

Modification of sulfur spring bitumen with Gilsonite: A step towards exploiting natural bitumen in flexible pavement production

Roaa Hamed Latief*

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

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Abstract

In recent years, there has been increasing interest in employing Natural bitumen (NB) as a modifier or bitumen in road construction due to its exceptional properties. This study investigates the ability to produce new asphalt binders by mixing two types of NB. In this work, NB from deposits in the western region of Iraq is selected. It consisted of two types, including the sulfur spring bitumen (NB-SS) and Gilsonite bitumen (NB-G). NB-SS modified with NB-G in percentages of 40, 50, and 60% by the weight of NB-SS. Conventional asphalt tests were conducted to examine the physical properties of bitumen. Furthermore, chemical tests, including Scanning Electron Microscopy (SEM), Energy Dispersive X-ray (EDX), and Fourier Transform Infrared spectroscopy (FTIR), were performed to examine the morphology and the chemical composition of bitumen. To examine the behavior of the asphalt mixture, the Marshall test was conducted to evaluate the mechanical and volumetric properties. In addition, the Cantabro test was performed in dry and wet conditions to assess the durability of PA and modified NB mixtures. The results reveal that NB-G can effectively harden NB-SS and improve its overall properties. Based on chemical tests, the microstructure of NB-SS and refinery asphalt is mostly flat, but it is more complex for NB-G. Moreover, NB-G has a high content of trace elements, and new functional chemical groups are produced in modified NB samples. Additionally, 50% of NB-G is the optimal blend content. Compared to a conventional mixture, modified NB-50% has a stiffness 34% higher with lower resistance to raveling, especially in dry conditions. However, the modified NB-50% mixture is more resistant to raveling in a moisture condition by approximately 52%. In summary, using NB-G as a modifier is an effective way to enhance the properties of NB-SS and make it more appropriate to use in asphalt pavement.

Keywords: Natural bitumen, Gilsonite, Sulfur spring, Petroleum asphalt

1. Introduction

The global energy demand is anticipated to grow due to the increasing population and expanding urbanization. However, the community is becoming progressively aware of the reducing reserves of fossil fuels alongside the environmental concerns. Thus, road scientists have turned to using Natural Bitumen (NB) as a partial substitutes (up to 50%) or modifiers for petroleum asphalt and not as full replacement. However, it is difficult to meet the entire world demand for pavement construction mainly due to limited quantities, variable composition, extraction and processing costs, logistical challenges, and lack of standardized research [1]. Additionally, it is essential to pay attention to the fact that the future of NB industries may depend on various factors, such as environmental regulations, market demand, availability of alternative materials, and technological advancements. Most of the research performed using NB is in the regions of petroleum and geology [2].

The word “bitumen” originates from the language of the ancient Akkad civilization, and it indicates “asphalt” [3]. It has been noted that more than 90% of the global road network is identified to be constructed of asphaltic pavement [4]. The changes in the oil sector, especially oscillation in crude oil prices and the approach of oil depletion would directly affect the construction of asphaltic pavements. Asphalt binder is produced in both refined and natural forms [5]. Many types of asphalt are made by nature and known as “Natural Bitumen”. NB may exist naturally in many forms, such as rock asphalt, sulfur spring bitumen (NB-SS), and Gilsonite (NB-G) [6].

NB-G is made naturally and has been discovered beneath the ground from crude oil for thousands of years. During these years, the light components are vaporized while the heavy and hard components remain, including ash, wax, sulfur, non-aromatic substances, and other minerals. Approximately 100 million tons of global NB-G reserves are found in countries like the USA, Iran, Canada, Russia, Iraq, China, Venezuela, Mexico, the Philippines, and Australia [7]. The first discovery of NB-G was in the early 1860s. In 1880, Samuel H. Gilson was searching for gold and silver. Instead, he discovered a glossy black material similar to coal and began to use this bitumen in wooden pilings as a waterproof coating [8]. NB-G is a safe, non-carcinogenic, and non-toxic material compared to synthetic products. Thus, its use in many applications is increasing, especially in road pavements. The stiffness of NB-G is approximately 50 times greater than that of petroleum asphalt (PA) [9].

*Corresponding author.

Email address: roaa.hamed@coeng.uobaghdad.edu.iq

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NB-G is used to modify asphalt and can completely or partially replace polymers. Modification with NB-G provides many benefits to asphalt binders, such as improved resistance to moisture damage and reduced temperature susceptibility, which are essential for preserving pavement quality over time. Using NB-G as an asphalt modifier leads to reducing the overall cost of asphalt binder and conserving the raw materials [10].

Until now, most road authorities in Iraq have hesitated to use NB in the pavement sector due to a lack of studies. NB-G modifier is limited use in Iraqi road pavement studies, with only two published papers conducted by Erkuş et al. [11, 12]. Based on these recent studies, NB-G is used as a performance-enhancing modifier for PA binder. PA is obtained from Turkish petroleum refineries, and NB-G is supplied from the north of Iraq (Zaho region), combined with PA at 0%, 20%, 35%, and 50% by weight of PA. In addition, the performance of modified PA with NB-G was compared to 2%, 3%, and 4% SBS polymer-modified PA. Ultimately, 20% of the NB-G modified PA exhibited similar performance to 3% of the SBS-modified PA [11]. Moreover, the mechanical properties of asphalt mixtures prepared with NB-G modified-PA were examined and compared with mixtures prepared with 4% SBS modified-PA. It was demonstrated that NB-G modification significantly enhanced the performance of unmodified PA mixtures in terms of resisting the effect of repeated loads. In addition, it was proved that modifying PA with NB-G instead of SBS provided significant economic savings without detrimentally influencing the overall performance of pavement since NB-G is 20% cheaper than PA. In contrast, SBS polymer is about five to six times more expensive than PA [12].

Sulfur spring is one of the vital sources of NB in Iraq. These springs are widespread in the west and north of Iraq. The origin of this NB is petroleum. Nevertheless, over the years, the hydrocarbon components of NB-SS were evaporated since crude oil was pushed from cracks and fractures and carried with groundwater and sulfur to the surface of the earth [13]. In fact, virgin NB-SS is not used instead of PA in asphalt mixture production since its rheological properties are unstable. Thus, most previous investigations focused on treated NB-SS to conform the specification requirements of PA. Nevertheless, limited studies have evaluated the impact of several treatments and modifiers on NB-SS properties. Some of them are presented briefly as follows.

Mohsin and Latief [14] evaluated the properties of NB-SS modified with limestone filler (LSF). The NB-SS is supplied from five sulfur springs: Atffa, Mamora, Askaree, Jabal, and Attat. Test results revealed that LSF improves the resistance of NB-SS against temperature susceptibility and increases its stiffness. In conclusion, the best addition of LSF is 40% for all sulfur springs. Only for the Atffa spring is the optimal content of LSF equal to 30% since NB from this spring is less soft than other specimens.

Albayati et al. [15] examined the performance of modified NB-SS with waste engine oil (WEO) at three additions: 10%, 20%, and 30%. The experimental outcomes reveal that 20% WEO is the optimal ratio and can effectively soften NB-SS, resulting in enhancing the ductility and workability of this bitumen. In addition, Albayati et al. [16] investigated the combined effect of WEO and sugarcane molasses (SM) on the performance of NB-SS. The results showed that a high WEO ratio significantly enhanced fluidity, while SM increased toughness and cracking resistance. Chemical and structural analysis demonstrated enhanced oxidation resistance and improved internal structure of the mixtures. Grayscale resolution analysis identified the 50% NB-SS, 40% SM and 10% WEO mixture as the best mix, given its balanced properties and superior overall performance, making it promising for sustainable bio-asphalt applications.

In addition, heat treatment is conducted by Ahmed et al. [17] to harden NB-SS and to gain behavior and properties similar to PA. In this treatment, NB-SS, from the Abu-Jeer region in Hit City, is subjected to heat at 163°C for different time periods. The best physical properties that conform to the specification limits are achieved by heating NB for 20 hours since NB undergoes molecular changes during the treatment process. Additionally, the chemical properties of PA and NB-SS are significantly different.

Despite the improvement in the NB performance achieved with the heat treatment, it needs further investigation since it may not be used for practical purposes for three reasons. The first is the tremendous cost required to apply heat treatment for many hours. Meanwhile, the second is the implications of emissions resulting from treating NB, and these emissions had a negative impact on the environment. Thirdly, heating NB for a long time at high temperature can change its rheological properties due to the aging effect. Furthermore, this NB will be exposed to even more aging during the service life of the pavement. Therefore, modifying NB with other additives is more effective in avoiding the complications of heat treatment.

Recently, scientific research on exploitation NB from Iraq in the road construction sector has steadily increased. NB-SS commonly has lower hardness and requires modifiers to enhance its performance and durability during application in asphalt pavement by preventing its premature melting. Nevertheless, no previous studies have been published concerning modified NB-SS with NB-G, and the aforementioned is the motivation behind this research. To this end, the main goals of this research are to fill this gap by examining the properties of these types of NB and to satisfy the requirements of asphalt specification. NB-G is used to modify NB-SS at 40%, 50%, and 60% due to its unique compatibility with softer petroleum products.

To achieve the goals of this research, firstly, bitumen tests, including purity tests, conventional asphalt tests, and chemical tests, were conducted to evaluate the purity, physical properties, surface morphology, and elemental composition of PA, virgin NB, and modified NB. Secondly, the Marshall test was performed to assess the mechanical properties and the stiffness of mixtures. In addition, the Cantabro test was conducted to assess the resistance of PA and modified NB mixtures to abrasion impact caused by transport loads.

2. Location of natural bitumen in Iraq

About 8.4% of the world's total largest proven crude oil reserves are in Iraq. Precisely, it is ranked fifth globally after Venezuela, Saudi Arabia, Canada, and Iran [18]. The Iraqi economy mainly depends on the oil and gas industry, and massive oil reserves contribute significantly to world energy supplies. Also, abundant natural resources, including NB deposits, are widespread in Iraq. Hit, Zaho, and Ramadi are some of the cities where the largest NB deposits are located. The Mesopotamian region, also known in modern-day times as Iraq and Iran, was the first region where NB leaks were discovered. Subsequently, NB leakage has been observed in various areas of the world [19]. Typically, NB deposits in Iraq are classified into three types: NB-SS, NB-G, and rock asphalt. Due to these reserves, Iraq can be one of the central suppliers in the global NB market.

As presented above, there is a great opportunity in Iraq to use NB in asphalt pavement. In this research, NB-G and NB-SS are extracted from deposits situated in the western region of Iraq, particularly in Anbar governorate. As illustrated in Figure 1, NB-G is collected from the Gilsonite mine located in Ramadi city, and NB-SS is collected from the Askaree sulfur spring located in Hit city.

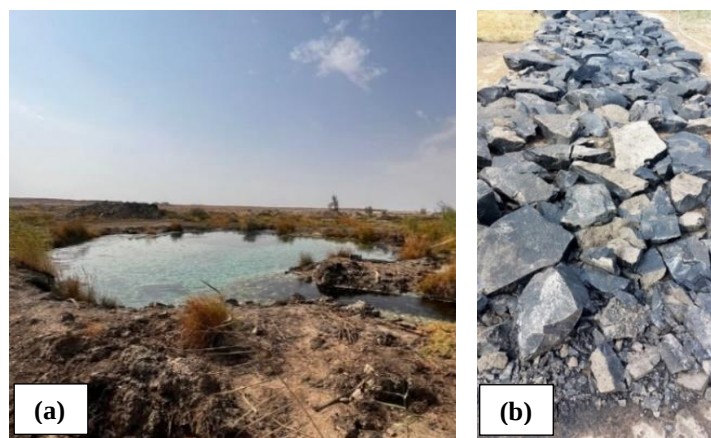


Figure 1 NB deposits: a) Askaree sulfur spring-Hit city, and b) Gilsonite mine-Ramadi city

3. Sampling and collection of NB

High-quality NB-G is required to enhance the efficiency of modification process, which can be obtained using Gilsonite with low impurity. The impurity of NB-G can vary depending on its source. Nevertheless, another problem during excavation, loading, and packing of Iraqi NB-G may increase the impurities of this bitumen. Figure 2 illustrates two samples of NB-G in lump form extracted from the Gilsonite mine. In this research, the sample in Figure 2(b) is used since it has fewer soil particles and impurities. On the other hand, heterogeneity of NB-SS is expected during the collection process from the sulfur springs. Thus, NB-SS must be collected from a high-purity sulfur spring, as illustrated in Figure 3(a). Otherwise, to improve its quality, unnecessary wastes and residue (Figure 3(b)) from the top of the sulfur spring should be removed prior to sample collection.

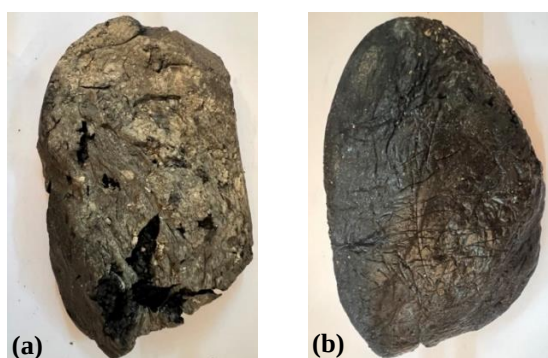


Figure 2 Purity of Natural bitumen-gilsonite: (a) low-purity and (b) high-purity



Figure 3 Purity of sulfur spring: (a) low-purity sulfur spring and (b) high-purity sulfur spring

4. Materials

4.1 Petroleum asphalt

Petroleum asphalt (PA) is a waste product derived from the distillation of crude oil. Hence, the characteristics of PA depend on the source of crude oil and the refinery operations. Typically, PA is the most common type of asphalt in the world due to its abundance, relatively low price, standardized properties, and ease of extraction. PA is supplied by Dora refinery oil, which is located in the southeast of Baghdad, Iraq. PA is presented in Figure 4.



Figure 4 Petroleum asphalt from Dora refinery oil

4.2 Natural bitumen-gilsonite

In this study, NB-G in lump form is used, as illustrated in Figures 5 (a) and (b). This is the most natural and untreated raw form. In addition, NB-G in mass is a black, solid, and brittle material that can be readily crushed into pelletized form, as observed in Figure 5(c). Also, it is very similar in appearance to the mineral obsidian, and its smell is very strong, precisely like that of heavy Gasoline.

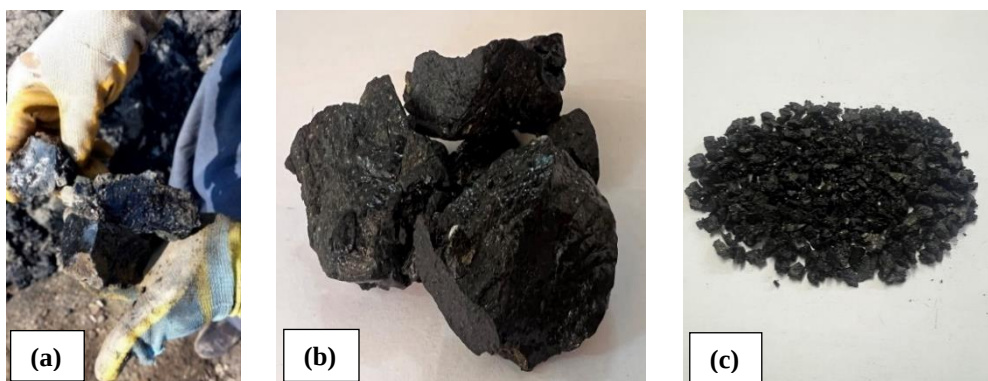


Figure 5 Natural bitumen-gilsonite: a) gilsonite lumps in mine, b) gilsonite lumps in laboratory, and c) gilsonite-pelletized form

4.3 Natural bitumen-sulfur spring

In its natural state, NB-SS is a highly viscous liquid and sticky form as it has a high content of hydrocarbons. Also, it is widely recognized for its soft texture as shown in Figure 6, shiny black color, thick, glossy appearance, and versatility. NB-SS smell is very heavy since sulfur spring is the source of Hydrogen Sulfide gas (H_2S).



Figure 6 Natural bitumen-sulfur spring

4.4 Aggregates

The aggregates, including fine aggregate, coarse aggregate, and mineral filler, were supplied from a local quarry in Baghdad, Iraq. The size of coarse aggregate was in the range of 4.75 mm to 12.5 mm, whereas for fine aggregate was in the range of 0.075 mm to 2.36 mm. Abrasion resistance of coarse aggregate was measured accordance to AASHTO T 96 standard method. Its average abrasion loss was 17.8%, which is less than 30%, the maximum permissible limit specified in road specification [20].

The mineral filler was limestone dust, which is obtained from a local factory with a bulk specific gravity of 2.72. In addition, the passing percentage from sieve size 0.075 mm is measured, and it was 88%, which is more than 70%, the minimum criteria. Figure 7 illustrates the aggregates gradation for surface layer.

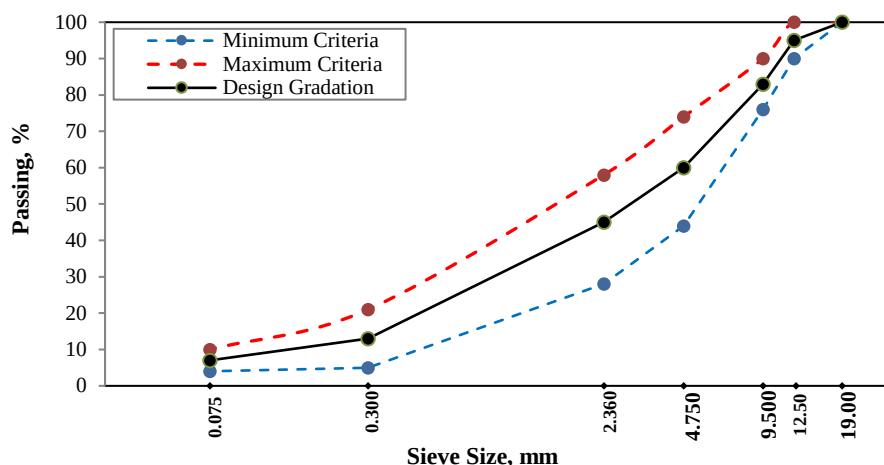


Figure 7 Aggregate gradation for design mixture-surface layer

5. Methods

5.1 Purity test

NB quality changes according to the amount of impurities combined. Therefore, a solubility test is conducted to detect the impurities in NB specimens. This test measures the solubility of asphalt in Trichloroethylene solvent, according to the standard ASTM D-2042. The purity of NB is crucial in determining the quality and export prices of NB.

5.2 Separation of water from NB

The water content in NB can be determined following test method ASTM D-95. Based on the test outcomes, NB-SS has a high water content of about 22% (by the weight of bitumen), whereas NB-G has a low water content equal to 0.514%. In this context, NB-G is not subjected to any treatment processes and is used directly in its crude form, unlike NB-SS, which is subjected to water separation processes.

Deposits with high water content, such as NB-SS from Al-Tayf deposit, have water content that is approximately equal to 37% [21]. This negatively affects NB-SS treatment as it needs more time to remove water, consequently increasing the treatment cost of NB.

Separation of water from NB-SS is a crucial step in improving its quality and facilitating the modification process with NB-G. Hence, the water separation process includes two stages: the first stage is saving NB-SS in storage containers without any treatment for periods ranging from three to seven days. Undoubtedly, the density of bitumen is higher than water. Thus, it will settle at the bottom of the container covered with water. Hence, removing water from the NB surface is easy at this stage. Nonetheless, NB-SS still has low water content and can be removed in the second stage by subjecting it to a temperature of 110°C using a convection oven for two and a half hours [22]. After these two stages, NB-SS is completely free of water, and now it is suitable to perform the modification process with NB-G.

5.3 Modification process

NB-G is added to NB-SS at various concentrations as follows: 40, 50, and 60% (by the weight of NB-SS). Compared to other modifiers, particularly polymers, there is no need for high-shear mixing [23] since NB-G incorporates perfectly and easily blends with NB-SS due to their compatibility. Initially, the NB-SS and NB-G were heated to 140°C for 30 min and 165°C for 60 min, respectively, to fluidize them [16]. Then, the NB-SS was poured into the mixer container, which has a temperature controller. Subsequently, NB-G was gradually added to NB-SS by portions and blended for 15 min at 150 rpm [24] until the mixing temperature reached 180°C. Finally, they were mixed at 180°C for 60 min after adjusted the mixing rate to 1,500 rpm [25]. Each 10 minutes, the temperature must be checked using thermometer to guarantee the melting of NB-G without causing premature aging for the NB-SS. This process ensures that samples are completely mixed together to establish a uniform and consistent bitumen.

In the case of non-uniform dispersion, chunks or large balls of undissolved material were formed and observed inside the modified bitumen at the end of the mixing process because the mixing time, temperature, or rotational rate were insufficient. For better dispersion, it is recommended to use micronized powder of NB-G (less than 0.3 mm), high-shear mixer, and sufficient blending time to ensure NB-G dissolves.

In addition, some difficulties are experienced since NB-G will quickly convert to a semi-solid form when the mixing temperature is slightly reduced to less than 180°C. After all, this bitumen contains polar molecules whose ability to disperse makes it soft at a temperature of more than 177°C [3]. Thus, a homogenous mixture cannot be obtained below this temperature, and this leads to inadequate coating of aggregates and poor compaction.

5.4 Conventional asphalt tests

It is crucial to conduct several tests on NB prior to applying the modification process to compare the properties of NB and PA. Additionally, these tests are performed for modified NB-SS to assess the impact of NB-G on its properties. Table 1 presents the asphalt tests.

Table 1 Conventional asphalt tests

Test	Unit	Method
Specific Gravity	Unit less	ASTM D-70
Penetration	0.1 mm	ASTM D-5
Softening Point	°C	ASTM D-36
Flash Point	°C	ASTM D-92
Ductility	cm	ASTM D-113
Color in Mass	Unit less	No Specification
Thin Film Oven	Unit less	ASTM D-1754

5.5 Surface morphology and elemental composition tests

The chemical characteristics of asphalt samples were investigated using Energy Dispersive X-Ray (EDX) inspection, Scanning Electron Microscopy (SEM) analysis, and Fourier Transform Infrared (FTIR) spectroscopy. EDX inspection was performed to identify the elemental composition of bitumen. Also, Thermo Scientific-Axia ChemiSEM is a high-performance imaging technique. It is designed to assess the morphology and characterization of various types of substances. In this research, this instrument is used to evaluate the microstructure of bitumen samples using distinct algorithms to process the SEM signals at 1,600x magnification.

FTIR spectroscopy was performed to compare and characterize the differences in the functional groups of asphalt specimens. Using FTIR in mineral analysis, particularly in bitumen inspection, has currently been gaining increasing attention among pavement researchers. The TENSOR-27 FTIR spectrometer from Bruker Corporation was employed in this research to obtain the spectra of the specimens. In the FTIR test, the molecules in the bitumen structure receive irradiation or power of a specific wavenumber to study the chemical interactions and establish the structure of bitumen particles. When chemical reactions occur, transmission peaks (functional groups) are observed in the FTIR spectrum. Also, the spectral range was 4,000 to 400 cm^{-1} with 0.4 cm^{-1} resolution.

5.6 Marshall test

5.6.1 Specimen preparation

The procedure of specimen preparation will be conducted in accordance with the ASTM D6926 standard. Also, the rotational viscosity test was performed at 135 °C and 165 °C following the AASHTO TP 48 standard to specify the mixing and compaction temperatures for PA and modified NB. The bitumen and aggregate were heated separately at the mixing temperature for two hours in a convection oven. The mixture components, including asphalt binder and aggregate, were first mixed for five minutes at 160°C for the PA mixture and 166°C for the modified NB mixture. Then, it was compacted at 155°C and 160°C for PA and modified NB mixtures, respectively, into cylindrical molds (diameter of 101 mm and a height of 63.5 mm) using a Marshall compactor. During compaction, 75 blows (heavy traffic level) were subjected to the top and bottom sides of each specimen using Marshall hammer. These specimens must be allowed to cool for 24 hours at room temperature. Consequently, after extracting these specimens from the molds, the volumetric properties for all specimens are computed before conducting stability and flow test.

5.6.2 Specimen testing

To obtain the values of Marshall Stability (maximum load) and flow (maximum deformation) for Marshall specimens, the test method ASTM D-6927 was followed and performed. The specimens height must be measured and these specimens are immersed in a water bath at a constant temperature of $60 \pm 1^\circ\text{C}$ for a time period varying between 30 and 40 minutes. Then, the specimens were placed inside the Marshall testing apparatus. A compressive load was applied in the diametrical direction of a cylindrical specimen at a loading rate of 51 mm/minute until it failed. The maximum load (stability) was recorded and at the same time, the overall amount of deformation (flow), is also measured.

To determine the optimum binder content (OBC) for PA and modified NB mixtures, this research considered fifteen specimens for each mixture. The OBC is determined for both mixtures using varying ratios of binder content (4, 4.5, 5, 5.5, and 6%). For each ratio, three specimens were prepared. The binder content at maximum bulk density, maximum stability, and air voids at 4% was averaged to compute the OBC.

5.7 Cantabro test

The Cantabro test indirectly evaluates the impacts of traffic abrasion, aggregate interlock or bonding, and cohesion on the raveling resistance of hot asphalt mixture. The test procedure this was performed in accordance with the AASHTO TP 108–14 standard. Two sets of samples are prepared in this study: the first set for the PA mixture and the second set for the modified NB mixture.

Marshall mold and compactor were used to prepare cylindrical samples of asphalt mixture with a height of 63.5 mm (2.5 inches) and a diameter of 102 mm (4 inches) at a heavy compaction level (75 blows). For each set, dry and wet conditions were adopted to assess the asphalt mixture response in its original state and after inducing moisture damage. The dry conditioning process required keeping these specimens for 24 hours at room temperature (25°C). Subsequently, for the wet condition, the dry-condition specimens were placed in a water bath at a controlled temperature of $60 \pm 1^\circ\text{C}$ for an additional 24 hours. Following, these specimens were left for another 24 hours at room temperature (25°C) and allowed to drain since, at low temperatures, the asphalt becomes stiffer and more susceptible to brittle fracture, leading to raveling failure [26].

Six specimens were prepared for each set (three for dry condition and three for wet condition). Before the test, the initial weight of the specimen (W_i) was recorded. Then, at a constant temperature of 25°C, one compacted specimen was placed into the Los Angeles machine drum without any abrasive loads (steel balls) and subjected to 300 revolutions, which takes approximately 9.1 minutes since the abrasion machine rotates at a regulated speed of 33 revolutions per minute. During the test, collisions occurred between the specimen and the inside metal wall of the drum. In fact, the above process approximately simulates the phenomenon of abrasion in the field. After that, the specimen is removed from the machine, and the loose particles are discarded from the surface of the specimen.

Ultimately, the final weight after the abrasion test (W_2) is measured. The percentage of Cantabro mass loss (CML) is an indicator to assess the raveling of the asphalt mixture and can be computed for both conditions by applying equation 1. A higher CML value indicates that the mixture has lower resistance to raveling. In addition, equation 2 was used to calculate the Cantabro loss index (CLI) [23]. During the Cantabro test, aggregate dislodgment and surface raveling were observed. Figure 8 illustrates the degradation of both mixtures before and after the test.

$$\text{CML (\%)} = \frac{W_1 - W_2}{W_1} \times 100 \quad (1)$$

Where W_1 is the weight of specimen before abrasion testing (gm), and W_2 is the weight of specimen after abrasion testing (gm).

$$\text{CLI} = \frac{\text{CML}_{\text{wet}}}{\text{CML}_{\text{dry}}} \quad (2)$$

Where W_1 is the weight of specimen before abrasion testing (gm); W_2 is the weight of specimen after abrasion testing (gm); CML_{wet} is the CML under wet condition (%), and CML_{dry} is the CML under dry condition (%).

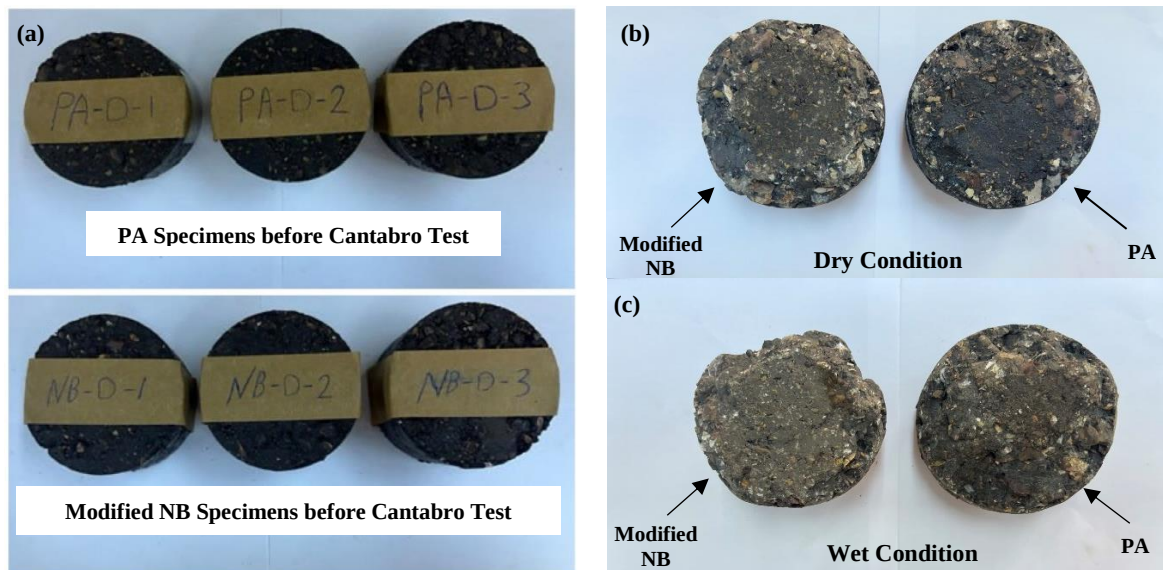


Figure 8 Cantabro test: (a) PA and modified NB specimens before test, (b) specimens after test in dry condition, and (c) specimens after test in wet condition

6. Results and discussion

6.1 NB purity

NB-SS has a high solubility in trichloroethylene solvent, about 98.6%, however, it is less than 99%, the minimum requirement [20]. Meanwhile, PA has a solubility equal to 99.52%, and that conforms to the minimum requirement. As a result, the solubility of NB-SS is slightly different than PA. Likewise, NB-G does not comply with this requirement since it has a low solubility around 73.5%. Correspondingly, NB-G has relatively high impurities and NB-SS has low impurities which approximately similar to PA. To clarify this, PA has high purity without impurities such as ash, wax, and other minerals. Moreover, the compound content of PA is controlled since PA is supplied from an oil refinery, and it undergoes further processing before being blown into the oxidation tower, where all harmful elements are segregated. Thus, the penetration and softening point parameters are controlled during the production of PA.

6.2 Physical properties of bitumen

Various physical parameters are crucial in assessing the suitability of NB for use in asphalt pavement. The physical properties outcomes are presented in Table 2. The properties of NB-G and NB-SS differ significantly due to their sources and distinct molecular structures.

Penetration degree is an indicator of asphalt hardness. Therefore, softer asphalt binders have higher penetration values and are used in colder areas. As presented in Table 2 and Figure 9, NB modified with 40%, 50%, and 60% NB-G experienced a 56.9%, 63.4%, and 72.4% reduction in penetration values, respectively. There is a remarkable increase in stiffness of NB-SS with the incorporation of NB-G. Sufficiently stiff NB is required to resist the applied loads and reduce the permanent deformation, whereas excessively stiff NB can cause cracking.

The softening point refers to the temperature at which asphalt begins to soften and flow under the application of heat. Additionally, it is a crucial parameter in identifying the acceptable application temperature range for NB and evaluating its ability to sustain temperature fluctuations during storage, transportation, compacting, and use [27].

Based on Figure 10, virgin NB-SS has a 37.9°C softening point, which is considered very low compared to 52.2°C, the softening point of PA. Thus, it flows and becomes liquid more easily at elevated temperatures than PA. In contrast, the thermal resistance is decreased with asphalt binders having lower softening point. For this reason, they are more suitable for use in colder regions. At room temperature, PA is solid and can be used in asphalt works in warm regions. Hence, NB-SS modifies with NB-G to transform it into a

stable and usable form. NB-G has a higher softening point (152°C). This makes it more appropriate for applications in regions with hot weather and high-temperature fluctuations, like Iraq. Addition of NB-G increases the softening point of NB-SS and the highest value was obtained when modified NB-SS with 60% of NB-G. In general, the addition of NB-G increased the softening point and reduced the penetration for modified NB-SS. In summary, PA and modified NB with 50% demonstrate slight variations in penetration and softening point.

Table 2 Physical properties results

Property	Bitumen Type						Specification Limits [20]	
	PA	NB-SS	NB-G	Modified NB-SS				
				40% NB-G	50% NB-G	60% NB-G		
Penetration, 0.1 mm	46	123	0	53	45	34	40-50	
Softening Point, °C	52.2	37.9	152	50.7	52.4	55.7	Not Specified	
Specific Gravity, unit less	1.0400	1.0603	1.0796	1.0659	1.0669	1.0678	Not Specified	
Flash Point, °C	249	176	348	225	239	252	232 minimum	
Ductility, cm	112	167	2	122	111	97	100 minimum	
Thin Film Oven	Retained Penetration, %	57	53	100	68	67	74	55 minimum
	Ductility, cm	81	82	0	78	66	54	25 minimum
	Color in Mass, unit less	Black	Black Shinny	Dull Black	Black Shinny	Black	Black	Not Specified

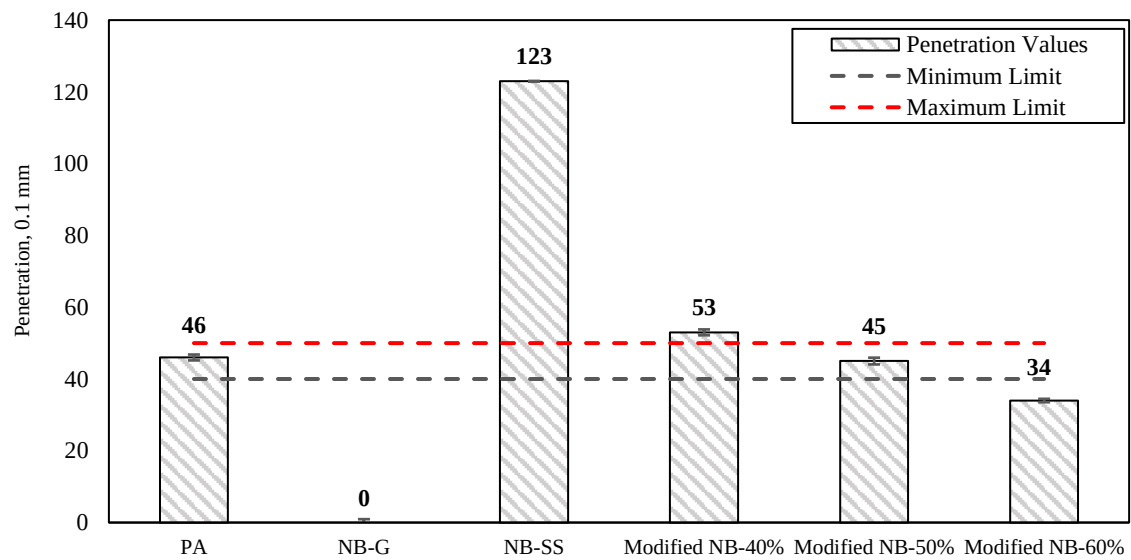


Figure 9 Effect of NB-G modifier on the penetration of NB-SS

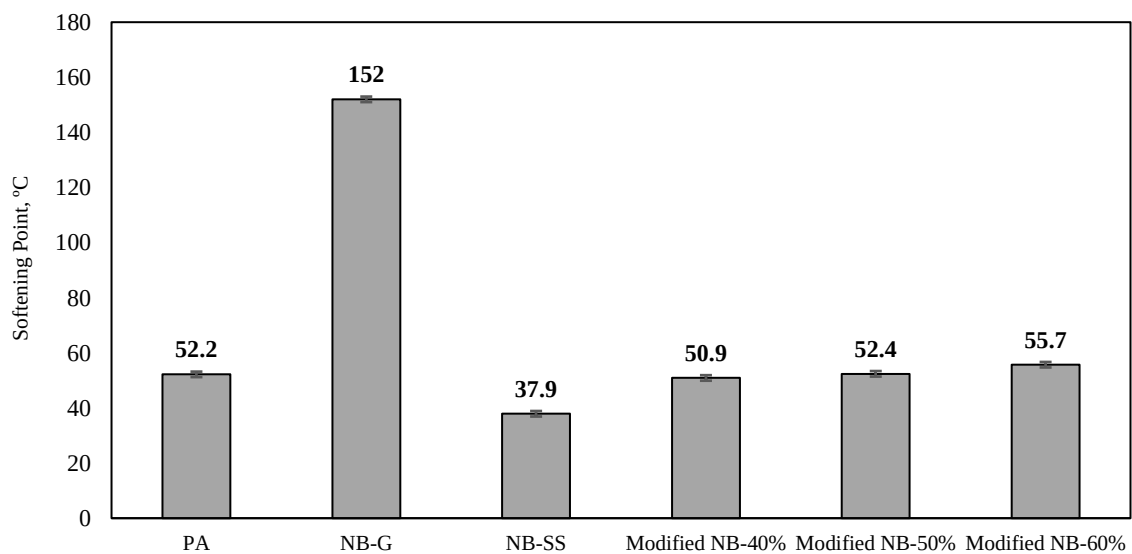


Figure 10 Impact of NB-G modifier on the softening point of NB-SS

Bitumen binder is a very sensitive material that is resistant to temperature fluctuations. Temperature susceptibility means the responses of bitumen to changes in temperature, and it is evaluated by computing penetration index (PI), which is a quantitative indicator that depends on the results obtained from penetration and softening point tests. The PI indicator is calculated by applying the equations below:

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

$$A = \frac{\text{Log Penetration at } T - \text{Log } 800}{T - SP} \quad (4)$$

Where T is the temperature of penetration test (°C) and SP is the softening point temperature for bitumen (°C). The PI indicator ranges between -2 and +2 (in most cases) for conventional PA binders. In this study, as displayed in Figure 11, PI is -0.854 for PA binder, and it is within the normal range. Bitumen has a high susceptibility to temperature change if the PI value decreases less than -2, like NB-SS, which has a PI equal to -2.774. Conversely, in the cases of PI values exceeding +2, bitumen will be less susceptible or sensitive to high temperatures, like NB-G, as it exhibits abnormally low temperature susceptibility. Nevertheless, the PI equation is inapplicable for NB-G since its penetration equals zero.

NB-SS has an unusually high sensitivity to temperature. It is necessary to enhance its response to high temperatures by modifying with NB-G. The results reveal that modified NB-SS with 60% NB-G has the maximum PI (-0.723), and modification with 40% NB-G achieves the minimum PI (-0.891). This suggests that increasing the addition of NB-G makes NB-SS less sensitive to high temperatures. Also, an additional 50% NB-G accomplished a PI equal to -0.856, which is approximately similar to the PI of PA. Thus, they have the same response to high temperatures.

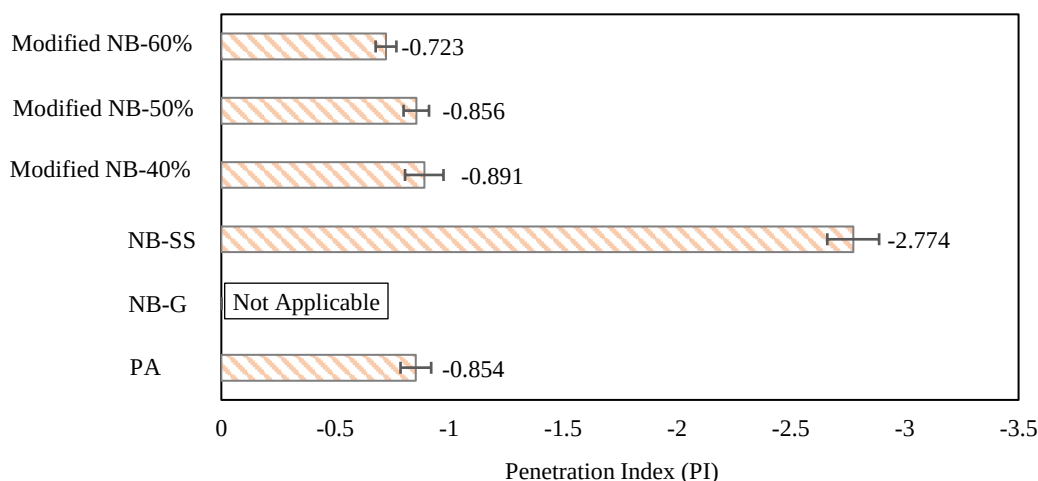


Figure 11 PI values for PA, unmodified NB, and modified NB

Specific gravity is an essential parameter to determine the density and assess the quality of NB. It can vary depending on the deposit and processing methods since specific gravity is affected by the impurity content and composition of NB. Both types of NB have a high specific gravity, more than PA, demonstrating that NB is denser due to containing heavy hydrocarbons in their composition since light hydrocarbons evaporate over time when exposed to air, pressure, and sunlight. Hence, NB is more durable and resistant to deformation. Moreover, PA has low specific gravity, and this may be related to the presence of lighter hydrocarbons in refinery bitumen, which makes PA less stable. An increase in the specific gravity of modified NB was observed. This is attributed to the fact that NB-G has a higher specific gravity than NB-SS since it contains a high wax content.

In addition, NB-G enhanced the flash point of NB-SS since it has a high flash point (348°C), thus reducing the risk of explosion and fire hazards. Also, the ductility of modified samples was reduced due to the hardness of NB-G. After short-term aging with a thin film oven test, all modified samples have considerable resistance to the combined impacts of air and heat since the retained penetration and ductility values are higher than the specified limits.

In conclusion, considering all physical properties of modified NB samples, it was revealed that the best results were obtained at the addition of 50% of NB-G, as they complied with the specification requirements for PA. Although the NB-G modifier led to enhance the properties of NB-SS, it is essential to understand that NB modification cost may not provide an economical solution. Thus, it is recommended to carry out the cost-benefit analysis [28, 29] in future investigations. This analysis helps in evaluating the economic loss or benefit obtained for the modified NB-SS.

Using MINITAB V17 software, a statistical analysis was also performed to determine whether the differences among the three modification levels (40%, 50%, and 60% NB-G) were statistically significant. A one-way Analysis of Variance (ANOVA) was conducted on the triplicate measurements of penetration, softening point, ductility, and flash point, as listed in Table 3. Furthermore, the coefficient of variability (CV) for each property was determined. It was considered acceptable, and this implies that the experimental data had high precision and consistency. The obtained results summarized in Table 4, indicate that all p-values were well below 0.05, confirming that the NB-G dosage had a statistically significant effect on the measured properties. The matter which demonstrates that the observed improvements in consistency, thermal susceptibility, and ductility are not due to random laboratory variation but are directly attributed to the increasing NB-G content. Particularly, the 50% NB-G blend consistently provided a balanced improvement in hardness, temperature resistance, and ductility compared with the 40% and 60% blends. While the 40% NB-G binder remained relatively soft, the 60% NB-G binder became excessively stiff and brittle. Therefore, the statistical evidence supports the selection of

50% NB-G as the optimal modification level, as it achieves the most desirable and balanced combination of physical properties while remaining within specification requirements.

Table 3 The triplicate measurements for each property

Property	Replicate No.	Bitumen Type					
		PA	NB-SS	NB-G	NB-40%	NB-50%	NB-60%
Penetration, 0.1 mm	1	45	122	0	52	44	34
	2	47	122	0	54	46	34
	3	46	124	0	53	44	35
	CV, %	2.17	0.94	-	1.89	2.59	1.68
Softening Point, °C	1	52.2	37.7	153	50.7	52.2	56.1
	2	52.4	37.8	152	51.0	52.3	55.6
	3	52.0	38.2	152	50.4	52.7	55.4
	CV, %	0.38	0.70	0.38	0.59	0.50	0.65
Ductility, cm	1	116	168	2	118	117	101
	2	112	165	2	122	106	96
	3	108	168	3	126	110	94
	CV, %	3.57	1.04	24.74	3.28	5.02	3.72
Flash Point, °C	1	240	179	341	232	244	249
	2	255	169	359	216	241	256
	3	252	180	344	227	232	257
	CV, %	3.19	3.46	2.77	3.64	2.61	1.72

Table 4 ANOVA results for the effect of NB-G dosage on physical properties of NB-SS

Property	F-Statistic	p-Value	Statistical Significance
Penetration	295.12	1.02×10^{-6}	Significant ($p < 0.05$)
Softening Point	200.59	3.20×10^{-6}	Significant ($p < 0.05$)
Ductility	23.55	0.00144	Significant ($p < 0.05$)
Flash Point	15.14	0.00452	Significant ($p < 0.05$)

6.3 Morphology and elemental composition analyses

6.3.1 Morphology inspection

Microstructural examination was performed and Figure 12 (a-f) presents the SEM images of PA, unmodified NB, and modified NB samples. From observation, there is a slight difference in surface morphology between PA and NB-SS since they have approximately the same structure (uniform microstructure), as displayed in Figures 12 (a) and (b). However, NB-SS has a dull black color as it has high water content. Nevertheless, the morphology structure of NB-SS is continuously variable which is mainly due to its natural formation process in sulfur spring deposit and the source-crude oil composition [3].

Also, it is observed that NB-G specimen has a rough surface and complex structure compared to PA and NB-SS (Figure 12c). For more clarity, it has a dense structure with curved lines feature and smooth conchoidal fracture arrangements, similar to those patterns of obsidian material. Additionally, NB-S appears as a continuous and single organic phase, especially in these fracture surfaces. The dense morphology of NB-G is related to its chemical composition since it mainly contains high contents of asphaltenes [30].

SEM imaging is effective and successful in observing the microstructural characteristics of modified NB. When the addition of NB-G is increased, the SEM image of modified NB demonstrates the existence of multifaceted and polyhedral particles with smooth and flat surfaces. This indicates that addition NB-G to NB-SS significantly changes the microstructure or morphology of NB-SS, as shown in Figure 12 (d) and (e). Finally, with addition of 60% NB-G, the characteristics of the NB-SS structure changed remarkably (Figure 12f). This means that NB-G has a dominant impact on the NB-S morphology.

6.3.2 Elemental composition analysis

In general, bitumen, either PA or NB, is recognized to comprise heterocyclic ingredients that consist mainly of carbon (C), hydrogen (H), sulfur (S), oxygen (O), and nitrogen (N). In addition, it is a visco-elastic substance comprised of a complex combination of asphaltene, aromatic, saturate, and resin with some trace elements such as magnesium (Mg), iron (Fe), copper (Cu), nickel (Ni), silicon (Si), calcium (Ca), chlorine (Cl), aluminum (Al), vanadium (V), and others. Trace elements and their contents play an essential role in the physical and chemical features of bitumen since they are mainly associated with the asphaltene fraction. This fraction is responsible for the viscosity of bitumen. The existence of trace elements can facilitate the bonding and clustering of the asphaltene particles. Elemental analysis using the EDX technique has been conducted. The findings of EDX are presented in Table 5.

In this study, four elements, Fe, Ni, Si, and Cl, are reported to be present in all bitumen samples. Also, it was noticed that each bitumen sample comprised different groups of trace metals, and the total number of metals varied from five to nine. Among all the elements detected, Cu (4.6%), Ni (1.8%), and Si (1.4%) are revealed to be the most abundant trace elements in PA. The appearance of these elements and the variation in their concentrations depend on the crude oil source. Meanwhile, Ca (7.4%), Si (3.8%), Cu (2.2%), and Ni (2.1%) are the most trace elements in the NB-G sample. This is attributed to the fact that Gilsonite has high ash content. Ash content in NB consists of inorganic components and waste substances like sulfates, clays, sulfides, etc. After combustion, these substances remain as ash. The major elements in ash are calcium, potassium, aluminum, magnesium, sulfur, iron, sodium, phosphorus, silicon, and titanium [31].

Very low amounts of trace elements were observed in NB-SS sample. In summary, Cu, Ni, and Ca have the highest contents in PA, NB-SS, and NB-G, respectively. NB-G is very rich in Ca, with a concentration of 7.4%. Meanwhile, PA and NB-SS do not contain

any detectable amount of Ca. The molecular weight and softening temperature of NB-G depend on the composition and the age of the mine.

Also, there is a significant divergence in the presence and the concentrations of some trace elements between these two types of NB. In other words, NB-G contains trace elements such as Cu, Mg, Al, and Ca, which do not appear in the element analysis of NB-SS. Iraqi NB-G is higher in C (64.1%) and also contains S (9.3%) and O (8%). Additionally, it can be observed that NB-SS demonstrated affinity with PA in element analysis. It is apparent that NB-SS is mainly of C (76.1%), and an approximately similar value is reported for PA (74.0%).

It can be observed that NB-G is characterized by a lower content of S (9.3%) compared to NB-SS (17.3%) and PA (12%). One of the major concerns of petroleum refineries is sulfur (S) since it is a highly poisonous element and can cause plant rusting and foul-smelling corrosion. Generally, most domestic oil crudes are free from dissolved H_2S . This indicates that NB-G seems to be of better quality than NB-SS. On this basis, NB-G may be considered an effective additive for NB-SS and PA modification. Regarding S quantity in modified NB-50% (9%), it is 25% and 48% lower than PA and NB-SS, respectively.

Moreover, NB-G exhibits the highest O concentration (8%), followed by NB-SS (5.6%), and then PA (5%). A large part of oxygen appears as phenols with minor amounts of carboxylic acids. Returning to the sulfur spring, the high concentration of O for NB-SS was related to the oxidation of the uppermost layer at the outcrop, especially in the early stages of development of these deposits. NBs are characterized by a high oxygen content as compared with PA.

Some trace elements are present in very small amounts, such as Mg, V, Cl, and Al. However, their existence is still very effective in the molecular interactions within the bitumen structure. Thus, for modified NB, the contents of trace elements are used as an indicator to evaluate the modification efficiency. The element composition of modified NB samples is variable, and no trends are observed when comparing one sample to another. However, it was noted that a decrease in C and S is associated with an increase in O, which is the most pronounced change induced by modification. Conversely, the differences in element contents are principally related to the content of NB-G and the efficiency of chemical reactions during modification.

A recent study [14] evaluated the element components of NB-SS. However, there are many discrepancies in the findings of the previous study. The reason might be sampling NB at different times and from different places within the sulfur spring and the Gilsonite deposit. Also, the purity of NB plays an essential role in the concentrations of elements.

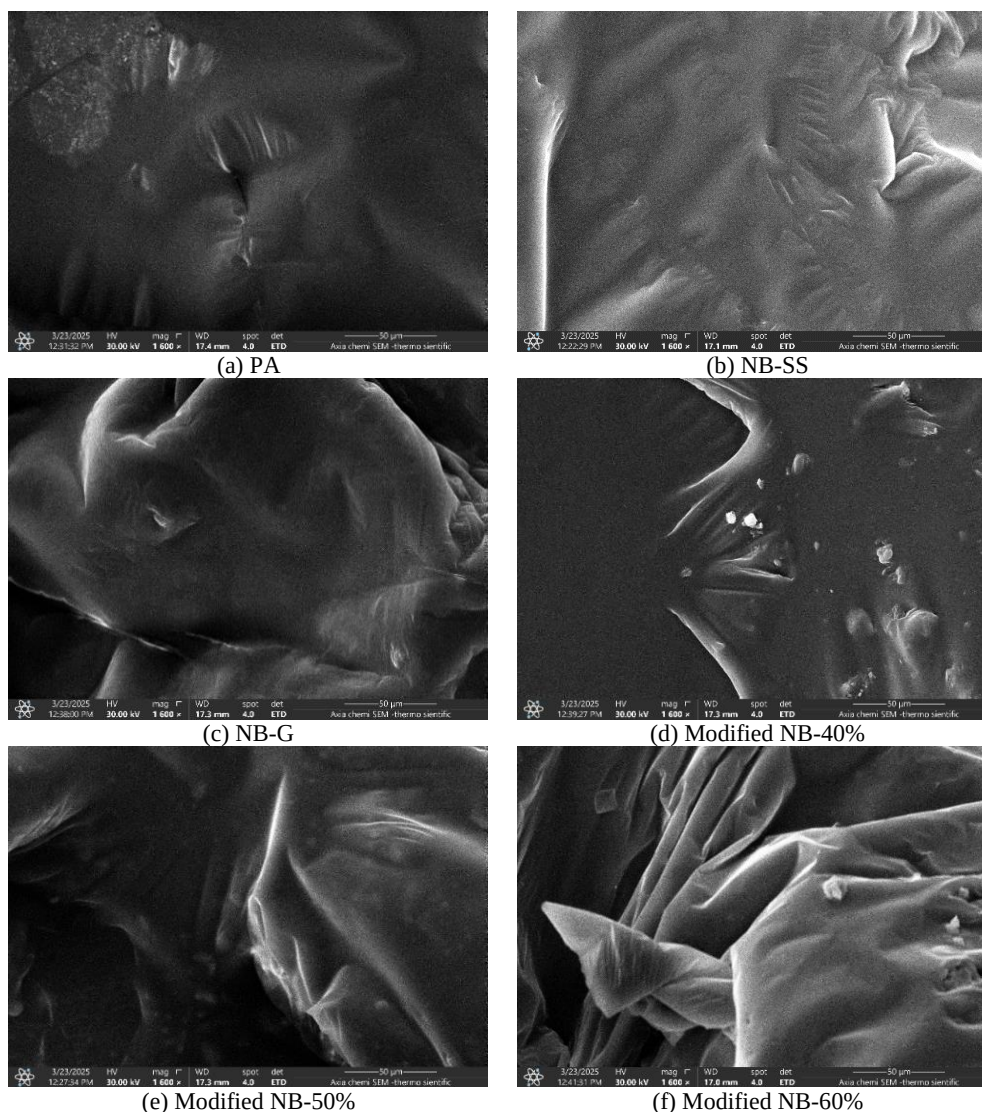


Figure 12 The morphology structure of NB-SS modified with NB-G

Table 5 Elemental analysis of PA, unmodified NB, and modified NB

Element	Bitumen Type					
	PA	NB-SS	NB-G	Modified NB-40%	Modified NB-50%	Modified NB-60%
	Weight, %					
Carbon (C)	74.0	76.1	64.1	73.0	69.6	67.8
Oxygen (O)	5.0	5.6	8.0	5.8	10.2	12.3
Sulfur (S)	12.0	17.3	9.3	15.6	9.0	7.1
Silicon (Si)	1.4	0.3	3.8	0.9	2.1	2.0
Chlorine (Cl)	0.1	0.1	0.8	0.4	0.4	0.5
Vanadium (V)	0.1	0.1	0.0	0.0	0.0	0.0
Iron (Fe)	1.0	0.2	1.3	0.7	1.7	1.3
Nickel (Ni)	1.8	0.3	2.1	0.7	1.7	1.6
Copper (Cu)	4.6	0.0	2.2	1.4	3.7	3.4
Magnesium (Mg)	0.0	0.0	0.5	0.1	0.1	0.3
Aluminum (Al)	0.0	0.0	0.5	0.0	0.0	0.0
Calcium (Ca)	0.0	0.0	7.4	1.3	1.5	3.7

6.3.3 Functional chemical groups

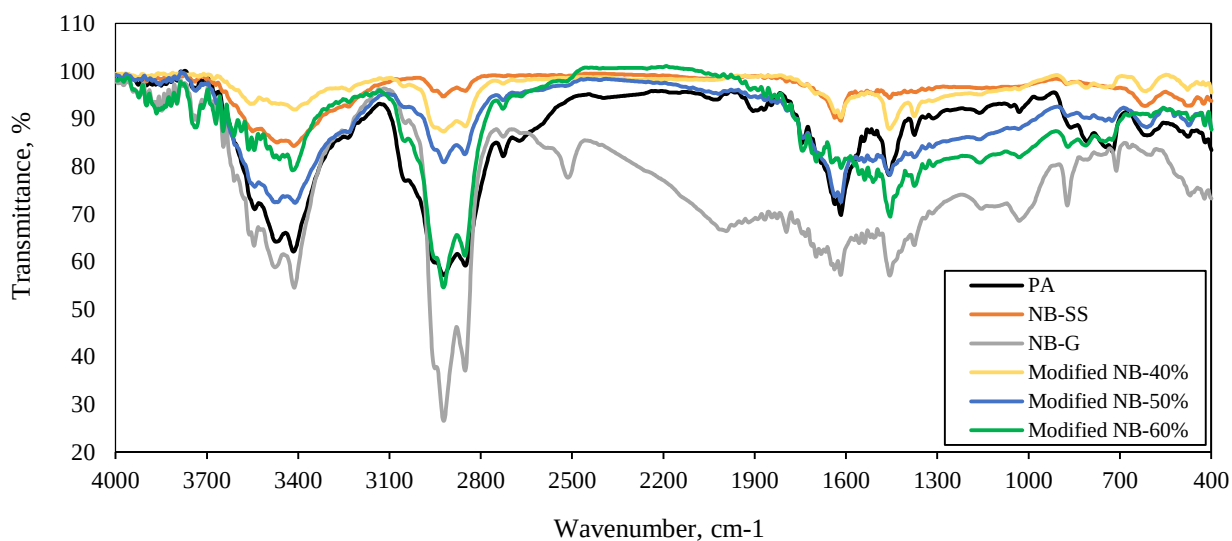
The absorbance spectra (FTIR) of NB types and PA have been studied and presented in Figure 13. Except for NB-G, which has a peak at $1,990\text{ cm}^{-1}$, it can be observed that the shape of the FTIR spectra is very similar from $2,500$ to $1,800\text{ cm}^{-1}$. However, in the range of wavenumbers between $1,900$ and $1,300\text{ cm}^{-1}$, the spectra of PA, virgin NB, and modified NB have different shapes. However, all reveal peaks at $1,373$, $1,450$, and $1,616\text{ cm}^{-1}$, which denote, respectively, the S=O stretch of the sulfoxide group, the CH_2 and CH_3 bend of the aliphatic group, and the C=C stretch of the aromatic skeleton. The peaks at the wavenumber range of 900 to 625 cm^{-1} appear in all samples and result from the bending vibration of C-H in the aromatic. This conforms to the findings of the recent study [32]. These similarities are due to the fact that both PA and NB are composed of hydrocarbons. Nevertheless, NB-G demonstrates an additional peak with respect to other samples. Specifically, a sharp peak at $2,511\text{ cm}^{-1}$ can be observed, which is completely absent in PA and other NB specimens' spectra.

Similar to the PA spectrum, the wavenumber of $1,028\text{ cm}^{-1}$ appeared in NB-G, NB-SS, and modified NB due to the stretching of the sulfoxide group. This finding is aligned with the results of a previous investigation [33]. The spectra of all samples had a band that was observed approximately in the wavenumber range of $3,800$ to $3,500\text{ cm}^{-1}$, which is dedicated to the vibration of free water or band water (OH) as well as CO_2 in ambient air. Due to the presence of OH, the chemical composition of the NB-SS can change when modified with NB-G. In addition, the strong peaks around $2,850$ to $2,922\text{ cm}^{-1}$ are associated with the stretching vibrations in the aliphatic chain of the bitumen. The outcomes of the previous investigation [34] supported this conclusion.

The change in the chemical composition of modified NB specimens can be determined by comparing the absorbance of the functional groups. A carboxylic acid has a carbonyl stretch $\text{C}=\text{O}$, and this stretch appears in the band from the wavenumber $1,760$ to $1,690\text{ cm}^{-1}$. The position of this band probably depends on the internal hydrogen bonding of carboxylic acid. The transmittance of the functional group $\text{C}=\text{O}$ for NB-SS is higher than other specimens. This indicates that the oxygen-containing functional groups, such as esters, ketones, aldehydes, and carboxylic acids, are formed during the oxidation process of NB-SS in sulfur spring.

By comparing the FTIR spectra of PA and NB samples, an absorption peak appears in NB samples at a range of 480 to 420 cm^{-1} , which corresponds to naphthalene functional groups. This group helps improve the structural strength of bitumen molecules, resulting in increased bitumen viscosity and enhancing its cohesion and adhesion with the aggregate surface.

Modified NB experienced changes in functional groups since the modification process resulted in the disappearance of some prominent peaks and produced new peaks in these specimens. This implies that the modification between NB-SS and NB-G is a chemical process. In other words, virgin NB-SS and modified NB have different peaks. This finding reveals that the addition of NB-G changes the proportion of each component in NB-SS due to the impact of the chemical reaction between them.

**Figure 13** Impact of incorporating NB-G modifier on the chemical properties of NB-SS

6.4 Marshall properties

Asphalt mixtures must meet a set of criteria and limits specified to control the production, laying operations, and service performance. Hot asphalt mixture requirements for the surface layer have been provided in Table 6. The mechanical and volumetric properties of PA and modified NB-50% mixtures are also listed in this table. It was observed that the properties of both mixtures were within acceptable limits.

OBC was determined to be 4.95 and 5.2% for PA and modified NB-50% mixtures, respectively. These OBC values fulfill all the criteria requirements as presented in Table 6. As expected, the modified NB-50% mixture achieved higher Marshall Stability compared to the PA mixture, with an increase of approximately 16%. On the other hand, the flow value and the air voids ratio of the modified NB-50% sample were less than those of PA, with a reduction percentage of nearly 12% and 4%, respectively. This confirms that the modified NB-50% is more rigid than PA.

Moreover, the Marshall Stiffness, also known as rigidity ratio or Marshall Quotient, is a preliminary evaluation of the mechanical characteristics of the bitumen mixture, and it can be computed for each mixture by dividing the stability over flow. This nearly represents the ratio of the maximum applied load to the maximum deformation at specific conditions to measure the resistance of bitumen mixture to permanent deformation in service. Excessive Marshall Stiffness means that the bitumen mixture has a brittle or stiff structure, and the cracks can occur under loading conditions [35]. In this research, it has been concluded that Marshall Stiffness for modified NB-50% mixture is higher than PA mixture with increasing rate of 34%. Regarding the aforementioned, this is another proof that the modified NB-50% mixture is more rigid than conventional (PA) mixture. This is because addition NB-G tends to strengthen the NB-SS matrix due to its high hardness.

Using modified NB-50% had a slight effect on the bulk density property of the bitumen mixture since modified NB-50% is denser than PA. This is attributed to the fact that both NB types have higher specific gravity than PA. In addition, a comparison between both mixtures exhibited a decrease in the voids in mineral aggregate value and an increase in the voids filled with bitumen value for modified NB-50%. The above findings demonstrate that the modified NB-50% mixture is stiffer and denser than the PA mixture.

Table 6 Properties of PA and modified NB-50% mixtures

Design Parameter	Unit	Mixture Type		Criteria Requirement [20]
		PA	Modified NB-50%	
Optimum Binder Content (OBC)	%	4.95	5.20	4-6
Marshall Stability	kN	11.24	13.03	8 (minimum)
Marshall Flow	mm	3.54	3.11	2-4
Marshall Stiffness	kN/mm	3.2	4.3	Not Specified
Bulk Density	gm/cm ³	2.334	2.343	Not Specified
Air Voids Ratio	%	4.10	3.94	3-5
Voids in Mineral Aggregate Ratio	%	14.23	14.17	14 (minimum)
Voids Filled with Bitumen Ratio	%	71.20	72.19	Not Specified

6.5 Abrasion loss resistance

For high-traffic highways, the recommended maximum limits of CML under dry and wet conditions are 20% and 35%, respectively. These limits were used to assess the durability of the asphalt mixture in terms of raveling resistance with the impact of traffic and environmental conditions [36]. Table 7 provides the CML results at dry and wet conditions for PA and modified NB-50% mixtures.

According to Table 7, all CML results for both PA and modified NB-50% mixtures were confirmed to the maximum permissible limits indicated for the Cantabro loss in dry and wet conditions. Additionally, it can be noticed that CML values of the specimens at dry conditions were higher than the values of specimens at wet conditions (the most critical situation) for both mixtures, which is as expected since the specimens exposed to moisture are weaker than those in dry condition.

Specifically, modified NB-50% mixture has 17.8% and 27.8% average CML for dry and wet conditions, respectively, which are higher than those of PA mixture (8.2% for dry condition and 26.7% for wet condition). This indicates that modified NB-50% mixture has less resistance to raveling and is less durable than PA mixture. The deterioration in durability, in both conditions, is not expected as the modified NB-50% mixture becomes more prone to disintegration. It is attributed to the fact that the interaction between NB and aggregate in the modified NB-50% mixture is less effective than PA mixture. In other words, modified NB-50% covers the aggregate with excessive film coating, as well as allowing the shifting of bitumen from the outside surface of aggregate. This weakens the cohesion power inside the bitumen matrix and the adhesion connection between bitumen and aggregates, resulting in a weaker internal structure. This finding is consistent with results from the recent research [37].

In addition, the variations in CML results may be partially caused by the rate of water accessibility in air voids. Nonetheless, in this study, no significant effect of this parameter was observed as the air void contents of PA and modified NB-50% specimens presented slight differences based on the data provided in subsection 6.4. In general, lower air void content leads to lower CML, particularly in moisture conditions [38]. On the other hand, CML values were affected negatively by the presence of unbonded or crushed aggregate particles induced by compaction, resulting in an asphalt mixture more prone to disintegration distress. This phenomenon does not influence the data comparison in this work since both mixtures were prepared with the same type and gradation of aggregate.

Further analysis was conducted to evaluate the abrasion resistance of PA and modified NB-50% mixtures using CLI, which provides a better evaluation of the water impact on the bitumen mixture. Notably, lower CLI values indicate that the bitumen mixture has greater durability under wet condition. As can be observed in Table 7, the CLI is 1.56 for modified NB-50% mixture, which is much less than 3.26, the CLI for the PA mixture. Thus, the water impact on the modified NB-50% mixture demonstrates the highest abrasion resistance against water action compared to PA mixture.

Table 7 Summary of cantabro loss test data

Mixture Type	Dry Condition			Wet Condition			CLI
	Specimen	CML, %	Average CML, %	Specimen	CML, %	Average CML, %	
PA	1	8.5	8.2	1	28.9	26.7	3.26
	2	8.2		2	26.7		
	3	7.9		3	24.5		
Modified NB-50	1	18.2	17.8	1	25.8	27.8	1.56
	2	17.7		2	28.3		
	3	17.5		3	29.3		

7. Conclusions and future recommendations

The research investigated the impacts of using two types of NB obtained from sulfur springs and Gilsonite deposits in asphalt pavement. Considering the analysis and comparison of the results obtained from the experimental tests, the following findings can be drawn:

- NB-G has unique properties that make it a suitable and perfect choice for NB-SS modification. It increases the stiffness and enhances the temperature susceptibility of NB-SS, resulting in improving the NB-SS's ability to resist deformation, yet at a slightly higher risk of cracking. Therefore, it is recommended that modified NB be applied in areas with high temperature climates. As observed, the physical properties of modified NB-50% are quite similar to those of PA. This is due to the fact that NB-G is an excellent hardening and reinforcing agent.
- The SEM examination emphasizes unique morphologies of both NB types. NB-SS has a relatively uniform and smooth surface microstructure similar to PA. Whereas, NB-G has a complex, irregular, and rough structure, indicating a heterogeneous matrix with a higher impurity level relative to NB-SS and PA.
- PA and NB-SS have approximately similar chemical compositions. On the other hand, NB-G has a high ash content since it contains many trace elements with significantly higher content than NB-SS and PA. Both NB types vary significantly in their chemical composition, which provides a base for identifying and characterizing them. The modification process changes the chemical composition of the modified NB.
- Based on FTIR analysis, two sharp peaks dominated the spectrum of NB-G. The dominant peaks were methylene stretches at around 2,922 and 2,850 cm^{-1} . This implies that the NB-G contains an appreciable amount of aliphatic hydrocarbons. In addition, new functional chemical groups are produced when NB-SS is modified with NB-G.
- Relative to the PA mixture, using modified NB-50% as a new binder notably increased the Marshall Stability of the bitumen mix by 16% and improved its stiffness by 34%, resulting in higher resistance to permanent deformation. Nevertheless, higher stiffness may cause crack failure. In addition, modified NB-50% leads to a slight reduction in air voids, improving mix compactness and durability.
- PA has adhesion to aggregate better than NB since modified NB-50% mixture achieved higher CML than PA mixture in dry and wet conditions, which is inconsistent with the expected behavior since modified NB-50% gains greater stiffness than PA. However, modified NB-50% mixture has a lower CLI value, implying that it is more resistant to abrasion in a moisture condition. Finally, modified NB-50% was not as promising in terms of having the desired raveling resistance.
- This paper is a starting point for future investigations on the use of NB-SS and NB-G in local road pavement. Nevertheless, it is subject to two main limitations. First, achieving consistent findings may be challenging since the composition and properties of NB are continuously varied depending on the deposit's location and weather conditions. Therefore, further studies can be performed to assess these changes in NB properties. Second, Gilsonite usually has a significant influence on the low-temperature cracking resistance of asphalt mixture, and this must be considered in future works. Finally, performing field trials is crucial and recommended to bridge the gap between field and laboratory performance.

In summary, this research provides a limited evaluation of modifying NB-SS with NB-G at three addition ratios: 40, 50, and 60%. Using modified NB enhances the strength of asphalt pavement against permanent deformation and achieves a limited improvement in raveling resistance under moisture conditions. Thus, NB may be a good alternative to PA for road pavement, which can reduce the dependency on PA and help to decrease carbon emissions.

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