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Effect of Natural Bitumen on the Performance of Hot Asphalt Mixture

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Highlights

- This paper focuses on using natural asphalt from sulfur springs in local asphalt mixtures.
- Heating treatment is applied to modify the properties of natural asphalt.
- Using natural asphalt instead of conventional asphalt can produce a good-performing asphalt mixture.

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Abstract

For over a century, the global consumption of asphalt binder in asphalt mixture production has been substantial. In the Heet region (west of Iraq), two distinct forms of natural asphalt (NA) deposits exist: rock asphalt and sulfur spring asphalt. This study focused on using NA sourced from sulfur springs. The aim was to investigate the potential of incorporating NA into local asphalt mixtures. To achieve this, NA was heated to 163°C for varying durations. After heat treatment, laboratory tests were conducted on NA. The findings suggest that by heating NA for 20 hours, it conforms to Iraqi specifications in terms of physical properties. Furthermore, compared to conventional petroleum asphalt, treated NA showed greater resistance to temperature fluctuations, making it ideal for hotter climates. The study also found that NA enhances mechanical properties significantly. Specifically, the NA mixture recorded the highest indirect tensile strength, with a tensile strength ratio of 81.2%, a slight increase of 0.37% over traditional mixes. In summary, NA mixtures exhibit commendable performance. Given its abundance and affordability in Iraq, after considering heat treatment costs and environmental impact, NA holds promise for the future of asphalt concrete production for the construction of flexible pavement.

1. INTRODUCTION

Enhancing the qualities of the asphalt mixture—by, for example, improving resistance to rutting, fatigue cracking, and moisture damage—will extend the service life of flexible pavement and thereby improve sustainability by reducing costs and pollution [1-5]. Hot-mix asphalt (HMA) is primarily composed of aggregate and asphalt binder [6]. One of the first building materials used by humans was asphalt. Since the early 1900s, the amount of asphalt binder consumed globally has rapidly increased thanks to the usage of large quantities of binder in asphalt mixtures [7]. Poor asphalt quality shortens the pavement lifespans and contributes to other problems such as a need for frequent repairs, early failures, higher maintenance costs, risky driving conditions, and decreased safety [8–10]. Therefore, developing new asphalt mixtures that use materials available in nature instead of conventional materials is necessary [11,12].

Heavy oil and natural asphalts (NAs) have emerged as significant sources of raw materials to meet the rising demand for fuels and petrochemical products because light and medium crudes are becoming depleted [13]. NAs have received a significant amount of attention in the last decade, and the recent studies have focused on developing NAs via physical and chemical processes for industrial purposes such as paving and roofing, moisture barricading and electrical insulation. These processes include anaerobic and aerobic oxidation, chlorination, and using links to increase adhesive properties, such as polymers, phosphoric acid, and metal salts [14].

In general, NAs are composed of carbon, hydrogen, nitrogen, oxygen, sulfur, and a trace amount of iron, nickel, and vanadium [15]. The most typical types of NA are Gilsonite, Trinidad Lake Asphalt (TLA), and

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rock asphalt (RA). TLA is too hard to be employed as a binder by itself in asphalt mixes, despite having a chemical composition similar to petroleum asphalt due to its high mineral and asphaltene content [16]. The characteristics of modified asphalt mixtures made with styrene-butadiene-styrene (SBS) and TLA were studied by Xu et al. According to the findings, SBS and TLA not only strengthened the asphalt's resistance to fracture but also extended its service life and aging resistance [17]. In the study of Yilmaz and Celoglu, the mixtures made with 40% pure bitumen and 60% TLA had the greatest values for Marshall stability, indirect tensile strength (ITS), hardness, and longest fatigue life [18]. Gilsonite is a natural asphalt with a ruptured surface that is serrated and quite black and lustrous in color. Its frail structure makes it easily crushable. It has a specific gravity that ranges from 1.03 to 1.10. It's softened between 120 and 175 °C and has a penetration value of zero [19-21]. Finally, RA is a natural building material that does not require chemical processing and is extremely compatible with bitumen. It enhances water resistance, durability, and high-temperature strength when employed in modified binders [22, 23].

NA is found in the geological regions in Iraq that are affected by the presence of a fault or crack in the earth's crust, particularly in the central region toward the west on the banks of the Euphrates River in Anbar Governorate and the northern region. This fault or crack causes groundwater to rise to the surface and sometimes carries tar material or deposits, in addition to salt water and sulfur. Some cities' names have been linked to the existence of natural tar. Historical sources claim that the city name (Heet) is derived from the Assyrian-Babylonian word (Hito), which signifies tar.

Few studies have focused on evaluating the performance of local HMA with NA supplied from sulfur springs in Iraq. Altameemi et al. [24] investigated the properties of NA deposits from five sites in the Heet region (Al-Awasl, Al-Tayf, Abu-Jeer, Al-Merj, and Al-Mamorh) and samples from the northern regions (Al-Qaiyarah and Hamam Al-Aleel). Experimental tests were performed to estimate the proportions of salty water combined with asphalt. Physical characteristics were also investigated to determine whether NA could be used for various construction projects after it received industrial treatments. Two treatment techniques for NA were used: heating to high temperatures and mixing with refined asphalt. It was found that combining NA with refined asphalt or heating it to 163 ^{0}C for varying periods of time, depending on the type of asphalt, could produce asphalt that met Iraqi requirements for asphalt used in road pavements.

Test results from the Abdul-Jaleel and Najres [25] study indicated that the thermal oxidation of NA from Abu Aljeer at high temperatures under atmospheric conditions improved its chemical and physical properties, increased the ratio of asphaltene and aromatic parts, and decreased the ratio of saturated and resinous parts. The study also revealed a relationship among the chemical content of asphalt, oxidation processes, and the stability of the colloidal state.

Abdul-Jaleel et al. [26] investigated the chemical properties of springs NA in the Al-Anbar province (in western Iraq). The main goal of this study was to apply two methods (column chromatography and extraction chromatography) to separate asphalt components. The separation results were 45.23% paraffinic, 28.39% aromatic, 21.66% resin, and 10.20% asphltene. The minor goal was to develop the rheological and thermal properties of NA by incorporating limestone at 5, 15, 25, and 35% concentrations. The results were compared to artificially created asphalt, such as asphalt cement from the Al-Dora refinery located in Baghdad, the capital of Iraq.

Widespread NA deposits in Iraq are being investigated to capitalize on them. NAs represent untapped national wealth at relatively low prices and are considered excellent alternative energy resources. NAs are extracted through simple mining and prospecting methods. Additionally, they effectively reduce emissions and energy consumption. These benefits are the motives behind the authors' interest in the subject. The study was conducted to identify the physical properties of NA as a new construction material and to demonstrate that industrial treatments can improve NA's characteristics to produce economical, ecological, and sustainable HMA.

2. MATERIALS

This work was accomplished using materials that were locally available, which is advantageous economically. The following materials were used:

2.1. Conventional Asphalt

The most popular asphalt for paving in the middle and south of Iraq is AC (40–50). It was supplied from the refinery in Al-Dora.

2.2. Natural Asphalt

Natural asphalt (NA), as illustrated in Figure 1, is brought from Abu-Jeer in Heet city in Al-Anbar province, located in the west of Iraq. The physical properties of NA differ significantly from the properties of conventional asphalt produced in highly controlled refineries. The initial state of NA is not usable because it contains a lot of impurities [24], despite the finding of a previous study by Muttar et al. [27], which stated that the specifications of spring NA are similar to those of industrial bitumen. Industrial treatments are required when using NA in pavement work.

Figure 1. *Abu-Jeer natural asphalt*

2.3. Fine and Coarse Aggregates

Al-Nibaie quarry provided the aggregates for this study. Following the Iraqi specification requirements, the coarse aggregate sizes have ranged from sieve No. ¾" to sieve No. 4 and from sieve No. 4, to sieve No. 200 are the fine aggregates [28]. In order to produce the chosen gradation, aggregates were sieved and then recombined in the laboratory to provide an identical and regulated gradation. Table 1 and Figure 2 show the selected gradation, while the physical characteristics of coarse and fine aggregates are listed in Table 2.

Table 1. *Aggregate grading for wearing course type IIIA*

Sieve Size	Sieve Opening, (mm)	% Passing by Weight	
		Selected Gradation	Specification Range [28]
3/4"	19	100	100
1/2"	12.5	95	90-100
3/8"	9.5	83	76-90
No. 4	4.75	59	44-74
No. 8	2.36	43	28-58
No. 50	0.3	12	5-21
No. 200	0.075	7	4-10

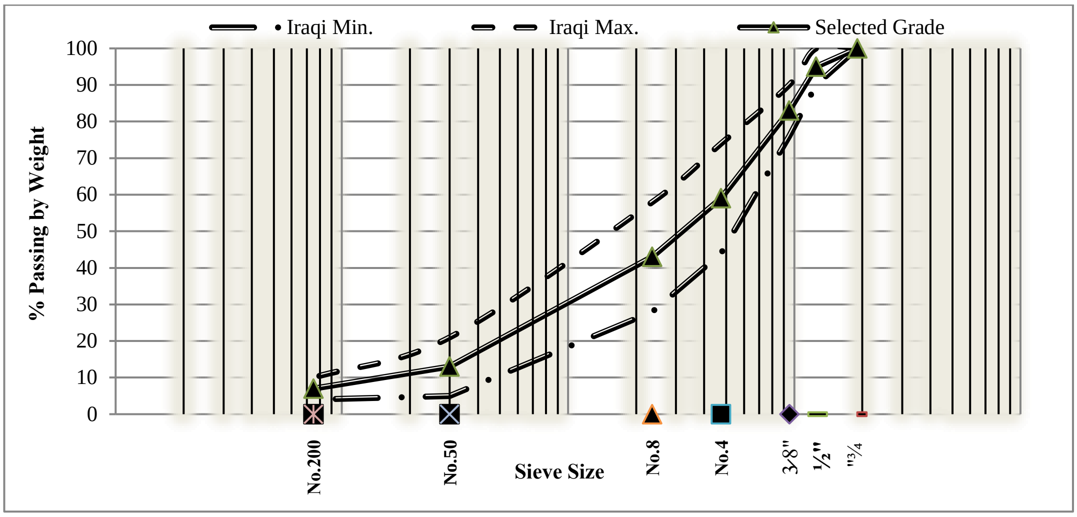

Figure 2. *Aggregate gradation curve*

Table 2. *Physical properties of coarse and fine aggregates*

Properties	Test Methods	Test Results	Requirements [28]
Coarse Aggregate			
Wearing (Los Angeles Abrasion), %	ASTM C-131	12.7	30 Maximum
Bulk Specific Gravity	ASTM C-127	2.61	---
Water Absorption, %	ASTM C-127	0.22	---
Fractured Pieces,%	ASTM D-5821	97	90 Minimum
Flat and Elongated Particles,%	ASTM D-4791	4	10 Maximum
Fine Aggregate			
Water Absorption, %	ASTM C-128	1	---
Bulk Specific Gravity	ASTM C-128	2.655	---

2.4. Mineral Filler

The southeast-located Iraqi city of Karbala is where the limestone filler was shipped from. Table 3 provides a list of the physical properties of limestone filler.

Table 3. *Physical properties of limestone filler*

Property	Value
Specific Gravity	2.7
Passing No.200, %	95

3. EXPERIMENTAL WORK

Laboratory tests were applied to assess NA performance and determine the suitability of using it in HMA instead of conventional asphalt type AC (40-50).

3.1. Treatment of Natural Asphalt (NA)

The physical and rheological properties of the binder have a significant impact on the performance of HMA. NA from Heet is unsuitable for industrial applications because it has poor rheological properties. Heating treatment is applied to NA to modify its properties and achieve the expected performance of HMA. The first stage of treatment for NA was done by stirring it in 500 ml steel containers during the two hours of oxidation in a heated oven at a temperature of 110 °C to ensure water expulsion. After removing the water, it is important to apply some tests to ascertain the physical properties and decide if using NA in local HMA is applicable.

NA was heated and subjected to a temperature of 163 ^{0}C during the second stage of treatment. To create asphalt with properties that meet the requirements of the Iraqi specification [28] for the types of asphalt used in the production of HMA for the construction of the base, binder, and surface layers in local paving roads. The NA was heated at different times (5, 10, 15, 20 and 25) hours.

3.2. Asphalt Tests

Asphalt tests such as penetration, softening point, flash point, ductility, and specific gravity were conducted for conventional asphalt and NA heated to 0, 5, 10, 15, and 20 hrs. A rotational viscosity test was also conducted for conventional asphalt, NA, and for NA that was heated for 20 hours. All tests are listed in Table 4.

Table 4. *Asphalt tests*

Test	Condition	Unit	Test Methods
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	4 ºC	°C	ASTM D-36
Flash Point	-	°C	ASTM D-92
Specific Gravity	25 ºC	-	ASTM D-70
Rotational viscosity	135 ºC	Pa.s	ASTM D-2170

3.3. Elemental Analysis Test

The chemical composition of molecular species significantly impacts the configuration of the internal structure of bitumen. Bitumen is a complex chemical combination comprising primarily hydrocarbon-based compounds, with small amounts of structurally similar heterocyclic species and functional groups with sulfur, nitrogen, and oxygen atoms. Metals like vanadium, nickel, iron, aluminum, and calcium, which appear as inorganic slats and oxides or porphyrine statures, are also found in trace amounts in bitumen.

The elements of AC (40–50) and NA were determined using the Vario EL III elemental analyzer from Elementar Analysensysteme GmbH (Hanau, Germany), as seen in Figure 3. The micro-combustion method is the test mechanism, and a particular test method called CHNS mode was used to test the four main elements, such as carbon, nitrogen, hydrogen, and sulfur.

Figure 3. *Vario-EL, III element analyzer*

3.4. Marshall Test

ASTM D6927-15 Marshall procedure is used to prepare, compact, and test specimens. The aggregate gradation for the wearing course is blended for each specimen. The asphalt cement and aggregates are then heated for two hours before being mixed at temperatures of up to 135 °C and 150 °C, respectively. Excessive heating and preheating of asphalt were avoided because they alter the properties of the asphalt.

When various amounts of asphalt (4, 4.5, 5, 5.5, and 6) percent by weight of aggregate were applied to the heated aggregate, the mixture was stirred for at least three minutes until it reached a homogenous state, covering all of the aggregates with asphalt cement. In the Marshall test, which uses conventional cylindrical molds with dimensions of 64 mm in height and 102 mm in diameter that have been heated to 130 °C, the mold is filled with the hot mixture, and 75 blows were applied to the specimen on each side by a Marshall hammer to compress it. The mix characteristics, such as flow value, Marshall stability value, proportion of air voids, voids filled with asphalt (VFA), voids in mineral aggregates (VMA), and optimal binder content (OBC), are determined for both AC (40-50) and NA after treatment for 20 hours mixes.

Thirty Marshall samples were used to obtain the OBC and assess the Marshall properties: fifteen samples for the AC (40-50) mixture and the same number of samples for the mixture prepared with NA after treatment for 20 hours. Figure 4 displays samples of AC (40–50) and NA mixtures. The Marshall apparatus, as shown in Figure 5, was used to test the stability and flow of the specimens.

Figure 4. *Marshall specimens*

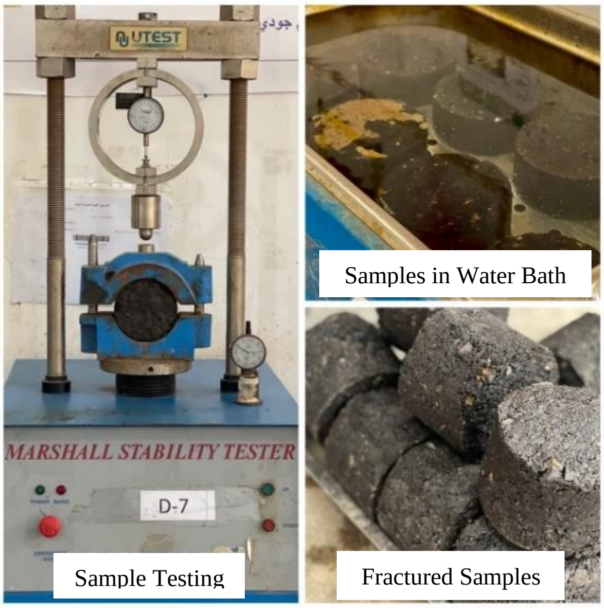

Figure 5. *Marshall test*

3.5. Moisture Damage Test

The tensile strength ratio was measured following ASTM D 4867M-96 to determine the sensitivity of compacted mixes to water damage. This test is recommended to show whether the presence of water is causing a decrease in asphalt and aggregate particle adhesion. Briefly stated, for AC (40-50) and NA after treatment for 20 hours, a set of Marshall Samples was prepared with OBC. The set was divided into two groups: the unconditioned group spent 20 minutes submerged in a water bath at 25 °C, while the second group (conditioned) underwent one freezing and thawing cycle before submerging for an hour in the same water bath at the same temperature. As shown in Figure 6, the specimens were tested at a rate of 50.8 mm per minute until the maximum load was reached and the specimen was totally fractured. The load was then recorded.

Figure 6. *Moisture damage test*

A tensile strength ratio (TSR) of at least 80% is recommended. The following equation can be used to determine the TSR value:

$$TSR = \frac{Ts}{Tss} * 100 \quad (1)$$

$$Ts \text{ or } Tss = \frac{2000 * P}{\pi\ H\ D} \quad (2)$$

where:
TSR = Tensile Strength Ratio, %
Ts = Average Tensile Strengths of the Conditioned Groups, Kpa
Tss = Average Tensile Strengths of the Unconditioned Groups, Kpa
P = Failure Load, N
H = Sample Height, mm
D = Sample Diameter, mm.

4. RESULTS AND DISCUSSION

4.1. Asphalt Properties

Tables 5 and 6 show the physical properties and chemical composition of AC (40-50) and NA after water removal. The conventional asphalt (40–50) employed in this research met Iraqi requirements, as shown in the relevant tables. However, significant deviations from the requirements were observed in NA because NA derived from sulfur springs has a soft texture, which causes high penetration, low softening point, and low viscosity. Also, the flash point for NA is below the lower limit of Iraqi requirements, which is critical for safety. As expected, NA cannot be used instead of AC (40-50) in paving construction. Notably, the hard texture of the bitumen helps the pavement bear traffic loads, while the soft texture helps increase its workability during mixing. The effect on ductility is negligible.

To make wide use of the NA for great performance, it is necessary to understand its chemical composition and determine if it is consistent with that of conventional asphalt supplied from the crude petroleum refinery. Asphalt's chemical composition can vary widely depending on the asphalt source. Nevertheless, the chemical properties of conventional asphalt and original NA are significantly different. This finding is not compatible with previous research findings [29].

***Table 5**. Physical properties of conventional asphalt (40-50) and original NA*

Test	Test Results		Requirements [28]
	AC (40-50)	NA	
Penetration, 1/10 mm	47	146	40 – 50
Ductility, cm	>150	103	≥ 100
Softening Point, ^{0}C	52	36	-
Flash Point, ^{0}C	245	180	232 min.
Specific Gravity	1.02	1.00	-
Rotational viscosity, Pa.s	0.488	0.233	-

***Table 6.** Chemical composition of conventional asphalt (40-50) and original NA*

Asphalt Type	Carbon (%)	Hydrogen (%)	Nitrogen (%)	Sulfur (%)
Conventional Asphalt	70.82	7.652	0.519	6.048
Natural Asphalt (NA)	68.05	6.020	0.938	8.460

To adjust the properties of NA to meet the local requirements of asphalt, heating treatments are applied for periods of 5, 10, 15, 20, and 25 hours. The physical properties of NA were tested after heating treatment and are listed in Table 7. The chemical composition of NA heated for 20 hours is also listed in Table 8. It can be concluded that when NA is heated at high temperatures for more than 20 hours, oxidation begins on its surface. Continuous mixing and heating causes all of the asphalt molecules to oxidize, which changes the material's texture and gives the NA a solid, glossy and dark-black color.

Both NA and conventional asphalt ductility were greater than 100 cm. However, the ductility of all NA samples increased rapidly with the increased heating treatment time. The increase in ductility was attributed to a high sulfur concentration that increased from 8.460% to 8.971% with the increase in heating treatment time to 20 hours. Furthermore, the sulfur concentration of conventional asphalt (AC 40–50) is 6.048%, which is less than that of NA. This result is consistent with previous investigation outcomes [30]. Sulfur concentrations for conventional asphalt and NA are illustrated in Tables 6 and 8. Accordingly, the use of NA in the Abu al-Jeer region in the construction of paving layers will improve the tensile strength of the resulting paving. Also, the specific gravity of NA increased by 3.3%, 3.4%, 3.48%, 3.6%, and more than 5.2% by heating for 5, 10, 15, 20 and 25 hours, respectively.

One of the physical properties of NA related to safety and security when dealing with bitumen grades high temperature is the characteristic of the flash point, where the flash point represents the highest temperature at which the bitumen can be heated without volatilizing vapors and gases that pose a threat to the safety of those dealing with the bitumen. Table 7 illustrates that the flash point values by heating for 5, 10, 15, 20 and 25 hours increased by 11.1%, 27.8%, 30.6, 38.9%, and 47.78%, respectively. Accordingly, the heat-treated NA is safe in terms of health and can be dealt with.

Petroleum asphalt exposed outdoors for a long period of time experiences rapid oxidation within a few years due to the impact of Ultraviolet Rays (UV) light. On the other hand, NA has good resistance to weathering because, when the asphalt mixture extraction experiment was carried out, the black trichloroethylene on the aggregate surface did not drop after several washings [31, 32]. This is because NA has a strong affinity with stones and contains a high content of nitrogen. In this study, the nitrogen content of NA after treatment for 20 hours was 1.48%, which is more than that of conventional asphalt by 1.85 times.

The physical properties result showed that NA treated by heating to 163 °C for 25 hours would not be used in HMA pavements because NA penetration was below the lower limit of the Iraqi specification requirements [28]. It is worth noting that NA becomes stiffer during the heating process by more than 20 hours due to the chemical conversion of light components to heavy components. In conclusion, NA after treatment for 20 hours fulfills the requirements of Iraqi specifications for asphalt that used in the construction of flexible paving.

The result showed that an increase in oxidation time improved the physical characteristics of the NA by decreasing the degree of penetration (increasing stiffness) and increasing viscosity. The viscosity of NA at 135 ^{0}C increased by 113 percent with a 20-hour heating treatment. At the same time, the increase in NA viscosity was only 1.63 percent more than the viscosity of conventional asphalt and, this was consistent with the softening point testing results.

Table 7. *Physical properties of NA after heating treatment*

Test	Heating Period in hr. at 163 ^{0}C						Specification Limits [28]
	0	5	10	15	20	25	
	Results						
Penetration	146	119	67	60	46	12	40 – 50
Ductility	103	109	115	137	>150	>150	≥ 100
Softening Point	36	39	47	49	53	65	-
Flash Point	180	200	230	235	250	266	232 min.
Specific Gravity	1.000	1.033	1.034	1.0348	1.036	1.052	-
Rotational viscosity	0.233	-	-	-	0.496	-	-

Table 8*. Chemical composition of 20 hrs. heated NA*

Asphalt Type	Carbon (%)	Hydrogen (%)	Nitrogen (%)	Sulfur (%)
20 hrs. heated NA	69.12	5.657	1.480	8.971

4.2. Temperature Susceptibility

The penetration index (PI) refers to the binder's temperature sensitivity, which is a quantitative measure (dimensionless value). The rate at which the consistency of a binder varies is known as temperature susceptibility. Equations (3) and (4) were used to compute PI in order to assess the asphalt binder's possible temperature sensitivity:

$$PI = \frac{20-500A}{1+50A} \quad (3)$$

$$A = \frac{\text{Log pen.@T} - \text{Log } 800}{T - T_{R\&B}} \quad (4)$$

where:
T = Testing Temperature
$T_{R\&B}$ = Ring and Ball Softening Point Temperature

Based on penetration at 25 ^{0}C and the softening point temperature, the PI value was calculated. The PI value was calculated for NA and conventional asphalt (40-50). Figure 7 displays the PI for conventional asphalt AC (40-50) and NA after 0, 5, 10, 15, and 20 hours of heating treatment, respectively. It is evident from the outcomes in Figure 6 that the heat sensitivity of NA decreases because the softening point is raised, and the penetration value is reduced with increasing heating hours. The susceptibility of the binder to temperature increases with decreasing penetration index. The PI values for conventional asphalt and 20 hr. heated NA are -0.853 and -0.668, respectively. This significant fact indicates that NA is less temperature sensitive and beneficial in terms of pavement deformation resistance. Thus, HMA with NA is better suited to hot climates in Iraq.

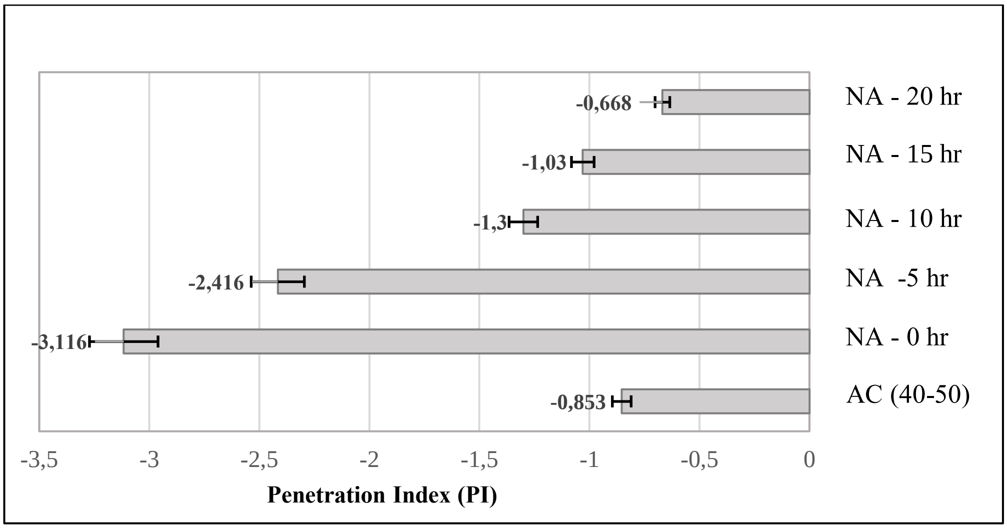

Figure 7. *Penetration index*

4.3. Marshall Properties

The Optimum Binder Content (OBC) is 4.96% and 4.93% (by weight of the aggregate) for conventional asphalt AC (40-50) and NA after treatment for 20 hours, respectively. The Marshall characteristics of both mixes (conventional mix and NA mix) are listed in Table 9. The performance indicators of HMA in the Marshall test are stability and flow. The results show that the NA mix has better flow and stability than the conventional mix. When compared to the AC (40–50) mix, the stability of the NA mix increased by about 17.6%. The reason may be that NA has a higher sulfur content than AC (40–50), and at temperatures over the melting point of sulfur (119 °C), sulfur chains are chemically bound to the organic molecules of the binder to produce polysulfides (R-Sn-R). In other words, sulfur chains serve as cross-linking agents between nearby organic molecules; this can result in polysulfides, which can stiffen the asphalt matrix [33].

According to the results, the Marshall flow for the NA mixture increased by 14.8% compared with the AC (40–50) mixture. The high flow indicates that the asphalt mixture has little resistance to the effects of loading from traffic.

The Marshall quotient (MQ) measures the rigidity of the mixes. A mix with a high MQ value will be stiffer, better able to distribute the applied load, and less susceptible to creep deformation [34, 35]. The MQ of the NA mix is greater than that of the conventional mix, indicating that the NA mix is more resistant to permanent deformation.

The air void (Vv%) limitations are (3-5%) as specified by Iraqi specifications [28]. Low Vv in asphalt mix (less than 3%) causes bleeding, and high Vv (more than 5%) makes the mix less resistant to the effects of moisture and fatigue. The Vv% for NA and AC (40–50) are within the limitations. However, the Vv% of the NA mix increased by 2.44% when compared to the AC (40–50) mix, as illustrated in Table 9.

Voids that are filled with asphalt in the spaces between the aggregate in the compacted mixture are known as void-filled with binder (VFB). The VFA aims to provide high durability for the mixture made from the thin asphalt films on the aggregate. The results demonstrate that the VFB% for the NA mix decreased by 2.7 % when compared to the AC (40–50) mix. The voids in the mineral aggregate (VMA) are important for the mixture's performance and must be high enough to confirm that there is enough bitumen in the mixture to meet the needs of durability. However, a mixture is more susceptible to the stability issue with increasing VMA. The results in Table 9 showed that the VMA of the NA mix was comparable to that of the AC (40–50) mix.

***Table 9**. Marshall characteristics*

Marshall Property	Conventional Mix	NA Mix	Specification Requirements [28]
Optimal Binder Content, %	4.96	4.93	4-6
Stability, kN	10.53	12.38	8 minimum
Flow, mm	3.18	3.65	2-4
MQ, kN/mm	3.31	3.39	-
Bulk Density, gm/cm3	2.34	2.34	-
Vv, %	4.10	4.20	3-5
VMA, %	15.70	15.70	14 minimum
VFB, %	74.00	72.00	-

4.4. Moisture Damage Assessment

Figure 8 depicts the unconditioned and conditioned mixes for indirect tensile strength (ITS). The highest ITS values were found in the NA mixture, with the NA mixture being higher than the conventional mixture by 9.1% and 9.5% for unconditioned and conditioned states, respectively. Higher ITS values indicated that the NA mixture was more resistant to tensile stresses that cause crack formation and lead to a longer service life for pavement. This result is in line with the previous investigations [36, 37].

In fact, TSR is important performance indicator of HMA. The TSR value of 0.80 is considered the minimum threshold for HMA by the Superpave design technique. Figure 9 displays the TSR values for mixtures. The NA mixture satisfies the Superpave requirements for the 80% lower TSR value limit. Moreover, it is a little bit higher (by 0.37%) than the TSR of the conventional mix.

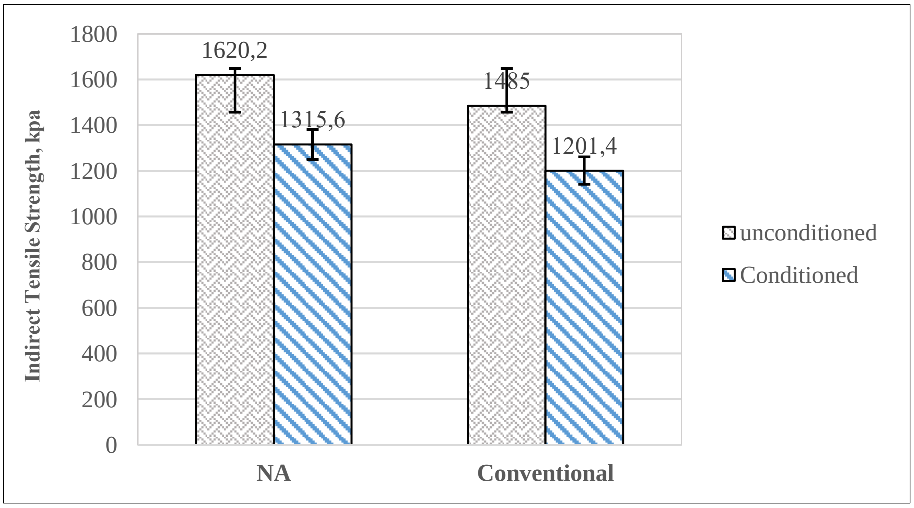

***Figure 8**. ITS for unconditioned and conditioned mixtures*

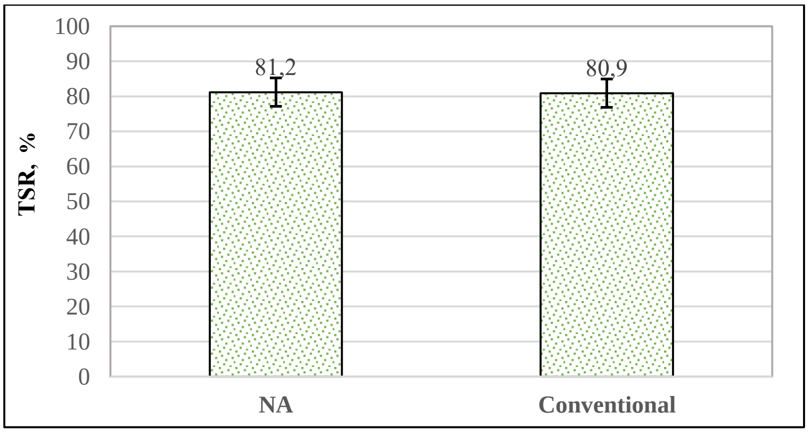

***Figure 9.** TSR values for conventional and NA mixtures*

4.5. Cost and Environmental Concerns of NA Application in Paving Work

Oil is still one of the world's primary natural resources used as an energy source; it accounts for more than 30% of total energy consumption. Venezuela (17.5%), Saudi Arabia (17.2%), Canada (9.8%), Iran (9.0%), Iraq (8.4%), and Russia (6.2%) are among the countries with the largest proven oil reserves [38]. Generally, around 10% of crude petroleum is converted into asphalt material and other products during the distillation process in petroleum refineries. Then, asphalt product manufacturers buy the asphalt from these refineries to produce retail products such as asphalt emulsions, asphalt paving mixtures, coatings, tar roofing, and other products. Refinery production is not the only factor affecting the cost of asphalt materials for paving work; it is considered a minor factor. In fact, the decisions made by the 12-nation Organization of the Petroleum Exporting Countries (OPEC) and the events around the world are the major factors that play an effective role in determining the cost of asphalt. According to the United States Bureau of Labor Statistics [39], the cost of asphalt over the last six decades has experienced a rise of more than 20-fold. This is because the asphalt material is a byproduct derived from petroleum refining; consequently, the cost of asphalt tracks crude oil prices. In the last decade, the petroleum sector has also changed its overall refining processes. Furthermore, many refineries are increasingly using a process called "coking to break down" or "crack" to produce high-grade petroleum distillates (like diesel fuel, gasoline, and other products) instead of asphalt because of the higher profits of these products. In turn, the recent advancements would further affect the quantity and quality of available asphalt, which directly represents a hard challenge to the road industry in the future to find new alternatives. Eventually, even if crude oil prices fall, the cost of asphalt will not recover and will continue to increase [40].

Peak oil, or oil depletion, is a serious concern related to asphalt costs that is important to discuss. Peak oil is the point in time when crude oil production around the world reaches its maximum rate, and production after this point will begin an irreversible decline because the petroleum reserves are finite and the extraction of crude oil will not be economically feasible. This is due to the low price of selling crude oil relative to the cost of the extraction process because the accessible reserves will deplete and there will be reductions in demand due to environmental regulations. Peak oil is predicted to begin in 2030 [41].

Because of concerns over the high cost of asphalt production, decreasing asphalt production in petroleum refineries, increasing crude oil prices, peak oil, climate change, and pollution, new alternatives have become more popular. In this context, NA may be economical and a good alternative to petroleum bitumen for road construction. This has led to studies on the feasibility of using NA in paving works around the world [42]. Especially in Iraq, NA is available in massive quantities at a low price.

Throughout the asphalt manufacturing process, the environment is carefully considered. Emissions from petroleum refineries and asphalt manufacturers should be strictly controlled [43]. Carbon emissions reduction during asphalt pavement production and transportation is receiving more attention around the world. To achieve this goal, using low-carbon technologies has become urgent, and searching for naturally present materials that are more environmentally friendly is a prerequisite to controlling and reducing greenhouse gas emissions as much as possible. Therefore, NA is a unique material, and using it may be suitable for reducing carbon emissions. In this study, the original NA after treatment for 20 hours improved the softening point and viscosity while decreasing the penetration value. This causes higher temperatures during treatment, mixing, and compaction, which raises energy use and related CO_2 emissions. This may be avoided by modifying conventional asphalt with the original NA. This will be the next research project planned by the authors. However, researchers in recent studies [44, 45] revealed that the production of modified petroleum asphalt with NA would reduce carbon emissions as well as improve the overall performance of the asphalt mixture. Also, modifying the original NA with additives offers more favorable economic and environmental outcomes [46–48].

For applying NA in paving work, the physical properties, chemical composition, heat treatment, and asphalt mixture performance were mainly performed and discussed in this study. It is thus necessary and worthy of future research to study the economic benefits and carbon emissions ratio for using Iraqi NA in HMA during production, transportation, and construction, then compare it with conventional asphalt.

5. CONCLUSIONS AND RECOMMENDATIONS

Throughout this research, an extensive set of tests was performed on both conventional and NA-based mixtures, with subsequent comparative analysis on their performances. The key findings include:

1. After the extraction of water, the inherent physical characteristics of NA are not in compliance with the Iraqi standards for asphalt cement. In its natural state, NA is not suitable for pavement applications. To address this, heat treatments over varying intervals (5, 10, 15, 20, 25 hrs.) are administered to NA, adjusting its properties to meet the desired performance levels of HMA.

2. Physical assessments of asphalt reflected that heat treated NA samples, generally exhibited lower penetration, augmented ductility, increased softening and flash temperatures, improved specific gravity, and heightened viscosity compared to their untreated counterparts. Remarkably, NA can met the Iraqi asphalt cement specifications when subjected to heating at 163°C for a duration of 20 hours. However, prolonged heating beyond this duration can render the NA excessively stiff, thus complicating the compaction processes in asphalt concrete placement during pavement construction.

3. The chemical composition of NA has an effect on its physical properties. After heat treatment for 20 hours, the ductility of NA increased because the sulfur concentration increased. Also, NA has good resistance to weathering because it has a strong affinity with stones and contains a high content of nitrogen, which is more than that of conventional asphalt by 1.85 times.

4. Penetration index assessments indicate that NA exhibits a reduced temperature sensitivity compared to the conventional asphalt AC (40–50). This characteristic enhances its capacity to resist pavement deformation.

5. The mixture incorporating NA demonstrated a 17.6% increase in Marshall stability as compared to conventional asphalt cement. This enhancement could attributed to NA's elevated sulfur content; during the preparation phase, when the asphalt blend is exposed to temperatures surpassing sulfur's melting point (119°C), the mixture gains additional rigidity. Complementing this, MQ showed a superior rigidity in the NA blend the matter which reflect a potential role for NA in improving the properties of HMA at hot climatic conditions.

6. The results of the Indirect Tensile Strength (ITS) and Tensile Strength Ratio (TSR) substantiated the suitability of NA as an effective binder material. Specifically, the NA mixture had the highest indirect tensile strength values and a tensile strength ratio of 81.2%, which was 0.37% higher than the TSR of the conventional mix.

Further research in the following areas could provide a deeper understanding and augment the insights of the current study:

- The rheological properties of NA can change due to the aging effect after applying the heat treatment at 163 ºC for 20 hours. Therefore, aging and rheological tests should be applied to NA for future studies before using it in HMA.

- Since the economic benefits and environmental impact were not considered in this study, there is a need for more studies to estimate the economic feasibility and investigate the environmental implications, particularly in terms of emissions resulted from using NA in flexible pavement construction.

- Additional studies are required to assess the characteristics of NA sourced from periodic asphalt extractions. Factors such as the age of the deposit, depth of extraction, and proximity to sulfur springs should be considered, as they might significantly influence the properties of NA.

CONFLICTS OF INTEREST

No conflict of interest was declared by the authors.

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