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Residues from The Mishraq Sulfur Mine and The Nano-Sulfur Derived from Them as Modifiers for The Mechanical and Thermal Properties of Iraqi Asphalt

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Abstract: This study included the modification of asphalt using sulfur waste (foam) resulting from the purification of sulfur from the Al-Mishraq field through a chemical oxidation method. In addition, it utilized nano sulfur prepared from these wastes in the modification of asphalt. The study initiated with a comprehensive analysis of foam composition and employed various diagnostic techniques. It then proceeded to prepare calcium polysulfide, sodium polysulfide and potassium polysulfide from the foam, followed by the preparation of nano_sulfur from these salts. The sulfur's nano-sized particles were characterized using scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX) to determine sulfur content, and the particle size range was (83.40nm), (45.13nm) and (38.51nm). The prepared nano-sulfur was used to modify the properties of Iraqi asphalt. The original and modified asphalt's rheological properties were determined by measuring properties such as Ductility, softening point, penetration, penetration index, Marshall stability, chemical immersion, and aging. The modified asphalt showed rheological properties that qualified it for use in paving operations, especially in terms of resistance to acid rain and stability.

Keywords: Asphalt, Nano_sulfur, Foam, Polysulfide.

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1. INTRODUCTION

Asphalt is a complex mixture of various hydrocarbon compounds with high molecular weights, in addition to containing atoms of sulfur, oxygen, and nitrogen (1,2). Asphalt is widely used in road paving due to its high adhesion to different metals and its viscosity property, in addition to its low cost (3). Traditional asphalt mixtures can perform well in paving operations. However, in recent years with the growth of traffic volume, heavy loads, harsh weather conditions and economic cost, all of this has led to an increased demand to improve the mechanical properties of traditional asphalt materials by modification using various high-quality and more reliable additives (4-7). Referring to previous studies, there have been many recent studies that focused on modifying asphalt and improving its specifications using various additives, including sulfur, by increasing the viscosity of asphalt binders, thus improving the adhesion of asphalt to aggregates and crushing. And resistance to aging (8-11).

(Al-Hadidy, 2023) (13) was able to use sulfur waste in asphalt mixtures, as the resulting binders were subjected to tests of penetration, ductility, softening point, absolute viscosity, modulus of elasticity, and durability. The study showed, in general, good results conforming to the standard specifications for Iraqi asphalt (12).

Abass and Hamdoon (2021) (12) used a mixture of synthetic polymer (EVA) and natural polymer (wood) to modify asphalt with the addition of 1% by weight of sulfur at a temperature of 180 degrees Celsius for one hour. The rheological properties of the modified samples were measured. The study gave good results in the field of asphalt resistance to deformation processes (13).

Nguyen & Le studied the effect of sulfur on the performance of asphalt mixtures by investigating the engineering and morphological properties of sulfur-modified asphalt using various tests, scanning electron microscopy (SEM) was also used in addition to conducting Marshall stability testing and indirect tensile (IDT) testing to evaluate the resistance of sulfur-modified asphalt mixtures. This study found that sulfur-modified asphalt leads to significant improvements in the mechanical properties of asphalt and crack and fatigue resistance (14).

(Zhang, J., & Tan, Y) (15) studied the rheological properties of asphalt by adding sulfur and styrene-butadiene-styrene (SBS), The results showed an increase in compatibility between the additives and asphalt in addition to improved performance of asphalt after adding sulfur to SBS-modified asphalt (15).

Zhang et al, 2022 were able to use four percentages of sulfur (0%, 15%, 30%, 45%) by weight of the asphalt binder. It was noted from the results that adding sulfur at a low percentage, such as 15%, leads to softening of the asphalt binder (16).

Singh et al. (2020) (17) and his team were able to improve asphalt properties represented by penetration, viscosity, Ductility and increased aging resistance, as well as Marshall stability through the addition of sulfur to asphalt and its use in asphalt modification (17).

This study attempted to benefit from the wastes from sulfur refining in Al_Mishraq mine (Nineveh, Iraq) resulting from chemical treatment represented as (foam) in an attempt to produce asphalt with rheological properties suitable for the environmental conditions in our country Iraq – Nineveh.

2. EXPERIMENTAL SECTION

2.1. Preparing The Raw Material

The raw material used in this study, which consists of the foam material, was obtained from the Mishraq Sulfur mine (Mishraq Sulfur Company) in the form of gray powdered pellets.

2.2. Chemical Analysis

Numerous chemical analyses were conducted to determine the components of the Foam. The elemental sulfur content in sulfur wastes was estimated. Additionally, the sulfate ion percentage was determined in a solution containing calcium sulfate, sodium sulfate, and potassium sulfate according to standard methods (18,19).

2.3. Foam as Asphalt Modifier

A weight of 100 grams of asphalt with the specified properties in Table (1) was placed in an asphalt processing device. Various proportions of foam were added with continuous mixing, and the temperature was gradually raised to the range of 170-180 °C. Mixing continued at this temperature for 60 minutes. Afterward, necessary measurements were conducted to determine the properties of the modified asphalt, including ductility (20), penetration (21), softening point (22), penetration index (23), Marshall test (24), stripping (25) and aging (26).

2.4. Preparing Calcium, Potassium, and Sodium Polysulfide Solutions from Foam

Given the high sulfur content in sulfur waste (Foam) according to the analyses conducted in section 2-2, these solutions can be prepared from it according to standard methods (27).

2.5. Preparing Nano-Sized Sulfur Powder

Nano sulfur powder was prepared in three ways as follows (27):

25 mL of sodium polysulfide solution was placed first, 25 mL of potassium polysulfide solution second, and 25 mL of potassium polysulfide solution third, Each addition alone in a beaker, and diluted hydrochloric acid was added with continuous stirring until the solution turned light yellow, The solution was then centrifuged at 4000 revolutions per minute for 30 minutes and washed thoroughly with distilled water to remove any sulfide ions. The resulting precipitate was dried and weighed. The models were then analyzed using (SEM), (EDX) and (XRD), and this precipitate represents nanoscale sulfur.

2.6. Nano-Sulfur as A Modifiers Materials for The Rheological Properties of Asphalt

The prepared nanosized sulfur was utilized in the asphalt modification process by taking 100 grams of asphalt. Various proportions of nanosized sulfur, obtained from the previous sections were added to the asphalt. The mixture was gradually heated with continuous stirring to a temperature range of 170-180 °C. It was then kept at this temperature for 60 minutes, Subsequently, the rheological properties were measured as outlined in section 2-3.

3. RESULTS AND DISCUSSION

The Mishraq mine is located 45 kilometers south of the city of Mosul on the western bank of the Tigris River. Sulfur is extracted from this field using the well-known Frasch method. Due to its geological nature, the sulfur contains approximately 1% of hydrocarbon materials, which are removed through chemical oxidation using concentrated sulfuric acid. This method produces solid sulfur compounds with a sulfur content of around 88%. To obtain asphalt with specifications suitable for road paving, resistance to various environmental conditions, and the ability to withstand heavy and repetitive loads, numerous researchers have explored the use of various additives to improve asphalt properties. Our study focused on utilizing locally available byproducts from the Sulfur Mashreq (foam) and its derived materials to modify asphalt and enhance its rheological characteristics. This was achieved through several steps.

3.1. Rheological Properties of Original Asphalt and The Paving Asphalt

The rheological properties of original asphalt and the paving asphalt were measured. Table 1 and 2 below shows the rheological properties for these two types of asphalt.

Table 1: Rheological properties of original asphalt.

Properties	Result
Ductility (cm)	+150
Penetration (100gm.5sec.25°C)	46.1
Softening point (°C)	50
Penetration index (PI)	-1.376

The rheological properties of the asphalt suitable for use in paving operations, in a way that suits the climate conditions of our country Iraq, were also determined according to the standard specifications shown in Table 2:

Table 2: Rheological Properties of paving Asphalt (23).

Properties	Range ASTM (23)
Ductility (cm)	≥100
Penetration (100gm.5sec.25°C)	40-50
Softening point (°C)	50-58
Penetration index (PI)	-2 _+2

3.2. Foam Components and Their Use in Improving Asphalt Properties

To know the components of the foam resulting from the purification of Mishraq sulfur in Iraq using the chemical oxidation method, we carried out chemical analyzes of this material and Table 3 shows the components of these residues (foam) according to the analyzes conducted.

Table 3: Components of the foam.

Parameters	Wt (%)
Free Sulfur	88.15
Bonded sulfur	**2.58**
Carbon	**1.86**
Acidity	1.53
Ash	**2.23**
Moisture	**3.65**

Also, energy dispersive X-ray spectroscopy (EDX) was used, which enables obtaining the composition or chemical analysis of the material as (EDX) technique provides the nature of the elements contained in the material and their percentages (28). Figure 1 shows the main elements in the foam.

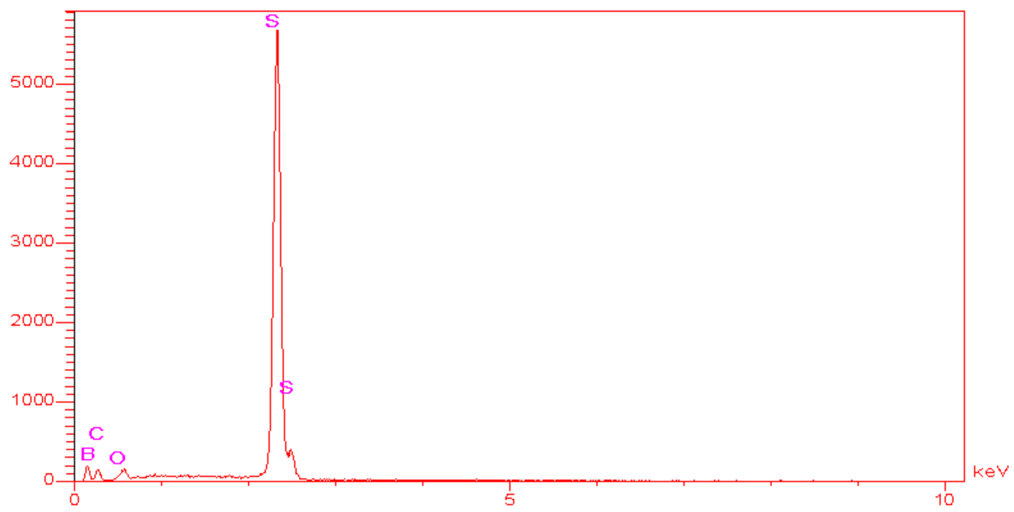

Figure 1: Energy dispersive X-ray spectroscopy (EDX) spectrum of the (foam).

Table 3 shows the results of the chemical analysis of the foam. Figure 1 represents the analysis using the energy dispersive X-ray spectrum (EDX) of the foam. Through the analyzes it is clearthat the foam material consists mainly of sulfur with minor amounts of other elements. On this basis, this material was used to modify asphalt at a temperature ranging between 170-180 °C for 60 minutes after determining the optimal conditions of temperature and time used in the modification process. Table 4 and Figure 2 illustrate the results obtained.

Table 4: The rheological properties of the asphalt modified by the foam.

Sample	Foam ratio %	Ductility (cm.25 °C)	Penetration (100gm.5sec.25°C)	Softening Point (°C)	Penetration index (PI)
AS1	0.5	+150	47	54	-0.391
AS2	1	+150	45.1	57	0.165+
AS3	1.5	91	41.2	61	+0.756
AS4	2	81	39.2	63	+1.016

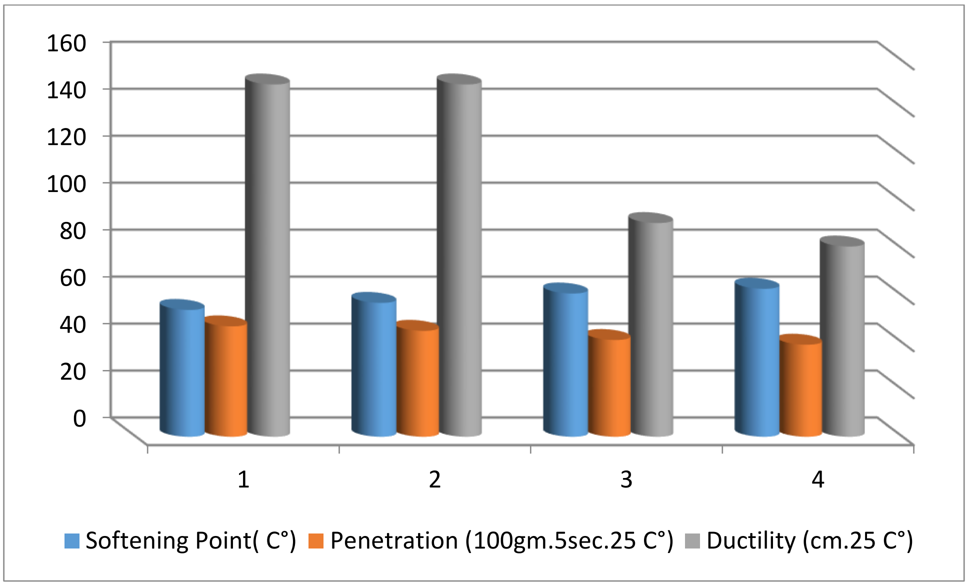

Figure 2: The rheological properties of asphalt modified by the foam.

We note from Table 4 and Figure 2 that the rheological properties represented by Ductility, penetration and softening point were all within the standard Iraqi specifications at additions of 0.5% and 0.1% of the added material. The table also shows that the best addition ratio is 1%. At 1.5% addition, Ductility and softening point were observed to be outside the required specifications. At 2% percentage, all these properties were observed to be outside the standard specifications. Therefore, foam material can be used to improve the specifications of asphalt at the identified ratios and under the conditions identified for reaction temperature and specific time.

3.3. Prepared the Nano-Sulfur

Nano sulfur was prepared from the foam using three methods as outlined in section (2-5) of the experimental part. The prepared nano sulfur was then characterized using scanning electron microscopy (SEM) to determine the nanoscale sizes. Energy dispersive X-ray spectroscopy (EDX) was also used to determine the sulfur content.

3.3.1. Nano-Sulfur prepared from calcium polysulfide

Nano-sulfur was prepared from calcium polysulfide (CaSx), as shown through (SEM) and (EDX) measurements.

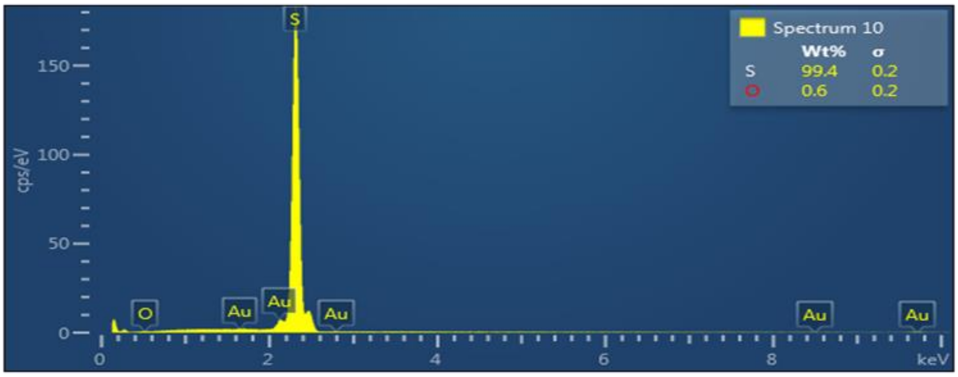

Figure 3: (EDX) spectrum for the nano-sulfur prepared from CaSx.

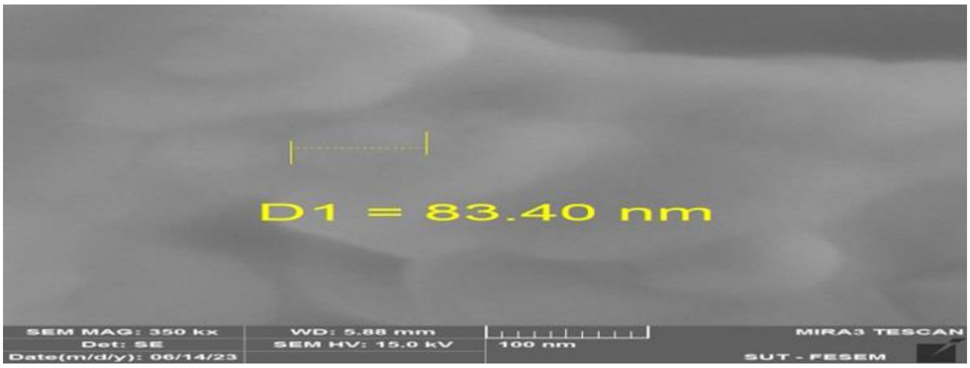

Figure 4: (SEM) of the nano-sulfur prepared from CaSx.

We note from Figure 3, which represents the energy dispersive X-ray spectroscopy spectrum, and Figure 4, which represents the scanning electron microscope examination, that the nano sulfur prepared from potassium polysulfide was 99.4%, which is a high percentage indicating the purity of the prepared sulfur. The remaining 0.6% of oxygen represents moisture since hydrogen is not shown in this measurement (29). We also note from the (SEM) measurement that the nanoscale size of sulfur was 83.40nm. To utilize the prepared nano sulfur in one of the industrial fields, it was used to modify asphalt under the same optimal conditions used to modify asphalt using the main material foam at a temperature of 170-180 °C for 60 minutes. Table 5 and Figure 5 illustrate the results obtained.

Table 5: The rheological properties of asphalt modified by nano-sulfur (83.40nm).

Sample	S-NP%	Ductility (cm.25 °C)	Penetration (100gm.5sec.25°C)	Softening Point (°C)	Tenetration index (PI)
AS5	0.5	+150	46.4	54	-0.421
AS6	1	+150	45.6	55	-0.238
AS7	1.5	+150	44.3	57	+0.190
AS8	2	135	39.3	61	+0.647

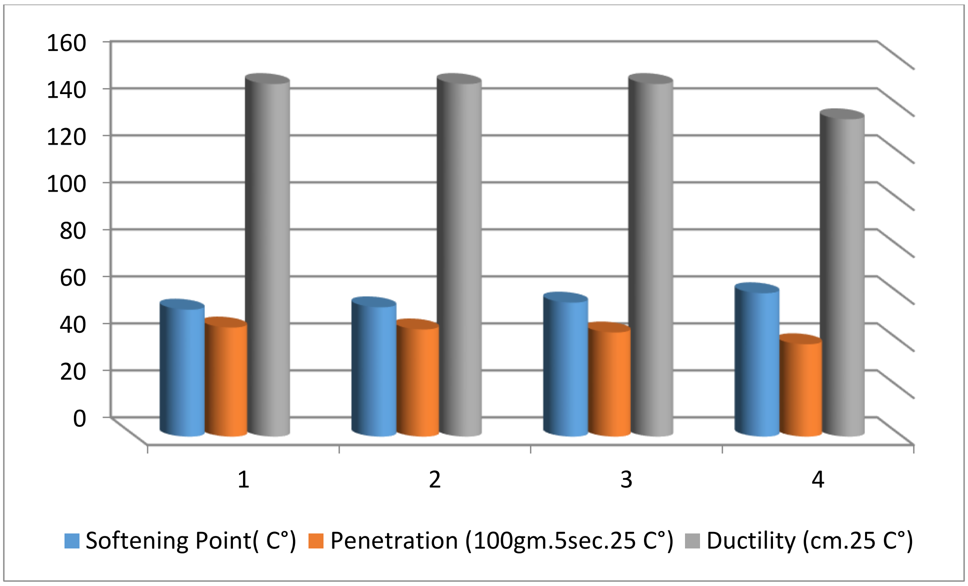

Figure 5: The rheological properties of asphalt modified by nano-sulfur (83.40nm).

From Table 5 and Figure 5, it can be observed that the ratios were satisfactory, except at the percentages of 2%, where the softening point was outside the desired specifications

3.3.2. Nano-Sulfur prepared from potassium polysulfide

Nano-sulfur was prepared from potassium polysulfide (K2SX), as indicated by (SEM) and (EDX) measurements shown in the following figures.

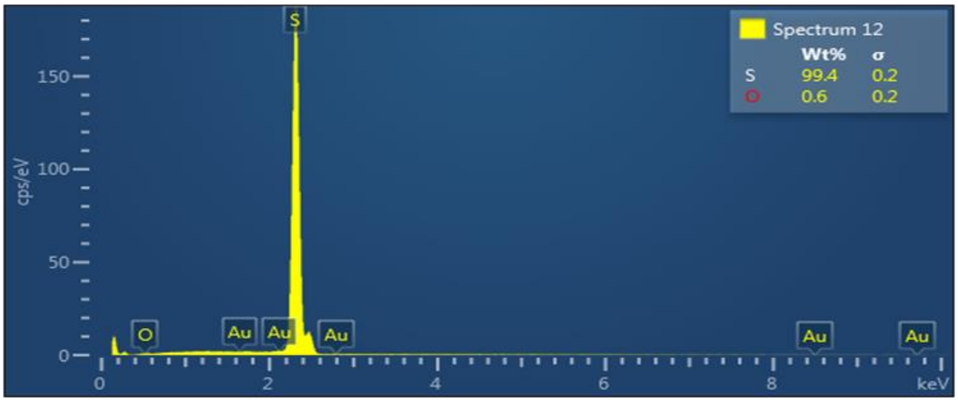

Figure 6: % (EDX) spectrum for the nano-sulfur prepared from K2SX.

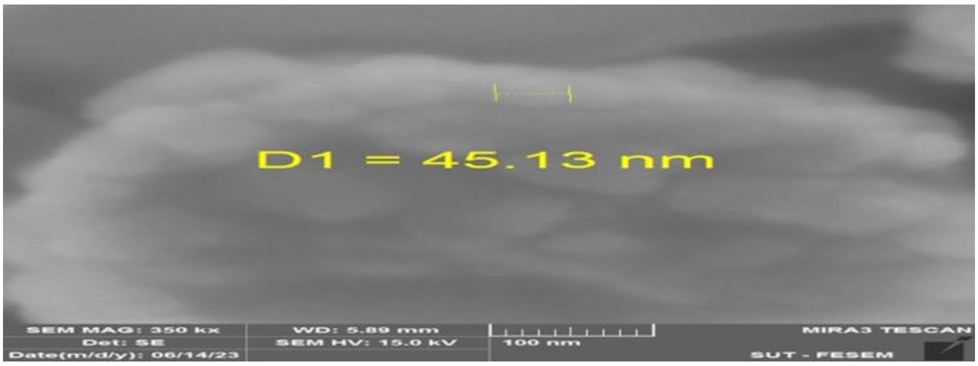

Figure 7: (SEM) of the nano-sulfur prepared from K2SX.

From these measurements, it can be observed that the sulfur content is 99.4% and the size of the nano-sulfur is 45.13nm. Table 6 and Figure 8 illustrate the results of using this type of sulfur in asphalt modification.

Table 6: The rheological properties of asphalt modified with nano-sulfur (45.13nm).

Sample	S-NP%	Ductility (cm.25 °C)	Penetration (100gm.5sec.25 °C)	Softening Point (°C)	Penetration index (PI)
AS9	0.5	+150	47.1	53	-0.614
AS10	1	+150	45.8	55	-0.228
AS11	1.5	+150	43.4	57	+0.076
AS12	2	141	40.1	60	+0.501

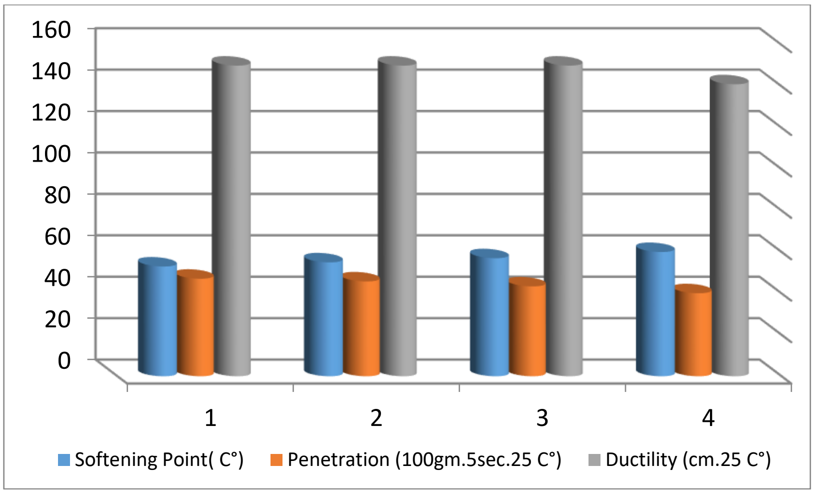

Figure 8: The rheological properties of asphalt modified by nano-sulfur (45.13nm).

3.3.3. Nano-Sulfur prepared from sodium polysulfide

Nano-sulfur was prepared from sodium polysulfide (NaSx), as indicated by SEM and EDX measurements shown in the following figures.

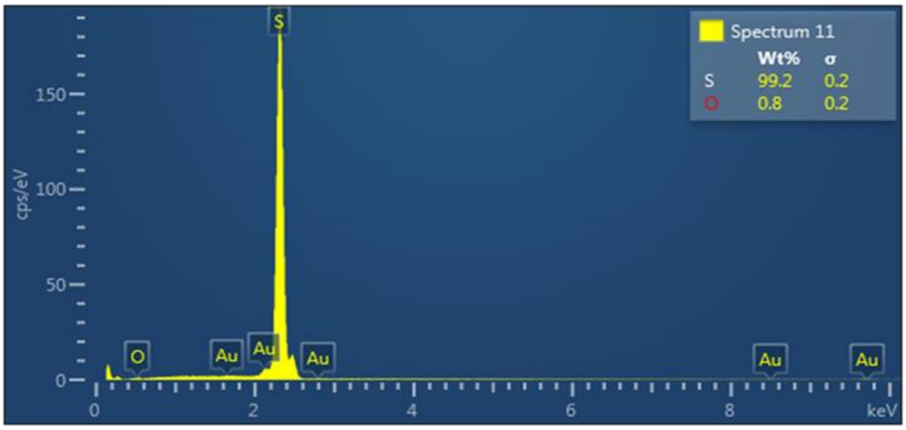

Figure 9: (EDX) spectrum for the nano-sulfur prepared from Na2Sx.

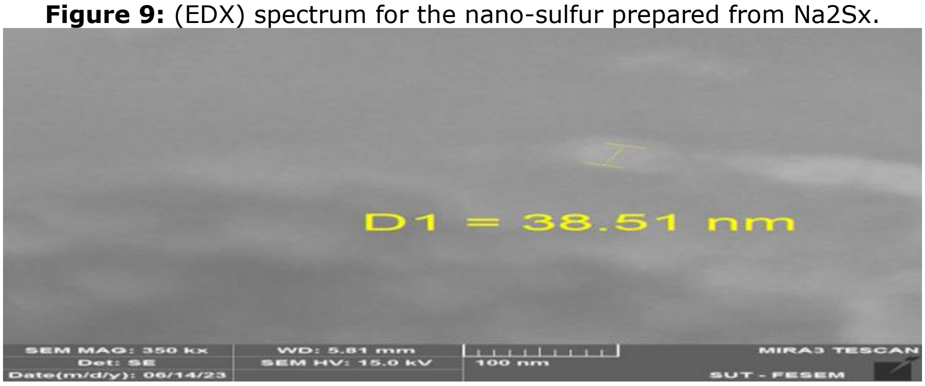

Figure 10: (SEM) examination of the nano-sulfur prepared from Na2Sx.

Through these figures, it can be observed that the nano-sulfur prepared from sodium polysulfide has a sulfur content ratio of 99.2%, indicating high purity of the prepared sulfur. Additionally, the nano-sulfur has a size of 38.51nm for the purpose of comparing the effects of different nano-sized particles, this prepared nano-sulfur was used in asphalt modification. Table 7 and Figure 11 illustrate the results obtained.

Table 7: The rheological properties of asphalt modified with nano-sulfur (38.51nm).

Sample	S-NP%	Ductility (cm.25 °C)	Penetration (100gm.5sec.25 °C)	Softening Point (°C)	Penetration index (PI)
AS13	0.5	+150	47.3	53	-0.604
AS14	1	+150	46.6	54	-0.411
AS15	1.5	+150	45.3	56	-0.036
AS16	2	145	42.6	59	+0.442

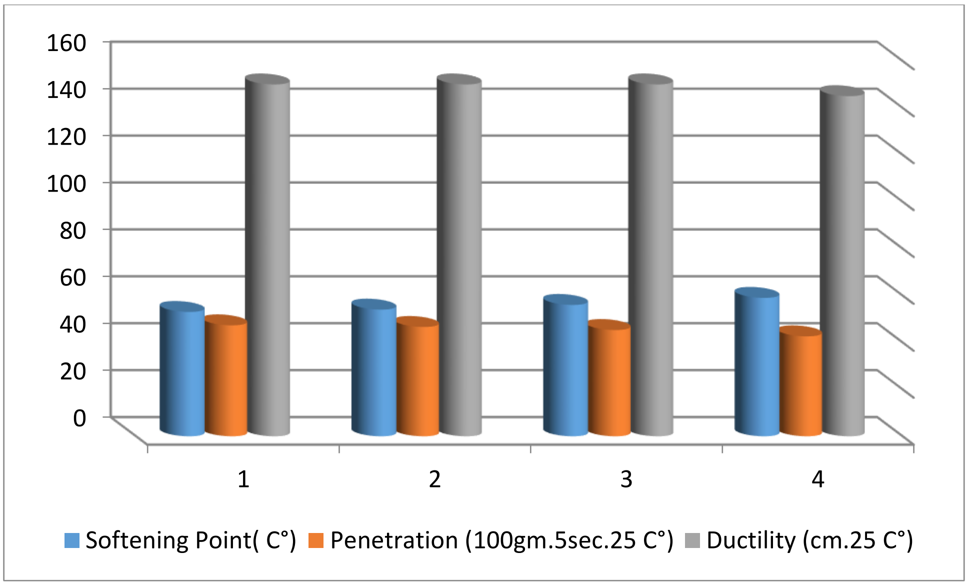

Figure 11: The rheological properties of asphalt modified by nano-sulfur (38.51nm).

We note from the results obtained from asphalt modification using different nanoscale sizes that good models were obtained that can be used in asphalt paving according to the rheological properties measured. We also note that nano sulfur of 38.51nm gave the best results, as it was observed from the results that all ratios of the additive (0.5, 1, 1.5, 2)% were within the standard specifications for asphalt used in paving. Next was nano sulfur of 45.13nm, although at a 2% additive ratio, the softening point and penetration were at the at the minimum properties As for nano sulfur of 83.40nm, the results showed that at a 2% additive ratio, the softening point and penetration were outside the standard specifications, along with a decrease in Ductility. By observing the results, it can be said that the smaller the nanoscale size of sulfur, the better results it gives for modifying asphalt properties. The ratio of the additive with the smaller size can also be increased as a result of greater dispersion and interaction of nanoscale particles with the asphalt.

To confirm the success of these asphalt models in paving roads and withstanding repeated traffic loads as well as resisting environmental conditions, acid rain and time degradation, four well-modified model samples represented by AS14, AS2, AS6, and AS11 were selected and subjected to Marshall testing, chemical immersion testing, and aging testing in addition to measuring the properties of the original asphalt to determine the suitability of these models for road paving works.

The Marshall Stability and Flow values are determined from this test and the Marshall Quotient is calculated by dividing the Marshall Stability by the Flow value (30). Table 8 illustrates the stability and flow values for the tested samples compared to the original model and according to the specifications of the Iraqi Public Works Department (S.C.R.B) inimum limit of the standard specifications for asphalt used in paving.

Table 8: The stability and flow values for the original asphalt, modified asphalt, and the specifications of the Iraqi Roads and Bridges Authority (S.C.R.B).

Sample No	Asphalt (%)	Marshal Test		
		Stability (KN)	**Flow (mm)**	**MQ**
AS0	4.5	10.7	5.2	2.05
AS2		14.3	2.9	4.93
AS6		15.2	3.0	5.06
AS10		16.2	3.1	5.22
AS15		16.6	3.0	5.53
AS**		7 Minm.	2-4	3.5 Minm.

AS* the specifications of the Iraqi Roads and Bridges Authority

It is evident from Table 8 above that all the modified models are better than the original model if used as paving asphalt. The MQ value of the modified models can also be observed to be higher than the MQ value of the original asphalt. We also note that asphalt modified with nano sulfur of 38.51 nm has the best stability value. Therefore, the modified models would be more resistant to permanent deformation and repeated loads than the original asphalt.

To determine the resistance of the asphalt after mixing with aggregates to acid rain and high temperatures, the same samples that underwent Marshall testing were subjected to chemical immersion (stripping) testing and compared to the basic model. Table 9 illustrates the stripping results of the original asphalt (AS0) and the modified models from aggregates after chemical immersion.

Table 9: Chemical Immersion.

No. of modified sample	Percentage of Na_2Co_{3gm}	R&WNO	R&WNO for the original asphalt	R&WNO for the modified samples
	0.025	1		
	0.041	2		
AS0	0.082	3	3	
	0.164	4		
AS2,,AS6	0.328	5		5
AS10	0.656	6		6
AS15	1.312	7		7
	2.624	8		

We note from Table 9 and the stripping values (the amount of sodium carbonate at which asphalt began to strip or separate from aggregates), that the modified models began stripping at a higher amount of sodium carbonate compared to the original asphalt. Number (1) refers to the amount of sodium carbonate which is 0.025 grams in 50 mL of distilled water, and number (8) refers to the highest amount of sodium carbonate which is 2.624 grams. Possessing higher stripping values enables the modified asphalt to better adhere to aggregates compared to the original asphalt, providing greater resistance to acid rain and high temperatures.

To determine the resistance of the prepared asphalt models to environmental conditions and aging over time, aging testing was conducted on both the original model (AS0) and the well-selected modified models chosen for testing. The testing was conducted according to standard specifications as shown in Table 10.

We note from Table 10, which represents aging test results, that the effect of aging conditions such as heat and oxidation on the rheological properties of the modified asphalt models was less than the effect on the original asphalt. This indicates that the modified asphalt models are characterized by great resistance to environmental conditions, aging over time and stress processes. This is a positive aspect of modification. The modified asphalt models showed less change and degradation of rheological properties compared to the original asphalt when subjected to aging testing, indicating higher resistance to aging factors such as heat and oxidatio.

Table 10: Aging testing.

Sample no.	Rheological properties	Before test	After test
AS0	Ductility cm. 25 °C	+150	+150
	Penetration (100gm.5sec.25°C)	46.1	42.2
	Softening point (°C)	50	53
AS2	Ductility cm. 25°C	+150	+150
	Penetration (100gm.5sec.25°C)	45	44.6
	Softening point (°C)	57	58
AS6	Ductility cm. 25 °C	+150	+150
	Penetration (100gm.5sec.25°C)	45.6	45.1
	Softening point (°C)	55	53
AS10	Ductility cm. 25 °C	+150	+150
	Penetration (100gm.5sec.25°C)	45.8	45.4
	Softening point (°C)	55	54
AS15	Ductility cm. 25°C	+150	+150
	Penetration (100gm.5sec.25°C)	45.3	45.1
	Softening point (°C)	56	55

4. CONCLUSION

1- The solid sulfur materials (Foam) resulting from the chemical purification method of sulfur in the General Company for Sulfur in Al-Mishraq contain a high percentage of elemental sulfur that can be utilized in various fields
2- The possibility of preparing calcium sulfide, potassium polysulfide, and calcium polysulfide solutions from sulfur waste materials as a raw material.
3- Preparation of nano-sized sulfur powder with varying nano-scale dimensions.
4- Using sulfur wastes materials (Foam) and the nano sulfur prepared from these wastes materials in modifying asphalt and improving its rheological properties.
5- Improvement of the rheological properties of asphalt is more pronounced as the nano-sulfur particle size decreases.
6- Obtaining good asphalt samples that can be used in road paving due to their ability to resist various environmental conditions, including repeated loads, acid rain, and aging, as evidenced by Marshall testing, chemical immersion, and aging tests.

5. CONFLICT OF INTEREST

There are no conflicts of interest.

6. ACKNOWLEDGMENTS

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