

Research Article

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Performance and rheological characteristics of hot mix asphalt modified with melamine nanopowder polymer

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Abstract: The increasing price of asphalt modifiers and the compulsory need for them make it necessary to find an alternative to improve the rheological properties of asphalt binders using economical polymeric materials. The aim of this study is to improve the rheological properties of asphalt binders and mixtures using nanopowder for polymer formaldehyde melamine (“NPFM”). The binder asphalt result showed that adding 4% NPFM to reference asphalt enhances the performance grade (PG) from PG (64-16) to PG (82-10). Additionally, the difference between the softening points of the upper and lower parts showed superior storage stability of less than 2.2%. As for Marshall properties, the results of the asphalt mixture modified with “NPFM” showed higher resilience, with a stability ratio increment of (17%) compared to the original mix. As for the tensile strength ratio (TSR), TSR effects showed a high resistance level (85.44%), higher than that of the styrene-butane-styrene mixture. Adding “NPFM” to the reference asphalt gave a high resistance against permanent deformations. Rutting resistance increased by 67.6% at a temperature of 45°C, 80.6% at 55°C, and 60.5% at 70°C compared to other asphalt mixtures.

Keywords: modified asphalt, formaldehyde melamine, rheological properties, sustainability, nanopolymer polymer formaldehyde melamine nPFM

1 Introduction

Superpaving and the revolution of modern roads require new elements to improve the properties of asphalt mixtures, especially when the asphalt binder is the essential feature of the internal structure of the asphalt mixture in road networks [1]. Therefore, an increase in $(G^*/\sin \delta)$ is required, representing the internal structure's fundamental component as a function of binder bonding to resist permanent deformations [2,3]. Adding polymer to the asphalt binder enhances performance, manufactures modified asphalt with high shear modulus, and improves the adhesion between the asphalt and the rest of the asphalt mixture components [4,5]. It is essential to account for the stability of the polymer during its mixing with asphalt due to the oxidation and separation factors [6] and achieve sustainability based on meeting consumer requirements and keeping pace during technology development at the lowest cost [7,8]. Therefore, the new economic polymers compatible with asphalt were interesting reasons for developing the rheological properties of asphalt and thus improving the mixtures against different types of failures [9]. Due to the fine particle properties of polymeric materials, one essential consideration that enhances the workability and structural bonding is the modification of asphalt binder with nanomaterial [10]. The application of counting nanomaterials to asphalt is the most crucial discovery of researchers. It led to improvements that significantly improved the performance of asphalt and positively affected Marshall properties, with increased adhesion ability between the binder and the aggregate [11]. The percentage of polymers added to modify the asphalt binder ranges from 4 to 8% higher than the optimal asphalt content in the asphalt mixture, provided that it does not exceed this percentage, causing difficulty in its work [1,12]. There is a constant search for polymers similar to the styrene-butane-styrene (“SBS”) polymer, an exciting polymer used to improve the rheological properties of asphalt. Still, it is expensive and complicated due to continuous agitation during mixing [13,14]. Melamine formaldehyde polymer (“MFP”)

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was selected to add to the asphalt binder, which is considered one of the formaldehyde polymer applications in asphalt mixtures [15]. In addition, civil engineering used melamine formaldehyde polymer waste, achieving results consistent with the specifications [16].

2 Materials and laboratory testing

The results of the performance grade for the asphalt binder used in this study are shown in Table 1. PG (64-16) met the required specification in Iraq PG (82-10), and the performance grade should be increased. Therefore, the thermal stable polymer formaldehyde melamine (“PFM”) was used as a modifier. To ensure the compatibility and stability of mixing the modifier with asphalt, PFM was converted to nanomaterial with particles smaller than “100 μm ” to increase the interaction by increasing the surface area and the chemical activity while raising its interacting surface through unrestricted electrons. Meanwhile, the atoms inside the material are more bound [17]. The granular size of the nanopolymer formaldehyde melamine (“NPFM”) is represented in Figures 1 and 2. All “NPFM” characteristics are tabulated in Table 2.

2.1 Conversion of PFM to NPFM

To prepare the melamine formaldehyde from the damaged dishes, the following steps were taken:

1. The damaged dishes were broken into small pieces and parts so that the particles could pass through sieve No. 4 (4.75 mm) and ground into powder by “Herson Mill,” as shown in Figure 3.
2. After crushing, the melamine formaldehyde powder was sifted through sieve No. 325 (45 μm) by washing according to [18].
3. After that, the particles were exposed to heat to dry, where they were inserted into an oven at 100°C until the sample was dehydrated, finally, the dried particles were crushed until we got Nanopowder using “Ball Mill,” as shown in Figure 4 to obtain Nano “PFM” powder shown through Figure 5.

2.2 Method of preparing modified asphalt

The compatibility of asphalt with polymers and the properties of the polymer are essential in producing modified asphalt with good rheological properties due to the chemical composition of each polymer and asphalt. The chemical composition differs from one polymer to another. The chemical composition of asphalt varies from one country to another according to the type of oil extracted from it. The mixing temperatures and pressure were determined according to the ASTM D 4402 Standard Test Method [19], which is used for measuring rotational viscosity. Two different percentages of melamine (2 and 4%) were used.

Table 1: Rheological characteristics of base asphalt binder

Performance grade of base asphalt binder				
Parameter		Temperature measured	Parameter measured	Requirements
Aging	Standard specification	Original binder		
Rotational viscosity (Pa s), Dynamic shear rheometer (DSR), $G^*/\sin \delta$, kPa	ASTM D 4402	@135°C		3 Pa s, Max
	ASTM D 7175		0.542	1 kPa, Min
		@64°C	1.36	
		@70°C	0.96	
Flash point (°C)	ASTM D 92	...		
Penetration (0.1 mm)	ASTM D 05	@25°C	291	230 min
Ductility (5 cm/min)	ASTM D 113	@25°C	42	...
			134	Greater than 100 cm
Aging		Rolling thin film oven (RTFO) residue		
DSR	ASTM D 7175	@64°C	2.55	2.2 kPa, Min
$G^*/\sin \delta$, kPa	ASTM D 2872	@70°C	1.85	
Mass loss (%)	ASTM D 2872	0.53	...	1, max
Aging		Pressure aging vessel (PAV-110_C) residue		
DSR	ASTM D 7175	@25°C	6,125	5,000 kPa, Max
$G^*/\sin \delta$, kPa	ASTM D 6521	@28°C	4,425	

Effective Diameter: 94.2 μm
 Polydispersity: 0.107
 Baseline Index: 9.5

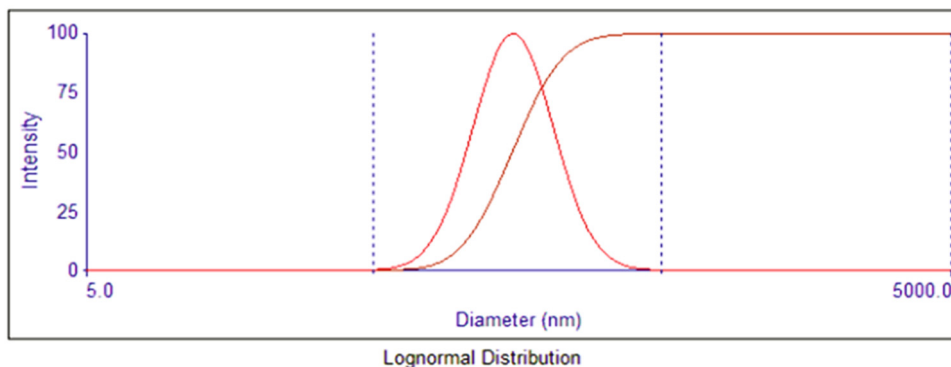


Figure 1: Effective diameter of NPfM.

Mixing was carried out at automatic temperatures ranging between 160 and 170°C for 2 h. Figure 9 represents the mixing of melamine with asphalt. The interaction of melamine polymer with the asphalt is one of the strong interactions, complementing the many properties of melamine. It works on a strong ionic crosslinking between it and the asphalt. Nitrogen and hydrogen from melamine with (O=C–OH) form carboxylic acid groups within the asphalt element, as shown in Figure 6, forming epic coordinating bonds from six sides, and then these ramify in turn so that each component has a pair of bonds or electrons union to make a strong crosslinking between melamine and asphalt. Hence, it becomes an influential group that causes high stability. As it is known through the partial composition of asphalt, it is full of bonds, Figure 7, and can unite with “NPfM” quickly because there are hundreds of sites to link them from all sides. As a result of the strong interlocking

between the asphalt and “NPfM,” it gains preference compared with the component “SBS,” which has a partial hexagonal structure, with one double bond in its composition ($-\text{CH}=\text{CH}-$), Figure 8. The ion bonding between SBS and asphalt occurs at a single site. Unlike NPfM, melamine cross-links with asphalt, bonding and union occurring at hundreds of sites. Unlike “NPfM,” the melamine crosslinks with asphalt, and hundreds of sites appear for the link. The “SBS” becomes ineffective when left in the atmosphere because its saturation in moisture will damage it, and the double bond turns into ($-\text{CH}_2-\text{CH}-\text{OH}-$), which is useless.

2.3 Mixing device

Mixing asphalt with other additives requires heat, high speed, shear mixing, and control of asphalt oxidation.

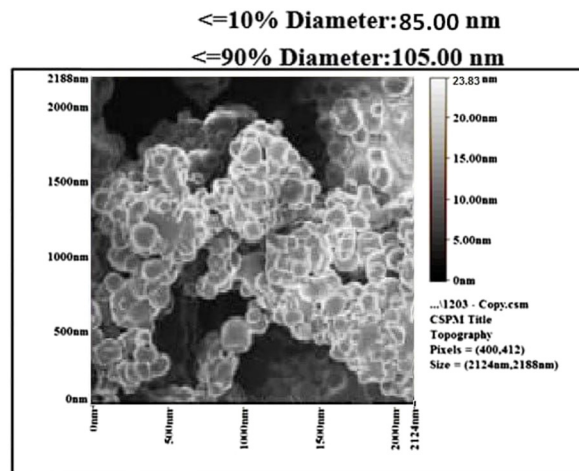
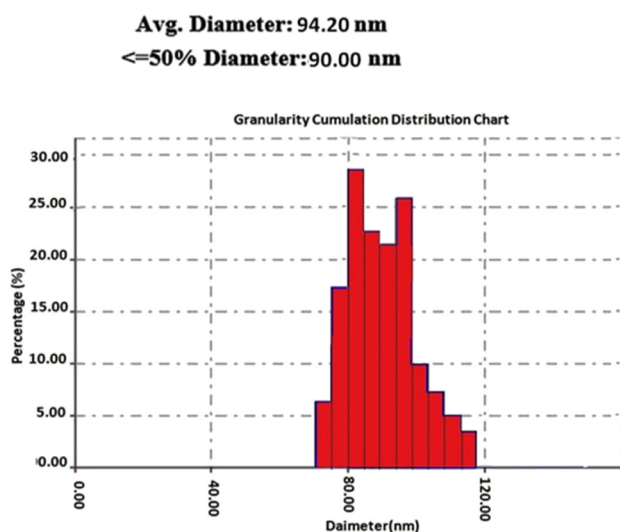


Figure 2: Particles diameter of melamine.

Table 2: Characteristics of NPFM

Measurement	Standard specification	Unit	Limitation	Result
Density	D792	...	1.5–1.55	1.573
Specific gravity	D792	...	1.5–1.55	1.5
Color	D792	...		white
Tensile strength	D638	MPa	55–83	58
Elongation at break	D638	%	0.3–0.9	0.6
Melting temperature	NTP1992 ICSC1154	°C	>300	345
Dielectric strength	D149	V/0.001in	160–240	200
Boiling point	NTP1992 ICSC1154	...	...	Sub limes
Water absorption	D570			
24 h at 24°C		Mg	10–50	20
30 h at 100°C		Mg	40–110	55

**Figure 3:** Herson Mill.

These requirements become efficient if the additive has nanoparticles. There are special devices for mixing polymeric materials with asphalt. They are added as units with the asphalt concrete production process. A local device simulating the shear mixer was manufactured for laboratory evaluation. The requirements for mixing depend on the relationship between temperature and viscosity. The mixing temperature was found through an asphalt viscosity test [20]. The mixture was mixed with two ratios of “NPFM” 2 and 4%, and the powder was added to the asphalt gradually at a temperature of 160–170°C with a speed of 2,220 rpm for 2 h until a good spread of the

**Figure 4:** Ball Mill.**Figure 5:** NPFM.

additive was obtained in the binder asphalt. Figure 9 shows the mixing device and mixing process. Morphology of the nanomaterial, homogeneous distribution, and non-agglomeration of the polymer particles in the asphalt link structure was achieved by scanning electron microscopy “SEM,” as shown in Figure 10.

3 Results and discussion

3.1 Asphalt binder tests

The results of the tests for adding melamine to the asphalt showed promising results, as shown in Table 3, when

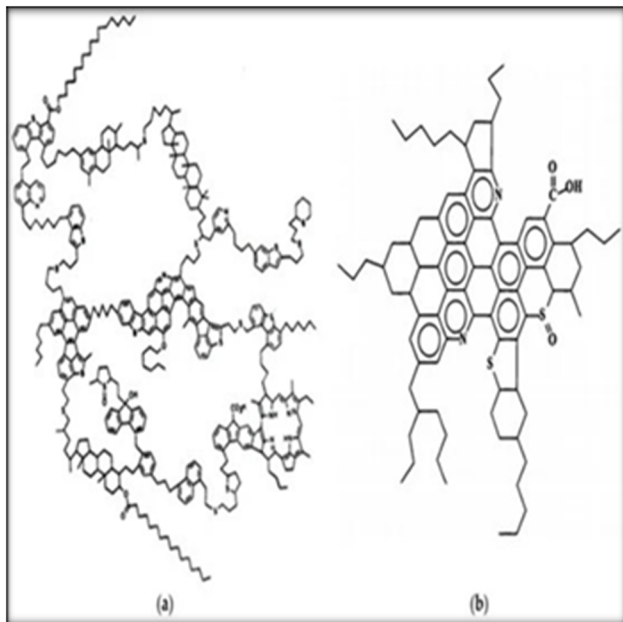


Figure 6: Structure of base asphalt binder. (a) Structural of asphalt binder and (b) fine section in asphalt binder.

compared with the SBS-modified asphalt. Table 4 describes the characteristics of SBS-modified asphalt. Storage stability results are tabulated in Table 5.

Table 5 shows the equilibrium thermodynamics and phase separation dynamics of polymer-modified asphalt based on different softening points according to standard practice [21]. Figure 11 shows the sample preparation process to examine the softening points of the upper and lower parts of the sample. The results showed that the asphalt polymer modifier's storage stability and separation behavior strongly depend on the composition and internal structure of the asphalt and the polymer. Modified asphalt "NPFM" has also been shown to maintain its rheological

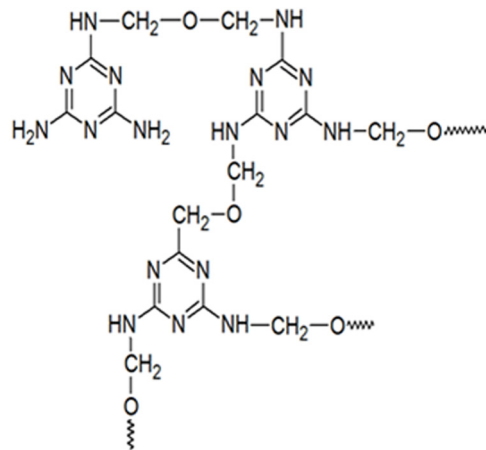


Figure 7: Shows sites to link in melamine.

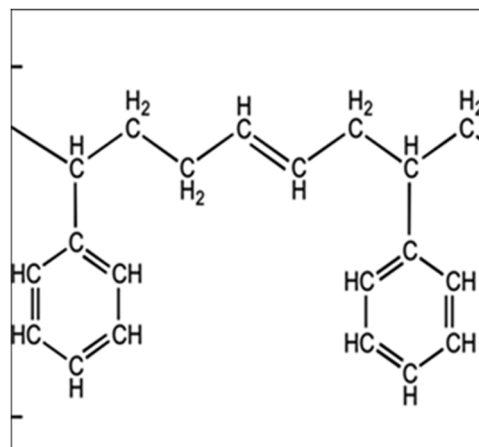


Figure 8: Shows sites to link in groups in asphalt SBS.

properties due to the strong compatibility between the polymer and the asphalt and the bonding network between them. It was an essential factor for non-separation and a

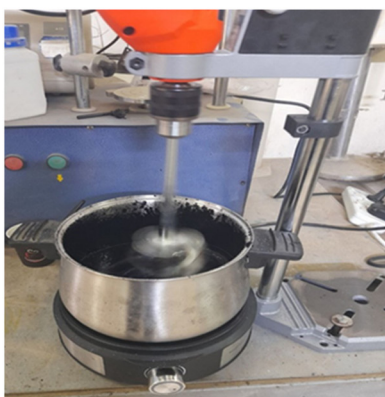


Figure 9: Shows the mixing device during the mixing process.



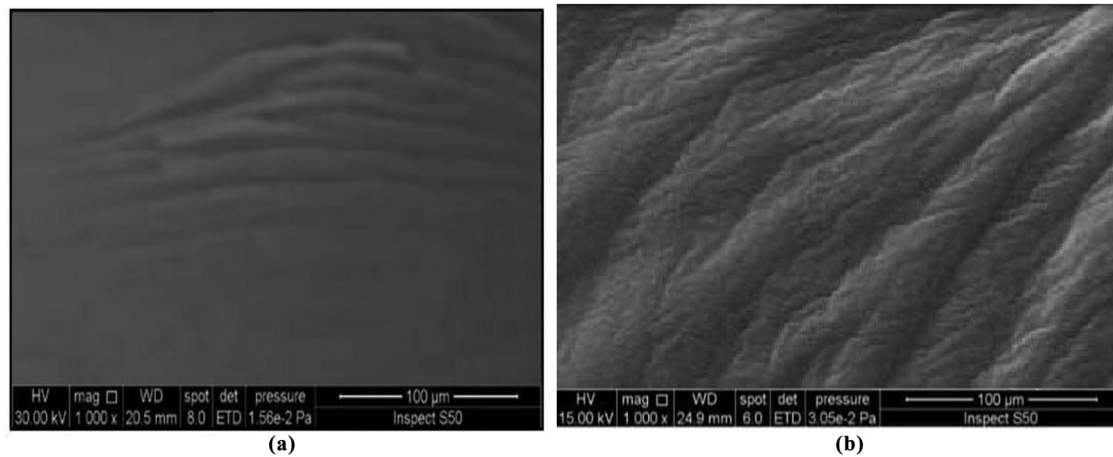


Figure 10: SEM image of (a) reference asphalt and (b) asphalt modified with nPFM.

reason for high storage stability. The results also showed that “NPFM” is better than “SBS” in terms of storage stability, significantly when the ratio of the additive to the asphalt is increased. It also showed the high effectiveness of “NPFM” and a better cohesion reaction. The added nanostructure stopped the sinking process. In addition, there was enough surface area to make the “NPFM” particles move randomly through the internal structure of the asphalt instead of up and down. Unlike asphalt, a bonding element enhanced storage stability. Modified asphalt “SBS” is considered unstable for extended storage periods due to the deterioration of “SBS” at high temperatures, especially when high percentages of it are added [22]. Therefore, other additives are added to the asphalt to reduce stability

and separation problems [23]. Due to the thermally stable properties of “NPFM,” it is not affected by the time it remains in the oven, but rather, the cohesive bonds between it and the asphalt are strengthened. The results also demonstrated increased bonding within 5 days, in contrast to SBS-modified asphalt.

3.2 Asphalt mixture tests

As per ASTM D6927 Standard Test Method [24], Marshall samples were made using the asphalt mixture consisting of limestone dust as a filler and three types of asphalt: reference asphalt, SBS-modified asphalt, and NPFM-modified

Table 3: Physical properties of asphalt modified with 4% PFM

Type of asphalt	Standard specification	Performance grade of modified asphalt		
Parameter		Temperature measured	Parameter measured	Requirements
Aging			Original binder	
Rotational viscosity (Pa s)	ASTM D 4402	@135°C	1.3	3 Pa s, Max
DSR	ASTM D 7175	@76°C	2.31	1 kPa, Min
$G^*/\sin \delta$, kPa				
Flash point (°C)	ASTM D 92	—	270	230°C, Min
Aging			RTFO residue	
DSR	ASTM D 7175	@76°C	5.72	2.2 kPa, Min
$G^*/\sin \delta$, kPa	ASTM D 2872			
Mass loss (%)	ASTM D 2872		0.68	1, Max
Aging			PAV-110 C residue	
DSR	ASTM D 7175	@37°C	4,330	5,000 kPa, Max
$G^*/\sin \delta$, kPa	ASTM D 6521			
Bending beam rheometer (BBR)	ASTM D 05	@0°C	135	300 MPa, Max
Creep stiffness, MPa				
BBR	ASTM D 6648	@0°C	0.37	0.3, Min
Slop m -value				

Table 4: Physical properties of asphalt modified with 4% SBS

Type of asphalt	Standard specification	Performance grade of modified asphalt		
Parameter		Temperature measured	Parameter measured	Requirements
Aging			Original binder	
Rotational viscosity (Pa s)	ASTM D 4402	@135°C	1.2	3 Pa s, Max
DSR	ASTM D 7175	@76°C	1.21	1 kPa, Min
$G^*/\sin \delta$, kPa				
Flash Point (°C)	ASTM D 92	—	270	230°C, Min
Aging			RTFO residue	
DSR	ASTM D 7175	@76°C	2.31	2.2 kPa, Min
$G^*/\sin \delta$, kPa	ASTM D 2872			
Mass Loss (%)	ASTM D 2872		0.66	1, Max
Aging			PAV-110 C residue	
DSR	ASTM D 7175	@37°C	3,950	5,000 kPa, Max
$G^*/\sin \delta$, kPa	ASTM D 6521			
BBR	ASTM D 05	@0°C	105	300 MPa, Max
Creep stiffness, MPa				
BBR	ASTM D 6648	@0°C	0.42	0.3, Min
Slop m -value				

Table 5: Result of storage stability

Asphalt type	Additive ratio (%)	After storage stability test @3 days softening point (°C) ASTM D7173-14		
		Top	Bottom	Difference in value
Asphalt (SBS)	2	61.0	60.3	0.7
Asphalt (SBS)	4	72.9	72.0	0.9
Asphalt (nPfM)	2	65.7	65.2	0.5
Asphalt (nPfM)	4	77.3	77.0	0.3
Asphalt type	Additive ratio	After storage stability test @5 days softening point (°C) ASTM D7173-14		
Asphalt (SBS)	2	58.4	56.7	1.7
Asphalt (SBS)	4	62.6	60.5	2.1
Asphalt (nPfM)	2	67.5	67.2	0.3
Asphalt (nPfM)	4	79.3	79.2	0.1

**Figure 11:** Demonstrates specimens for the softening point test.



Figure 12: Marshall's device with test samples.

asphalt. Marshall stability test was carried out using the device after removing the samples from the molds and putting them in the water bath for half an hour, as required by the specification of testing the Marshall properties, as shown in Figure 12. Figure 13 shows the gravel gradation of the used mix.

The stability results for reference and modified asphalt “SBS” and “NPFM” showed higher strength of the asphalt mixtures containing melamine-modified asphalt than other asphalt mixtures. The increase in the result is due to the strong bonding between the asphalt “NPFM” and the elements of the asphalt mixture, which forms a vital bonding component. It

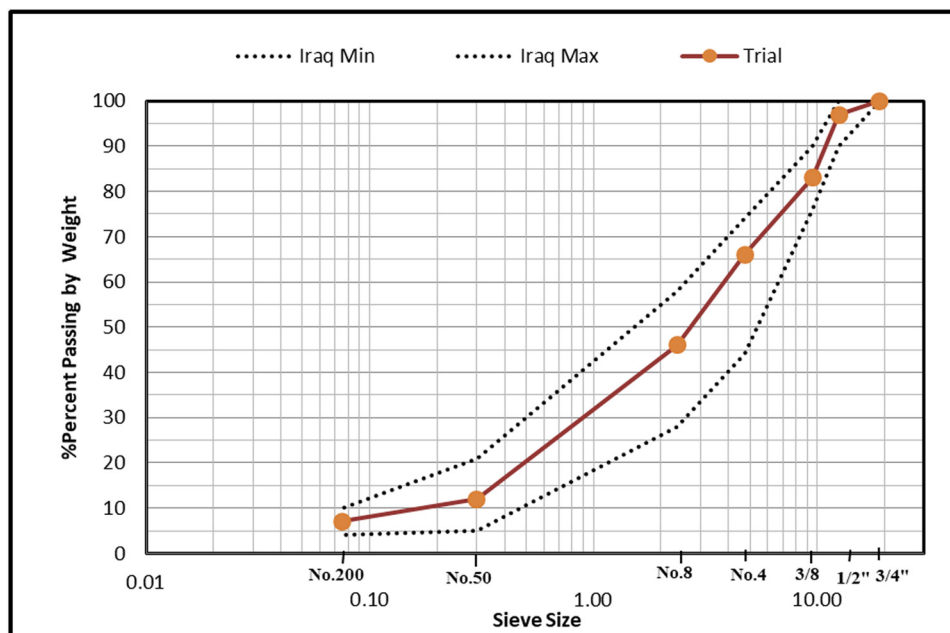


Figure 13: Grain size diameter (mm) of (SCRB, 2003) for surface course layer.

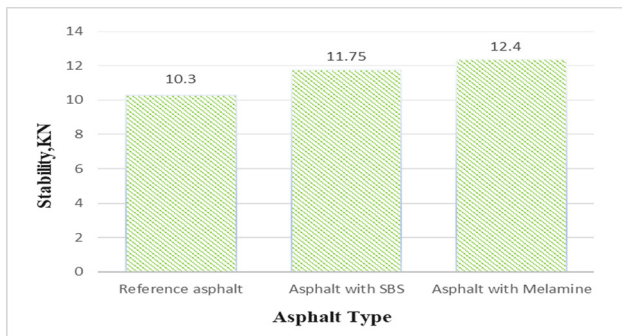


Figure 14: Marshall test results.

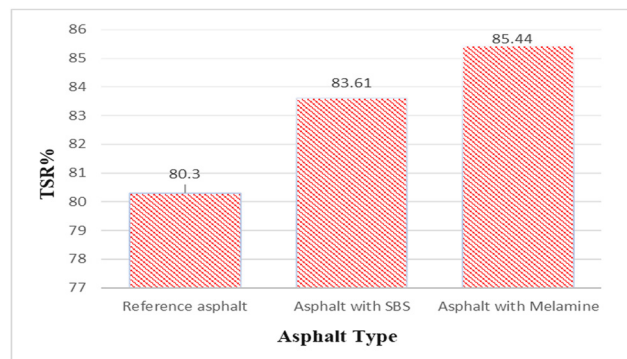


Figure 15: TSR% test results.

resembles a network with increasing viscosity, a crosslinking factor component forming a homogeneous mass. The “NPFM” element is considered one of the most substantial elements, as it complements a strong chain when added to the asphalt. Figure 14 shows the stability result.

A moisture damage test was carried out based on the specification [25], and the result showed that the tensile strength ratio (TSR) of NPFM-containing asphalt mixtures was higher than that of the asphalt mixture containing SBS-modified asphalt as well as the asphalt mixture that includes raw asphalt. Figure 15 shows the results of TSR with different types of asphalt. Moisture damage resistance when “NPFM” was used increased more than the TSR values of SBS-modified asphalt binders, all within a high improvement rate of more than 10%. This is attributed to “NPFM,” which improves the properties of asphalt due to the strong interlocking between the asphalt and “NPFM.” It is almost an insulating layer surrounding the components

of the asphalt mixture and prevents moisture penetration into the mixture and, thus, weakening it.

Compared to the original mix, the stability and TSR ratio of asphalt mixtures containing melamine-enhanced asphalt increase. The asphalt mixture containing SBS is attributed to the strong bonding between the melamine and asphalt particles from hundreds of places. This produces strong cohesion with high asphalt properties that effectively link aggregate particles as a solid homogeneous asphalt component at different conditions.

According to the rutting resistance test of asphalt mixtures containing modified asphalt “NPFM,” according to [26], was higher than that of the asphalt mixture containing SBS-modified asphalt and the asphalt mixture containing raw asphalt. The use of “NPFM” is more resistant than the rutting resistance for asphalt concrete with modified asphalt binder SBS, with a high rate of improvement.

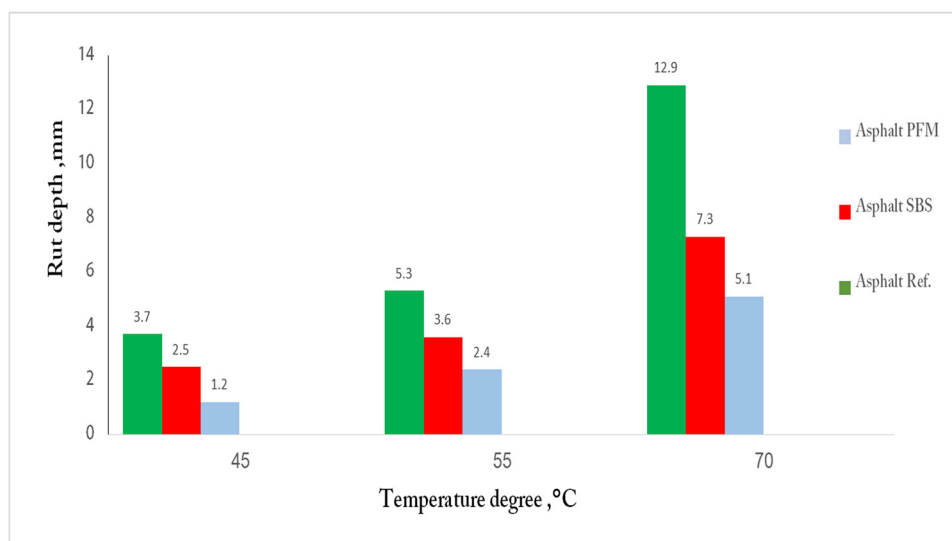


Figure 16: Shows the result of rutting resistance for the surface layer.



Figure 17: Rutting device.

Adding “NPFM” to the asphalt formed a strong mesh that can resist continuous heavy loads for long periods and high temperatures. Figure 16 represents the results of the

rutting test at 45, 55, and 70°C. The test was conducted using the rutting device (Figure 17). Figure 18 shows the paving of the samples in the mold, and Figure 19 shows the sample pressing device (Table 6).



Figure 18: Paving of samples in the mold.



Figure 19: Sample pressing device.

Table 6: Rutting results

Layer type	Temperature (°C)	Filler type	Asphalt type	N cycles	Rut depth (mm)
Surface	45	Lime stone dust	Reference asphalt	10,000	3.7
Surface	45	Lime stone dust	Asphalt SBS	10,000	2.5
Surface	45	Lime stone dust	Asphalt NPFM	10,000	1.2
Surface	55	Lime stone dust	Reference asphalt	10,000	5.3
Surface	55	Lime stone dust	Asphalt SBS	10,000	3.6
Surface	55	Lime stone dust	Asphalt NPFM	10,000	2.4
Surface	70	Lime stone dust	Reference asphalt	10,000	12.9
Surface	70	Lime stone dust	Asphalt SBS	10,000	7.3
Surface	70	Lime stone dust	Asphalt NPFM	10,000	5.1

4 Economic analysis

- In the case of using 4% modified asphalt (SBS), the cost is higher by 25% than when using regular asphalt, while in the case of using 4% modified asphalt (NPFM), the cost increases by 5% for sustainability purposes.
- If commercial melamine powder is used to produce modified asphalt, the cost increases by 10% over regular asphalt production.

5 Conclusion

Through this study, the properties of the local asphalt binder were strengthened and improved by adding “NPFM” at two rates of (4 and 2%). The rheological behaviors of the asphalt modified with “NPFM” were obtained through a series of tests according to the international classification superpave to determine the value of PG and compare it with asphalt modified with SBS. The effectiveness of the asphalt with the new additive was confirmed by testing in the asphalt mixtures. The results were extracted and analyzed as follows:

1. From the viscosity results, the improved material “NPFM” showed a higher increase in the binder asphalt's viscosity than with the modified asphalt SBS. The viscosity increased by 139.8% for melamine-modified asphalt, while the percentage increase for SBS asphalt was equal to 121.4%. Despite the higher viscosity of “NPFM,” the mixture sample needs a lower mixing temperature than the SBS asphalt mixing temperature, and this prevents damage to the adhesives during mixing and the production of modified asphalt mixtures. In addition, no such emissions occur when mixing “NPFM” with asphalt because melamine is thermally stable, so it maintains the viscosity of the mix.
2. The addition of “NPFM” to the local asphalt led to an increase and improvement in the performance to become PG (82-10) and thus led to an improvement in the durability of asphalt mixtures (stability) by large values to prevent the occurrence of scratches in Iraq according to “PG” required in the governorates of Iraq, which is in general PG (76-10). Therefore, adding this type of polymer provides excellent benefits for achieving the desired goal.
3. Moisture damage resistance was achieved when using “NPFM” more than the TSR values of SBS-modified asphalt binders, which were within a high improvement rate of more than 80%. The percentage increased (85.44%) in TSR % of the “NPFM” mixture compared to the “SBS” mixture, which was (83.61%).
4. The importance of the cost of asphalt modifiers has been estimated on the environment, especially when comparing

the asphalt mixture modified with “NPFM” with the modified asphalt mixture - SBS, through the recycling and use of damaged melamine plates. The results of the tests conducted on the mixture containing melamine proved that dispensing it with the modified binder SBS is possible. The result of the asphalt mixtures improved with melamine, which confirmed its high effectiveness during different conditions and gave better results than the modified asphalt mixture with SBS, especially the moisture damage test as one of the most critical problems leading to the destruction of asphalt concrete. This, in turn, reduces large amounts of road maintenance work.

5. Depending on the results of the fragmentation resistance of the asphalt modified with “NPFM,” the cracking resistance increased by 67.6% at 45°C, 80.6% at 55°C, and 60.5% at 70°C compared to the local asphalt mixtures modified with SBS, where the percentages were 32.4, 32.1, and 43.4% at 45, 55, and 70°C, respectively.

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Conflict of interest: Authors state no conflict of interest.

Data availability statement: Most datasets generated and analyzed in this study are in this submitted manuscript. The other datasets are available on reasonable request from the corresponding author with the attached information.

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