atomic and molecular levels [29]. The principle of operation of SEM involves generating a beam of electrons directed to the sample through a column containing electromagnetic lenses. The electrons then interact with the surface of the sample. As a result of this interaction, different signals are emitted. The detectors collect these resulting signals to generate three- or two-dimensional images with fine details, such as surface composition.

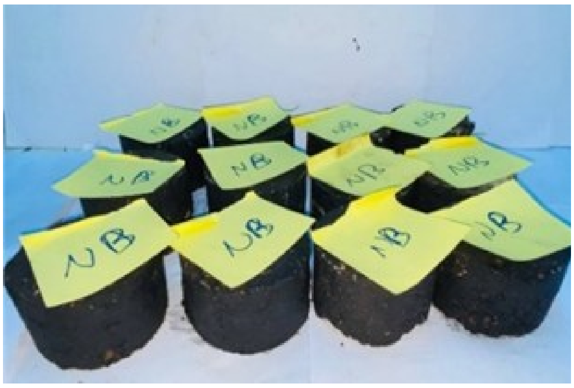
Figure 4. SEM-EDX device.

4.4. Marshall Test

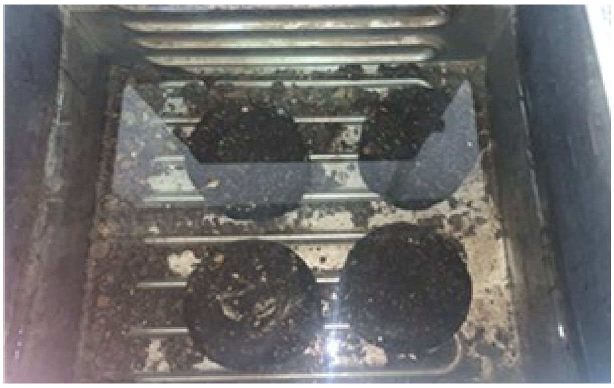
As specified in ASTM D6927-15, the Marshall test is employed to arrange, compress, and verify the samples. The asphalt and aggregate are heated separately for a couple of hours, with temperatures reaching 135 °C and 150 °C, respectively. Avoiding excessive heating and preheating of the asphalt is essential, as this can alter its properties and negatively affect the overall mixture. According to the local specification [26], the asphalt binder content for the wearing layer must be between 4% and 6%. Therefore, five asphalt proportions with a constant increment of 0.5% (4.0%, 4.5%, 5.0%, 5.5%, and 6.0% based on the aggregate weight) are combined with the heated aggregate to find the optimum binder content (OBC). The mixture must be agitated for at least three minutes to ensure consistent application of the asphalt cement on every aggregate particle.

The conventional cylindrical Marshall test moulds possess a diameter of 102 mm and a height of 64 mm. Before use, these moulds are heated to a temperature of 130 °C. The mould is then filled with the heated mixture and compressed using a Marshall hammer, applying 75 impacts from each side for heavy traffic roads. The compressed Marshall sample is permitted to cool. After a duration of 24 hours, the mould is unsealed, and the specimen is immersed in a water bath at a temperature of 60 °C for a period of 30 minutes before examination.

The mixture characteristics of petroleum asphalt and NB from the five springs post-treatment are evaluated. These characteristics include flow value, Marshall stability value, air void ratio, asphalt-filled voids, voids in mineral aggregate, and OBC. Figure 5 illustrates the detailed Marshall test procedures.



(a) Specimen preparation



(b) Specimens in a water bath

(c) Specimen testing

(d) Fractured specimens

Figure 5. Marshall test.

4.5. Moisture Damage Test

Moisture damage refers to the progressive decline in the performance of the pavement mixture caused by the weakening of the adhesive link between the bitumen and the surface and the reduction in cohesive resistance. Moisture leads to a loss of adhesion between the bitumen surfaces and aggregates. Consequently, the mixture undergoes separation, resulting in a decline in stability, strength, and hardness due to reduced cohesion.

The compacted mixtures' tensile strength ratio (TSR) was tested according to ASTM D 4867 M-96 to evaluate their susceptibility to water harm. This test is indicated to determine whether water reduces the adhesion between asphalt particles and aggregate. To achieve this,

https://doi.org/10.31436/iiumej.v26i2.3452

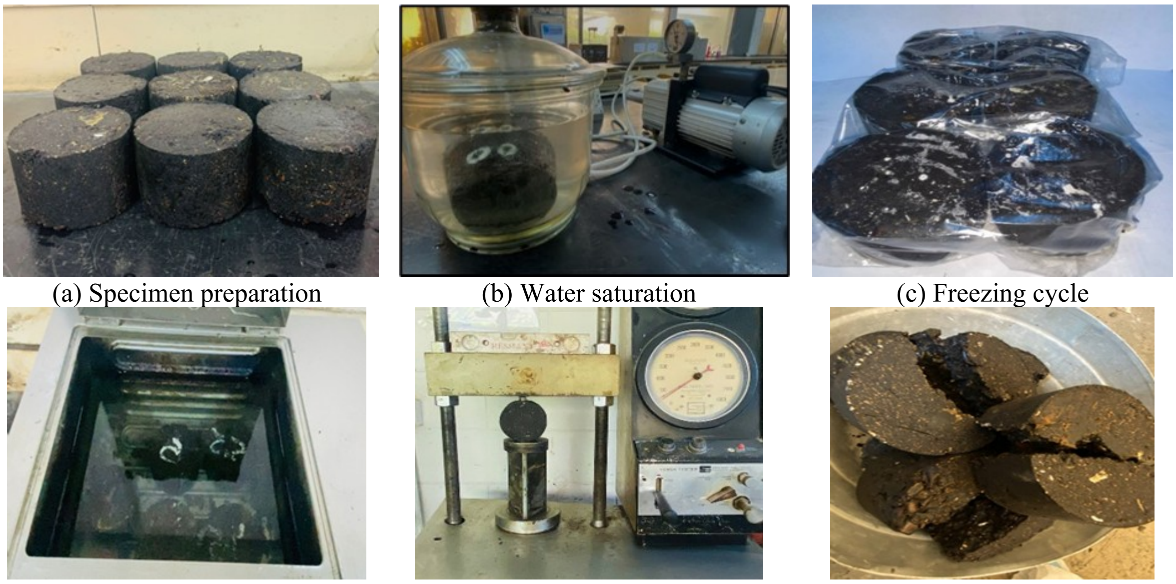
for the petroleum asphalt and NB after undergoing specific treatment durations for each spring, Marshall samples were prepared using the optimum binder content (OBC). These samples were categorized into two groups: the unconditioned group, which was placed in a water bath at a temperature of 25 °C for 20 minutes, and the conditioned group, which underwent one freeze-thaw cycle followed by immersion for one hour at the same temperature. The minimum requirement for HMA (Hot Mix Asphalt) TSR is more than 80%. TSR is calculated using the following equation:

$$TSR = \frac{IDT_{wet}}{IDT_{dry}} \times 100 \quad (1)$$

$$IDT = \frac{2000 \times P}{\pi t D} \quad (2)$$

where TSR is the tensile strength ratio (%), IDT_{wet} is the average indirect tensile strength of the conditioned groups-wet (kPa), IDT_{dry} is the average indirect tensile strength of the unconditioned groups-dry (kPa), P is the peak load, t is the specimen thickness (mm), and D is the specimen diameter (mm).

Figure 6 shows the procedures for conducting the TSR test as stated previously. Each specimen was tested by applying a rate of 50.8 mm per minute. After the load reached its maximum value, the specimen was fractured entirely, and this load was recorded.



(a) Specimen preparation (b) Water saturation (c) Freezing cycle

(d) Thawing cycle (e) Specimen testing (f) Fractured specimens

Figure 6. Moisture damage test.

5. RESULTS AND DISCUSSION

5.1. Asphalt Properties

The physical properties of untreated NB derived from springs and petroleum asphalt are shown in Table 4. Physical testing shows that petroleum asphalt meets all requirements stipulated in the local specifications [26], including penetration, flash point, ductility, and other tests. Compared to petroleum asphalt, the test results reveal that NB has a much higher penetration value and does not meet the specification requirements [26]. It needs to be treated to improve its qualities and make it conform to the local specifications for paving asphalt.

https://doi.org/10.31436/iiumej.v26i2.3452

Table 4. Physical properties of petroleum, asphalt, and untreated NB

Test	Results						Specification Requirements [26] for AC (40-50)
	Petroleum Asphalt	MS-NB	JS-NB	TS-NB	AS-NB	SS-NB	
Penetration, 1/10 mm	42	130	143	120	58	142	40-50
Ductility, cm	>100	68	63.7	66.7	65	64	>100
Softening point, ^{0}C	53.5	39	35	43.2	42.5	36.2	-
Flashpoint, ^{0}C	324	180	158	175	175	175	232 min
Specific Gravity	1.02	1.04	1.052	1.03	1.028	1.04	-

Table 5. Physical properties of treated NB

Test	Heating Period in Hours							Specification Requirements [26] for AC (40-50)
	0	5	10	15	17	-	-	
MS-NB Results								
Penetration	130	95	72	58.6	45	-	-	40-50
Ductility	68	82.9	94	>100	>100	-	-	>100
Softening point	39	45	48.7	50.4	52	-	-	-
Flash point	180	216	224	240	248	-	-	232 min
Specific Gravity	1.04	1.0415	1.042	1.043	1.0431	-	-	-
JS-NB Results								
	0	**5**	**10**	**15**	**20**	**25**	-	
Penetration	143	97	88.8	72	66	44	-	40-50
Ductility	63.7	75	80	88	>100	>100	-	>100
Softening point	35	41	46.7	48	51	53.5	-	-
Flash point	158	183	197	213	224	232	-	232 min
Specific Gravity	1.052	1.0524	1.0528	1.053	1.0532	1.054	-	-
TS-NB Results								
	0	**5**	**10**	**15**	**20**	**21**	**-**	
Penetration	120	83	72.6	65	50.8	44	-	40-50
Ductility	66.7	74	86	91	>100	>100	-	>100
Softening point	43.2	44.6	47	48.7	51	53.5	-	-
Flash point	175	188	190	210	230	240	-	232 min
Specific Gravity	1.03	1.031	1.0316	1.032	1.0323	1.0327	-	-
AS-NB Results								
	0	**2**	**5**	**-**	**-**	**-**	**-**	
Penetration	58	45	40	-	-	-	-	40-50
Ductility	65	>100	>100	-	-	-	-	>100
Softening point	42.5	52.3	54	-	-	-	-	-
Flash point	175	238	242	-	-	-	-	232 min
Specific Gravity	1.028	1.03	1.033	-	-	-	-	-
SS-NB Results								
	0	**5**	**10**	**15**	**20**	**25**	**26**	
Penetration	142	115	90.6	83	69	52	45	40-50
Ductility	64	70	80	88	97	>100	>100	>100
Softening point	36.2	40	43	47.6	50.1	53.5	56	-
Flash point	177	187	198	213	224	232	245	232 min
Specific Gravity	1.04	1.042	1.0426	1.043	1.0434	1.044	1.0441	-

Table 5 displays the physical characteristics of NB after treatment and reveals improvements in these properties. MS-NB is classified as liquid, with a penetration of 130 (0.1 mm). Also, it requires a heat treatment period of about 17 hours. Heating the asphalt for a longer period, for example, 20 hours, leads to poor results and increases its hardness due to a high rate of oxidation, which in turn affects the asphalt mixture and causes issues. Therefore, the ideal penetration rate was 45 (0.1 mm) after just 17 hours. Meanwhile, JS-NB has a high degree of penetration and was treated for 25 hours, after which the penetration became 44 (0.1

mm). This is within the limits of the local specifications [26] and is close to the penetration of the petroleum asphalt. Continuous mixing and heating cause the oxidation of all parts of the asphalt, which alters its properties, making it harder and giving it a dark black color. Thus, this affects the mechanical performance of the asphalt mixture.

In addition, the heating treatment for TS-NB took a total of 21 hours; if the treatment were carried out for 25 hours, the degree of penetration would be very low and not meet the limits of the standard specification [26]. At 21 hours, the penetration was 44 (0.1 mm), which complies with the specification because the heat increases the softening point of asphalt and enhances its hardness. Moreover, AS-NB is considered the best of the five types of NB because the bitumen extracted from this spring is considered solid, with a low penetration of 58 (0.1 mm), and contains a very small amount of water. Therefore, the treatment period was only two hours, the penetration became 45 (0.1 mm), and the ductility was greater than 100 cm. Therefore, this bitumen type will increase the resistance against cracking formation and extend the lifespan of the asphalt mixture. It conforms to the specification limits, and the improvement in properties is attributed to the heating process. The longer the heat treatment, the higher the percentage of sulphur, which improves the properties of asphalt.

Due to its fluid nature and high water content, SS-NB took a long time to dry. The longest heat treatment period required 26 hours to reach the required degree of penetration, which is 45 (0.1 mm). The material has a ductility exceeding 100 cm, a softening point of 56 °C, and a flash point of 245 °C, all meeting the specification limits.

The NB of Abu Al-Jeer spring was treated to improve the physical properties through the same heat treatment process for 25 hours, as conducted in the previous study by Ahmed et al. [25]. This is approximately compatible with the treatment time in this study for NB from JS and SS. In addition, Altameemi et al. [27] treated NB-MS by applying heat treatment for 20 hours to enhance its properties, which is not consistent with the treatment period in the current study, which is 17 hours. The variance in treatment periods for the same spring may be related to changes in environmental conditions over the years, especially temperature fluctuations and rainfall rate reductions, which lead to the production of different NB.

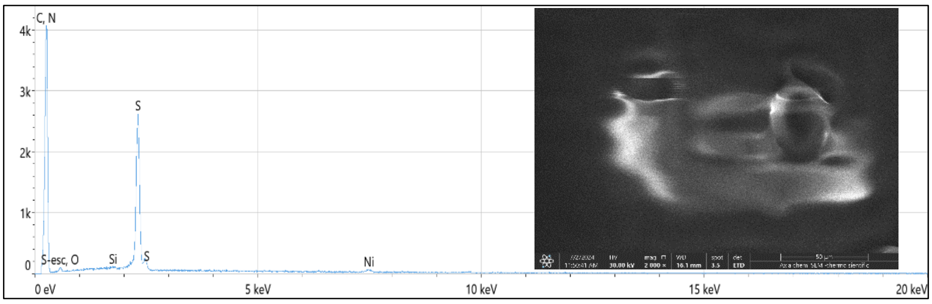
5.2. Morphology and Element Analysis Properties

Hydrocarbons, the main component of asphalt, have paraffinic, naphthenic, and aromatic structures. They also include nitrogen, sulphur, and cyclic or non-cyclic oxygen molecules. In addition, atoms of nickel, iron, and vanadium are present. Compounds containing sulphur, nitrogen, and oxygen react with these elements. Various component fractions affect bitumen compatibility and durability. However, the balance of these components gives bitumen its unique viscoelastic characteristics, which are crucial for its use as an asphalt binder in paving works. Thus, a lack of balance or compatibility between components leads to component phase separation and undesirable characteristics [30]. Minerals significantly impact the physical properties of asphalt because non-polar atoms cause molecular interference, which alters the material's solubility, boiling temperature, and viscosity. The SEM-EDX analysis for petroleum asphalt and treated NB is shown in Figure 7.

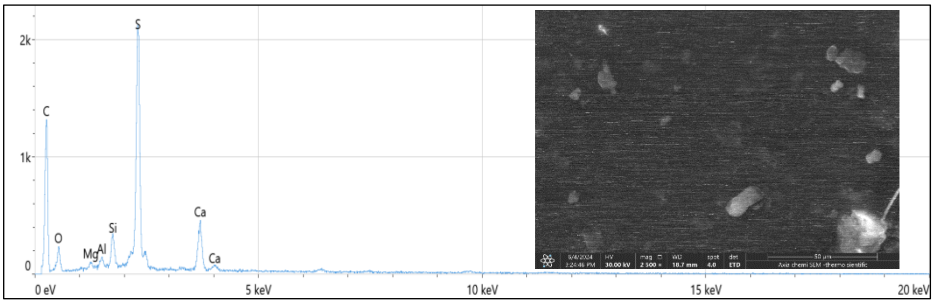
The microstructure of the asphalt binder significantly affects its properties, as this microstructure is closely linked to asphalt chemical composition [31]. It explains the changes that occur in asphalt due to chemical and environmental factors and helps detect defects and impurities within the asphalt that affect its properties and quality. It also helps determine the components' distribution, their homogeneity with the asphalt when applied, and their effect on performance. From the SEM images, petroleum asphalt is a mass of homogeneous hydrocarbons, and the surface of this asphalt appeared primarily plain and clear of any noticeable features in surface morphology. This is because the chemical composition of petroleum asphalt is less diverse in its compounds than that of NB samples. It can be seen that SS-NB is approximately similar to petroleum asphalt in morphology structure compared to NB from other springs. This may be due to its high carbon content (88.7%) and lower component diversity, with no trace elements in this NB. The percentage of chemical components in petroleum asphalt and NB (after treatment) for each of the five sulphur springs is illustrated in Figure 7 and listed in Table 6. Evaluating the structure information and molecular type is important for a fundamental understanding of how the element compositions affect the chemical reactivity and physical properties of bitumen.

Besides sulphur, most of the NB from sulphur springs consists of organic components such as carbon and oxygen, small amounts of trace components such as iron, magnesium, and silicon, and other contaminants. Also, NB is free of toxic elements such as lead, mercury, and fluoride, which is advantageous for environmental preservation.

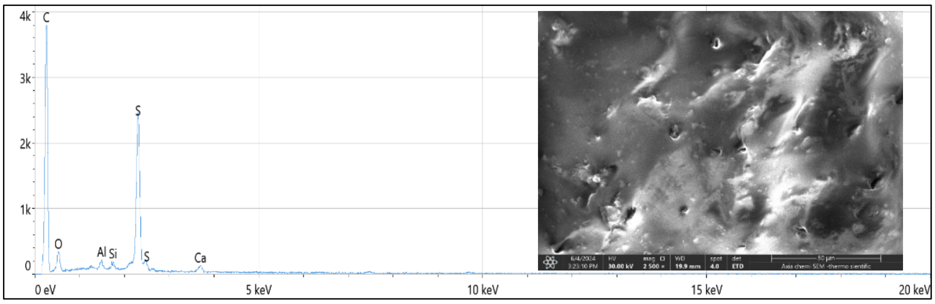
SS-NB had the highest carbon content, at 88.7. Increasing carbon has a positive effect on the asphalt mixture, improving its flexibility, which decreases the failure stress of asphalt at low temperatures. This is because carbon improves asphalt properties, especially anti-aging behavior, and thermal and electrical conductivity, enhancing the behavior of asphalt roads under different climatic conditions. Also, it enhances the asphalt's ability to resist deformation at high temperatures and cracking under cold conditions, ultimately improving the durability of the asphalt [32]. Asphalt contains sulphur, nitrogen, and oxygen, known as heteroatoms that encourage reactions in materials. Compared to the hydrocarbon moiety, heteroatoms are a minor component, and their concentrations are not constant and vary depending on the source of bitumen. This is attributed to heteroatom moieties generally imparting polarity and functionality to the molecules, contributing to differences in the physical properties between asphalt binders supplied from different sources. Furthermore, sufficient amounts of these heteroatoms made the hydrocarbon molecular structures more complicated; usually, one or more heteroatoms per molecule may be present [33].



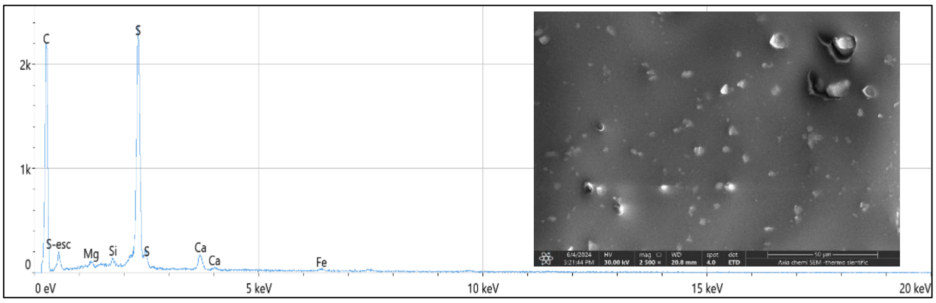
(a) Petroleum Asphalt



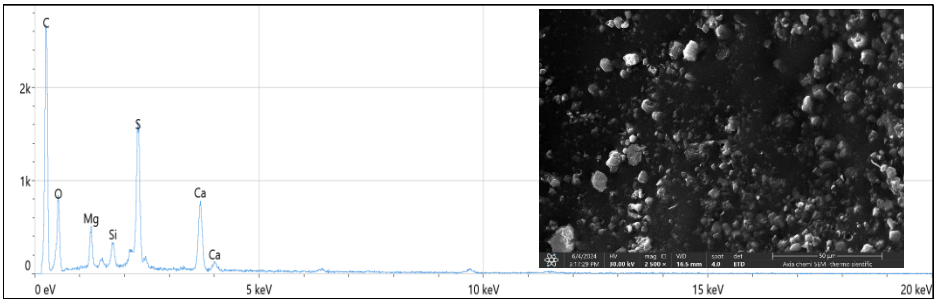
(b) MS-NB



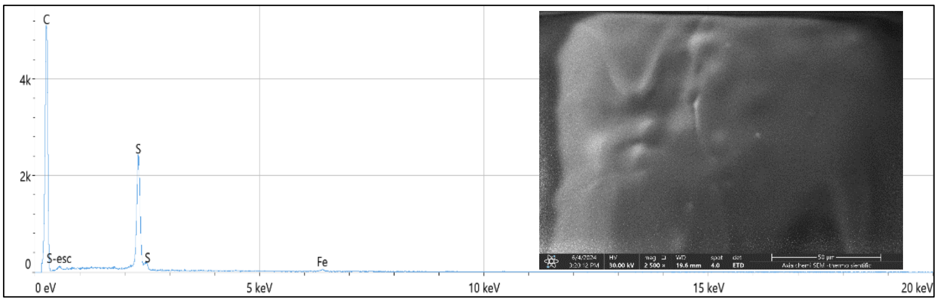
(c) JS-NB



(d) TS-NB



(e) AS-NB



(f) SS-NB

Figure 7. SEM-EDX analysis for petroleum asphalt and NB from five sulphur springs.

For each of the six NB samples, the percentage of sulphur in Table 6 varied and was not the same. AS-NB had the lowest content (6.2%), while TS-NB had the highest sulphur content (16.4 %). The high density of the sulphur elements increases the density of the binder, which, in turn, increases the service life of asphalt pavement and saves costs. For more clarification, sulphur reacts with indole to form polysulfides. It leads to a change in its colloidal structure, which leads to an increase in the proportion of asphaltenes/resins, which causes the structure to change to a more stable and cohesive gel structure, increasing the density. Additionally, the softening point rises with increased sulphur content, and this outcome aligns with the results of the previous study [34]. NB from the five springs does not contain nitrogen and nickel elements in its structure, compared to petroleum asphalt. Nitrogen increases the affinity of asphalt with aggregate, improving the resistance of the asphalt mixture against weathering, especially stripping and oxidation [25]. Meanwhile, nickel is one of the trace elements, and when heating the asphalt, higher concentrations of nickel can cause higher emissions of pollutants [35]. Furthermore, the presence of calcium and magnesium in NB positively affects its properties, as it increases hardness and improves stability [36].

Table 6. Elemental composition of petroleum asphalt and treated NB

Asphalt Type	Element, % *									
	C	**O**	**S**	**Si**	**Fe**	**Ca**	**Mg**	**Al**	**N**	**Ni**
Petroleum Asphalt	83.4	1.2	11.7	0.1	-	-	-	-	2.6	1.0
MS-NB	62.0	16.5	14.2	1.8	-	4.5	0.4	0.6	-	-
JS-NB	71.6	16.2	10.6	0.4	-	0.7	-	0.5	-	-
AS-NB	54.4	31.2	6.2	1.2	-	4.6	2.4	-	-	-
SS-NB	88.7	-	10.8	-	0.5	-	-	-	-	-
TS-NB	80.4	-	16.4	0.4	0.6	1.8	0.4	-	-	-

* Carbon (C), Oxygen (O), Sulphur (S), Silicon (Si), Iron (Fe), Calcium (Ca), Magnesium (Mg), Aluminium (Al), Nitrogen (N), and Nickel (Ni)

5.3. Marshall Properties

The OBC is 5% for a conventional mixture prepared with petroleum asphalt. After applying heat treatment to the NB samples, the OBC values are 5.2%, 4.9%, 5.3%, 4.93%, and 5.1% (by weight of the aggregate) for mixtures prepared with NB from AS, JS, MS, SS, and TS, respectively. The Marshall properties of both combinations (conventional mixture and NB mixtures) are shown in Figures 8 and 9. In the Marshall test, flow and stability are the most important performance indicators of asphalt pavement.

The results in Figure 8 showed that the stability of NB mixtures, except for the JS-NB mixture, was better than that of the conventional mixture. The results were (13, 10.8, 12.6, and 11.8) kN for mixtures with NB from MS, AS, TS, and SS, respectively. In contrast, the stability of the conventional mixture was 9.2 kN. Compared to the conventional mixtures' stability, NB mixtures' stability increased by 17.3%, 28.2%, 36.9%, and 41.3% for NB from AS, SS, TS, and MS, respectively. This might be because NB contains a high percentage of sulphur, which increases the hardness of asphalt by raising the temperature between 112 and 170 °C. Sulphur consists of chemical chains that bind with the organic molecules of the compound, thus forming polysulfates [16, 25, and 37]. However, the stability of the JS-NB mixture is 8.8 kN, which is 4.4% less than the stability of the conventional mixture. This may be because JS-NB is more affected by high oxidation rates than other NB samples. Generally, all mixtures' stability values meet the specification limits (8 kN minimum limit for wearing layer) [26].

Figure 9 shows the Marshall flow values of the conventional and NB mixtures, according to the Marshall test outcome. The flow values are close, except for the MS-NB mixture, which increases by 12.1% compared to the conventional mixture flow. According to the specification limits, the flow should range between 2 and 4 mm, and all the mixtures comply with this requirement.

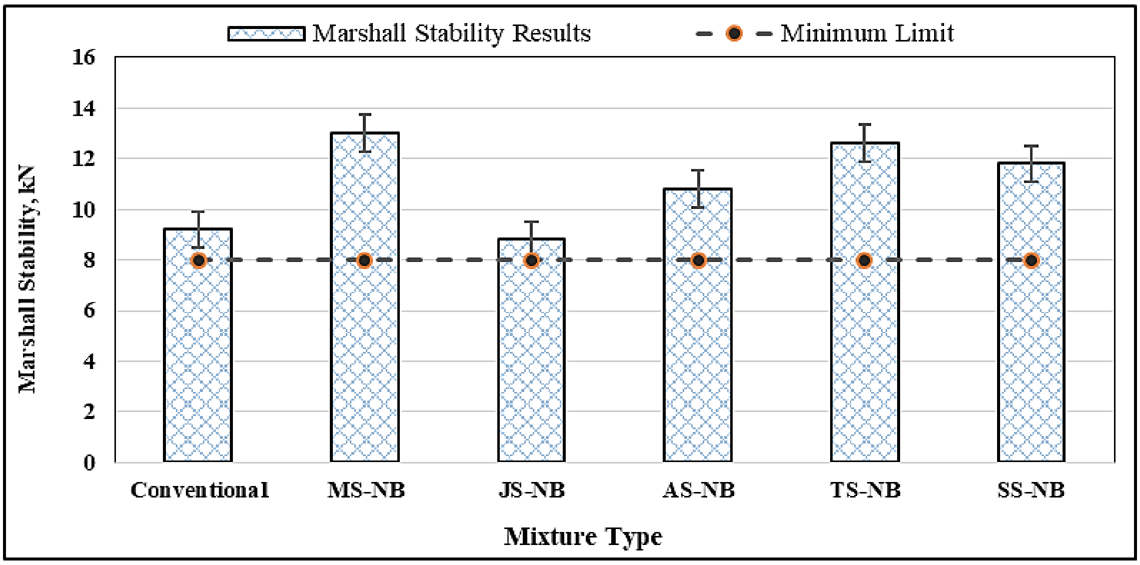


Figure 8. Marshall stability results.

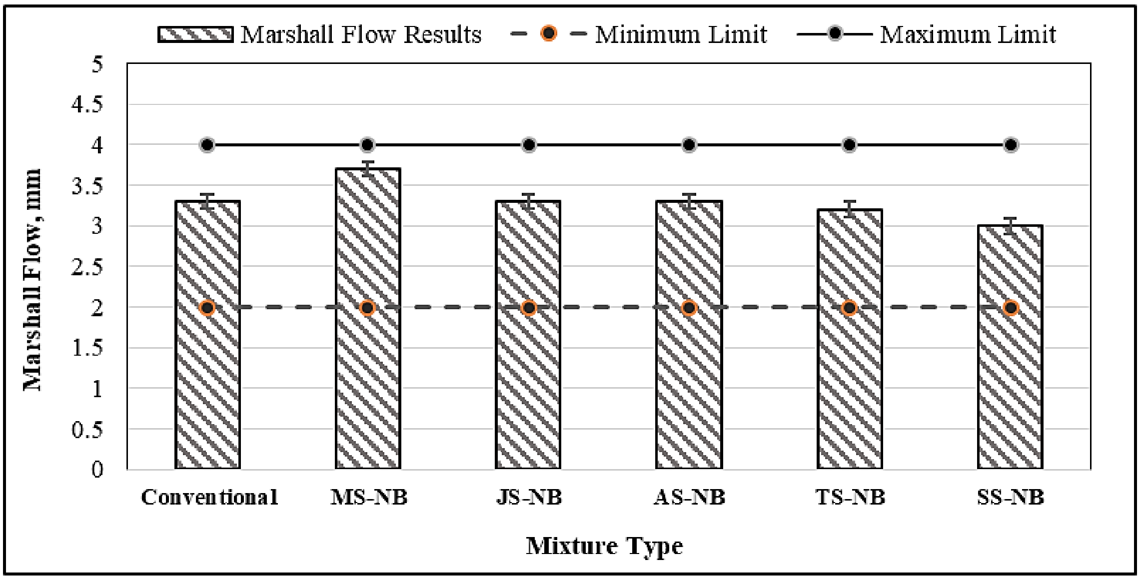


Figure 9. Marshall flow results.

Figure 10 shows conventional and NB mixtures' Marshall Quotient (MQ), or stiffness index. MQ refers to the proportion of Marshall stability to Marshall flow and is considered an indicator of the asphalt mixture's rutting resistance. Higher MQ values indicate better stability and more resistance to deformation. Therefore, an increase in MQ means the stability of the asphalt mixture has improved regarding its flow behavior. This is desirable for the durability of high-quality asphalt pavements [38].

The volumetric properties for conventional and NB mixtures are listed in Table 7. Based on local specifications [26], the air void (Vv) should be between three and five percent. An increase in voids leads to rutting of the road, rapid oxidation of the asphalt, and higher permeability, which leads to problems that shorten the road's lifespan. A decrease in the percentage of voids can also lead to creep and eruption of asphalt [39]. The Vv % in conventional and NB mixtures are similar, and both fall within the limits of the local specification.

The performance of a mixture depends on the voids in mineral aggregate (VMA), which should be sufficiently high to ensure adequate bitumen content to meet durability requirements. However, as VMA increases, the mixture becomes more susceptible to stability issues. The results indicated that the VMA values for NB mixtures slightly differ from those of conventional mixtures.

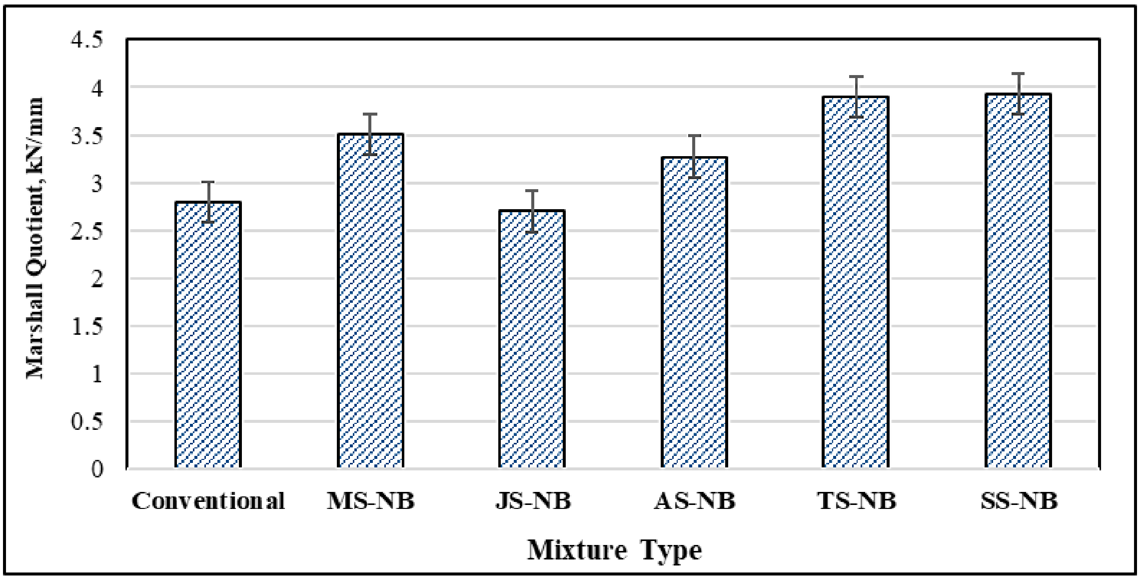


Figure 10. Marshall quotient results.

Table 7. Volumetric properties for conventional and NB mixtures

Property		Bulk Density g/cm^3	Vv (%)	VMA (%)	VFB (%)
Conventional Mixture		2.323	3.8	16.4	76.8
NB-Mixture	MS	2.340	3.6	16.1	77.6
	JS	2.330	4.0	16	75
	AS	2.325	3.5	16.5	78
	TS	2.335	3.4	16.1	78.9
	SS	2.332	3.6	16.1	77.6
Specification Requirements [26]		-	3-5	14 Minimum	-

The voids filled with binder (VFB) refer to the distances among aggregate particles in a compacted mixture filled with bitumen. These voids impact the durability of the mixtures. The findings reveal that VFB percentages for conventional and NB mixtures are either similar to or lower than conventional values, except for TS-NB, which showed a 2.7% increase compared to the conventional mixtures.

5.4. Moisture Damage Resistance

As a result, it is necessary to recognize the harmful effects of water and humidity and take measures to mitigate them, while also ensuring the mixture's hardness, durability, and resilience under various conditions [40]. TSR can be defined as a coefficient of moisture durability index, and its value should be at least 80%. TSR depends on the adhesion between the aggregate surface and bitumen and the cohesion properties of the asphalt binder. Cohesion refers to the overall integrity of the material when subjected to load and stress, and it is influenced by several factors, including the viscosity and chemical components of the asphalt binder, attraction inside the asphalt binder, and water penetration. Water can affect cohesion by saturating and expanding the system of voids due to freezing and thawing cycles. Asphalt adhesion to water can be affected by separation or displacement mechanisms.

Adhesion depends on the aggregate gradation, Vv, and permeability. Adhesion under moisture circumstances, adhesion may vary if any of these parameters change [41].

Figure 11 displays the TSR values for conventional and NB mixtures. The test findings show that the NB mixture had the highest resistance to moisture and water conditions. The mixture with SS-NB exhibited 5.72% greater resistance to moisture damage than the conventional mixture. This may be attributed to the large amount of carbon in SS-NB, since the high content of carbon elements increases the adhesion properties of the asphalt binder, which increases durability and provides better resistance to moisture damage. Specifically, carbon plays a crucial role. It is present in high concentrations and forms chains that help create strong bonds within the NB structure, increasing the stability of the asphalt and its ability to adhere to the aggregate. Furthermore, asphalt with high carbon content is an absorbent material, which helps trap moisture and enhances the performance and moisture resistance of the asphalt [32].

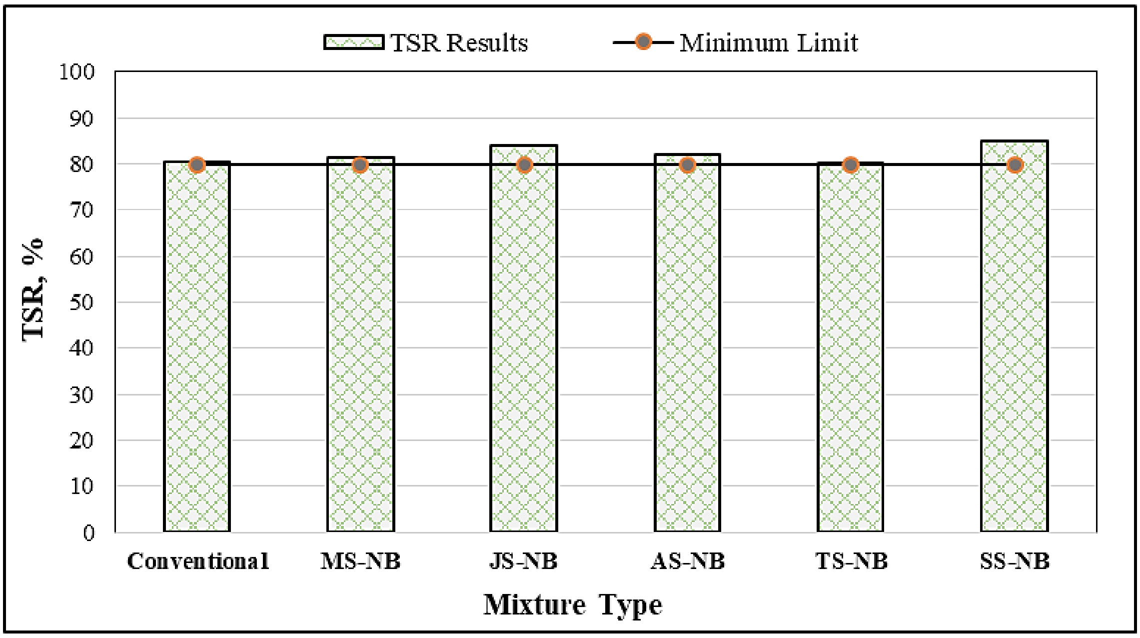


Figure 11. TSR for conventional and NB mixtures.

6. COST ANALYSIS

Developing innovative processes and product models that simultaneously deliver economic, technical, and environmental benefits is essential to ensure that policies effectively target sustainable development goals. Environmentally friendly and more economical alternatives must be discovered to achieve sustainable development. NB is considered one of the natural resources available in abundance in Iraq, and it can be exploited across various governorates, from north to south. Economic gains can be achieved by investing in NB or incorporating it into road paving operations. This is because NB is more economical than petroleum asphalt, which is derived from the distillation processes of crude oil, and it is a national resource. Oil, a major energy source, represents over 30% of global consumption. Countries such as Venezuela, Saudi Arabia, Canada, Iran, Iraq, and Russia possess large proven oil reserves. During the distillation process in oil refineries, around 10% of crude oil is converted into asphalt materials and other products through asphalt manufacture. This asphalt produces various retail products, including asphalt emulsions, paving mixtures, paints, tar roofing, and other products [42].

Due to the continuous change in oil prices over the past six decades, which affects asphalt prices, asphalt costs have increased by a factor of 20 due to oil refining operations. Asphalt prices follow crude oil prices; consequently, the quantity and quality of asphalt are impacted. For every 1% increase in crude oil, asphalt prices rise by 0.7%. The decisions made by the Organization of the Petroleum Exporting Countries (OPEC), which consists of 12 countries, have played an important role in determining the cost of asphalt [25, 43]. Some specialists have stated that the oil peak occurred between 2010 and 2020 and will decrease over the coming years due to the decrease in production quantities, the occurrence of a crisis at the global level, and a significant rise in prices. Meanwhile, geologist Marion King Hubbert, in the 1950s, predicted that the global productivity peak would occur between 2007 and 2037, according to estimates from primary energy sources, after which production would decline to less than a third of its previous levels. Therefore, the price of oil has a significant impact on asphalt production and its prices, prompting road engineers to study alternative synthetic asphalt materials to use instead that are more economical and environmentally friendly [44]. The environment is given serious attention during the asphalt production process. There must be strict regulations on emissions from asphalt producers and oil refineries. Reducing carbon emissions during the transportation and production of asphalt pavement has become a global priority. Finding more environmentally friendly natural materials is essential to managing and minimizing greenhouse gas emissions as much as possible. The use of low-carbon technology has become vital to achieving this goal. For this reason, there is an urgent need for future investigations to study the carbon emissions rate and the economic benefits of using NB in paving works.

7. CONCLUSIONS

This study conducted a series of experiments on natural bitumen (NB) derived from sulphur springs, petroleum asphalt, and their mixtures to evaluate the suitability of using NB in hot asphalt mixtures. Physical tests revealed that untreated NB does not meet the local Standard Specification limits, making it unsuitable for direct use in paving. However, when NB is properly dried and heat-treated at 163 °C for a duration dependent on the characteristics of each spring and the penetration level, its properties significantly improve. Overheating beyond 163 °C is discouraged due to potential degradation of performance. Post-treatment physical and chemical analyses showed enhanced characteristics, including reduced penetration, increased ductility and softening point, and improved flash point, bringing NB's properties closer to those of petroleum asphalt. Morphological observations highlighted that the surface microstructure of petroleum asphalt closely resembles that of SS-NB, differing notably from other NB types due to variations in elemental composition. Treated NB demonstrated higher stability than petroleum asphalt, likely due to its elevated sulphur content, which enhances mixture hardness and durability, with calcium and oxygen contributing further to performance. Nonetheless, the JS-NB mixture did not significantly improve Marshall stability due to its high oxidation rate, but did show a slight enhancement in water damage resistance with a 4.35% increase. Notably, the SS-NB mixture achieved the highest tensile strength ratio (TSR) at 85%, surpassing the conventional mixture by 5.7% and exceeding the 80% minimum threshold, indicating NB's strong potential in mitigating moisture damage. In conclusion, NB is a valuable and promising alternative for improving asphalt mixture performance. Economically, it is highly advantageous, being widely available nationwide and costing at least five times less than petroleum asphalt. As a national asset, NB warrants further economic and engineering research to optimize its use in infrastructure applications.

8. RECOMMENDATIONS

Future studies in several key areas may provide deeper insights and a more comprehensive understanding of the current findings. Further investigation is needed to evaluate the intrinsic properties of natural bitumen (NB), including its asphaltene origin, extraction process, and influencing factors such as sediment age, extraction depth, and proximity to sulphur sources. Environmental variables—such as rainfall, temperature fluctuations, and the geographical location of the spring—may also significantly impact NB's characteristics. Additional studies are encouraged to explore alternative treatment methods, such as enhancing NB with inexpensive, readily available materials like fillers or utilizing solar energy by exposing NB to sunlight over extended periods and periodically testing its properties for changes. Furthermore, it is crucial to assess the rutting resistance of NB mixtures under repeated loading and elevated temperatures and evaluate their fatigue cracking behavior to ensure performance under real-world conditions. From a sustainability perspective, economic and environmental assessments of NB usage should be conducted to determine its cost-effectiveness and environmental impact as a substitute for petroleum asphalt, including strategies to minimize emissions associated with its application.

REFERENCES

[1] Ng CP, Law TH, Jakarni FM, Kulanthayan S. (2019) Road infrastructure development and economic growth. IOP Conference Series: Materials Science and Engineering, 512(1):12045. https://iopscience.iop.org/article/10.1088/1757-899X/512/1/012045.

[2] Latief RH. (2019) Evaluation of the performance of glasphalt concrete mixtures for binder course. International Journal on Advanced Science, Engineering and Information Technology, 9(4): 1251-1259. https://doi.org/10.18517/ijaseit.9.4.5858.

[3] Chen J, Dan H, Ding Y, Gao Y, Guo M, Guo S, et al. (2021) New innovations in pavement materials and engineering: A review on pavement engineering. Journal of Traffic and Transportation Engineering, 8(6): 815–999. https://doi.org/10.1016/j.jtte.2021.10.001.

[4] Kołodziej K, Bichajło L, Siwowski T. (2021) Effects of aging on the physical and rheological properties of trinidad lake asphalt modified bitumen. Journal of Materials,14(10): 2532. https://doi.org/10.3390/ma14102532.

[5] Al-Azawee ET, Latief RH. (2020) The feasibility of using styrene-butadiene-styrene (SBS) as modifier in Iraqi bituminous binder. Journal of Engineering Science and Technology, 15(3): 1596–1607.

[6] Petrov SM, Kayukova GP, Goncharova IN, Safiulina AG, Lakhova AI. (2018) High-quality asphalt binders produced by deasphalting of natural bitumen. Section Oil and Gas Exploration, 14(1.4): 455-460. https://doi.org/10.5593/sgem2018/1.4/S06.060.

[7] Zhang XF, Zeng Y, Feng YN, Zhang CX, Zhang L. (2023) Carbon emissions analysis of producing modified asphalt with natural asphalt. Green Processing and Synthesis, 12(1): 20228146. https://doi.org/10.1515/gps-2022-8146.

[8] Khusnutdinov I, Goncharova I, Safiulina A. (2021) Extractive deasphalting as a method of obtaining asphalt binders and low-viscosity deasphalted hydrocarbon feedstock from natural bitumen. Egyptian Journal of Petroleum, 30(2): 69-73. https://doi.org/10.1016/j.ejpe.2021.03.002.

[9] Anupam K, Akinmade D, Kasbergen C, Erkens S, Adebiyi F. (2023) A state-of-the-art review of Natural bitumen in pavement: Underlining challenges and the way forward. Journal of Cleaner Production, 382(8): 134957. https://doi.org/10.1016/j.jclepro.2022.134957.

[10] Kök BV, Yílmaz M, Turgut P, Kulog N. (2012) Evaluation of the mechanical properties of natural asphalt-modified hot mixture. International Journal of Materials Research, 103(4): 506-512. https://doi.org/10.3139/146.110654.

[11] Nalbandian KM, Carpio M, González A. (2021) Analysis of the scientific evolution of self-healing asphalt pavements. Journal of Cleaner Production, 293: 126107. https://doi.org/10.1016/j.jclepro.2021.126107.

[12] Mahmood NS. (2011) Sand dunes modification by natural asphalt with cement and lime. Muthanna Journal of Engineering and Technology, 1(1): 66-75. https://muthjet.mu.edu.iq/wp-content/uploads/2022/08/Sand-dunes-modification-by-natural-asphalt-with-cement-and-lime.pdf.

[13] Bilski M. (2018) Natural asphalts – properties and use. Archives of Institute of Civil Engineering, 27(27): 25-36. DOI:10.21008/j.1897-4007.2018.27.02.

[14] Al Dulaymie AS, Hussien B, Gharbi MA, Mekhlif HN. (2013) Balneological study based on the hydrogeochemical aspects of the sulfate springs water (Hit-Kubaiysa region), Iraq. Arabian Journal of Geosciences, 6(3): 801-816. https://doi.org/10.1007/s12517-011-0385-5.

[15] Sobhi S, Yousefi A, Behnood A. (2020) The effects of Gilsonite and Sasobit on the mechanical properties and durability of asphalt mixtures. Construction and Building Materials, 30(238): 117676. https://doi.org/10.1016/j.conbuildmat.2019.117676.

[16] Mohammed FH, Latief RH, Albayati AH. (2024) Assessment of traditional asphalt mixture performance using natural asphalt from sulfur springs. Journal of Engineering, 30(1): 54-73. https://doi.org/10.31026/j.eng.2024.01.04.

[17] Mahmood BA. (2021) Influence of waste engine oil addition on the properties of natural asphalt. In Proceedings of the Second International Virtual Conference on Pure Science: 21-22 April 2020; Diwaniyah. pp 1-11. https://dx.doi.org/10.1088/1742-6596/1999/1/011001.

[18] Koshkarov V, Shunyaev I, Koshkarov E. (2023) Characteristics evaluation of modern bituminous binders in group hydrocarbon composition and their application in construction. Materials Sciences and Applications, 14(8): 407-415. https://doi.org/10.4236/msa.2023.148026.

[19] Shlimon AG, Mansurbeg H , Othman RS, Gittel A, Aitken CM , Head IM, et al. (2020) Microbial community composition in crude oils and asphalts from the kurdistan region of Iraq. Geomicrobiol Journal, 37(7): 635-652. https://doi.org/10.1080/01490451.2020.1753131.

[20] Farhan MM, Rabeea MA, Muslim RF, Zidan TA. (2021) Chemical composition (saturate fraction) of western Iraq natural bitumen. Journal of Materials, 42(5): 2527–2533. https://doi.org/10.1016/j.matpr.2020.12.574.

[21] Muttar AA, Zedan TA, Mahmood BA. (2009) Analytical comparison study for asphalt and water of Heet sulphurous springs. Journal of University of Anbar for Pure Science, 3(1): 45-56. https://doi.org/10.37652/juaps.2009.15453.

[22] Abdul-Jaleel T, Najres M. (2012) Oxidation influence on the chemical content and some rheological properties for natural asphalt to Abu Al Jeer-Al Anbar. Iraqi Journal of Desert Studies, 4(1): 1-15. DOI:10.36531/ijds.2012.68137.

[23] Abdul-Jaleel T, Salem JW, Najres AM. (2016) A new separation, fractionation and improving of Abu- Aljeer asphalt. Anbar Journal of Engineering Science, 7(1): 31-41. https://doi.org/10.37649/aengs.2016.124363.

[24] Mahmood BA, Al-Ani Y, Mohammed KA, Eyada SO. (2018) The feasibility of using plastic wastes to improve the properties of natural asphalt. Journal of Engineering and Applied Science, 13(21): 8929–8934. https://doi.org/10.3923/jeasci.2018.8929.8934.

[25] Ahmed F, Latief R, Albayati A. (2024) Effect of natural bitumen on the performance of hot asphalt mixture. Gazi University Journal of Science, 37(3): 1154–1170. https://doi.org/10.35378/gujs.1282239.

[26] State Corporation for Roads and Bridges. General Specification for Roads and Bridges (SORB/R9)-Hot Mix Asphaltic Concrete Pavement. Baghdad, Iraq: Department of Planning and Studies, Republic of Iraq, Ministry of Housing and Construction; 2003.

[27] Altameemi AA, Al-Jumaili MA, Mohammed AJ, Abdul karim A. (2016) Studying the possibility of utilizing the vast deposits of natural asphalt in Iraq in specific uses using as little as possible manufacturing treatment. The Iraqi Journal for Mechanical and Material Engineering, 1(1): 462-488.

[28] Tosun HB, Alver A, Baştürk E. (2022) Removal of exhaust gas with advanced solar photocatalytic asphalt applications. KSCE Journal of Civil Engineering, 26(1): 13-24. https://doi.org/10.1007/s12205-021-0654-0.

[29] Mithil M, Raju A, Ahmed W, Soon J. (2018) SEM and ESEM techniques used for analysis of asphalt binder and mixture. Journal of Construction and Building Materials, 186(1): 313-329. https://doi.org/10.1016/j.conbuildmat.2018.07.126.

[30] Liang T, Wang J, Kou X, Peng P. (2024) Influence mechanism of original components on mechanical properties and transformation behaviors of natural bitumen. Fuel, 371 (Part A): 131924. https://doi.org/10.1016/j.fuel.2024.131924.

[31] Zhang M, Hao P, Dong S, Li Y, Yuan G. (2020) Asphalt binder micro-characterization and testing approaches: A review. Measurement, 151: 107255. https://doi.org/10.1016/j.measurement.2019.107255.

[32] Yamaguchi K, Sasaki I, Nishizaki I, Meiarashi S, Moriyoshi A. (2005) Reinforcing effects of carbon black on asphalt binder for pavement. Journal of the Japan Petroleum Institute, 48(6): 373−379. https://doi.org/10.1627/jpi.48.373.

[33] Yao H, Liu J, Xu M, Bick A, Xu Q, Zhang J. (2021) Generation and properties of the new asphalt binder model using molecular dynamics (MD). Scientific Reports, 11: 9890. https://doi.org/10.1038/s41598-021-89339-5

[34] Khedaywi T, Haddad M, Mujalli R, Shareef S. (2023) Effect of sulfur on the asphalt cement and asphalt concrete mixture. Innovative Infrastructure Solutions, 8(11): 286. https://doi.org/10.1007/s41062-023-01264-w.

[35] Awadh SM, Al-Mimar HS. (2023) The effect of nickel, vanadium, asphaltene, NSO and sulfur on crude oil quality. Iraqi Geological Journal, 56(2B): 137-144. https://doi.org/10.46717/igj.56.2B.10ms-2023-8-19.

[36] Yuan Y, Chen H, Wang Y, Xu S, Xue B, Zou G, Wang B. (2023) How calcium and magnesium ions affect the stability of asphalt with anionic emulsifiers: A molecular dynamics study. Construction and Building Materials, 12(10): 132770. https://doi.org/10.1016/j.conbuildmat.2023.132770.

[37] Sakib N, Bhasin A, Islam MK, Khan K, Khan MI. (2021) A review of the evolution of technologies to use sulphur as a pavement construction material. International Journal of Pavement Engineering, 22(3): 392–403. https://doi.org/10.1080/10298436.2019.1612064.

[38] Sanja D, Miroslav S, Martina Z, Tatjana R. (2021) Laboratory evaluation of the properties of asphalt mixture with wood ash filler. Journal of Materials, 14(3): 575. https://doi.org/10.3390/ma14030575.

[39] Abd NI, Latief RH. (2024) Assessment of rutting resistance for fiber-modified asphalt mixtures. Journal of Engineering, 30(5): 98–113. https://doi.org/10.31026/j.eng.2024.05.07.

[40] Omar HA,Yusoff NI, Mubaraki M, Ceylan H. (2020) Effects of moisture damage on asphalt mixtures. Journal of Traffic and Transportation Engineering, 7(5): 600–628. https://doi.org/10.1016/j.jtte.2020.07.001.

[41] Al-Saadi AA, Ismael MQ. (2023) Improvement of moisture susceptibility for asphalt mixture with ceramic fiber. Journal of Engineering, 29(4): 78–91. https://doi.org/10.31026/j.eng.2023.04.05.

[42] Yildiz I. (2018) Chapter 1.12 Fossil Fuels. In Comprehensive Energy Systems, 521-567. https://doi.org/10.1016/B978-0-12-809597-3.00111-5.

[43] Kaylyn MC, Khalafalla M, Benavides JR. (2021) Protocol to assess the impact of crude oil price fluctuations on future asphalt prices. Journal of the Transportation Research Board, 2675(6): 294-305. https://doi.org/10.1177/0361198121992072.

[44] U.S. Bureau of Labor Statistics, U.S. Department of Labor. Website Available: https://www.bls.gov/data/.