POLYMERIC MIXTURES AS MODIFIERS FOR THE RHEOLOGICAL PROPERTIES OF PAVING ASPHALT

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Received 23.10.2024
Accepted 26.12.2024

Abstract: *Due to the significant impact on enhancing the rheological properties of asphalt, many researchers were eager to enhance the specifications of asphalt by employing different additives, particularly polymeric additives. This is attributed to their significant impact on enhancing the rheological properties of asphalt that make it more suitable for a variety of uses, particularly in the process of paving. In this research, ethylene vinyl acetate (EVA) and Noflake were combined in a polymeric mixture to modify the material of asphalt. Different weights of mix polymers were utilized in the aforementioned polymer combination. At a constant temperature of 150 °C for 60 minutes, the polymeric mixture (EVA with Noflake) was added to the asphalt in different weight percentages while anhydrous aluminum chloride was present as a catalyst. The findings showed that the amended asphalt's qualities had clearly changed. Overall, there was a possibility of benefiting from Noflake material in the rheological modification of asphalt by using it as a mixture with EVA in the modification of asphalt. Noflake alone failed in the rheological modification, and at the same time, gave good results when used as a mixture with EVA. It was concluded that several measurements were made on the samples whose physical and chemical properties have been modified, as well as on the original asphalt, which were represented by measuring the ductility, penetration, softening point, and percentage of the separated asphalt.*

Key words: *asphaltenes, catalyst, polymers, rheological, softening, ethylene vinyl acetate, plastic waste*
DOI: 10.32737/2221-8688-2025-4-493-505

Introduction

Asphalt is often referred to as bitumen, and is produced by the direct distillation of crude oil. It is typically a very viscous, black, sticky liquid or semi-solid that comes from petroleum. About 70% of asphalt is utilized as a binder with particles to make asphalt concrete, and it is thermoplastic glue that is commonly used for paving, mastic, and flattening. Since crude oil has a high molecular weight in comparison to other components of crude oil and a high density of 1.0–1.1 g/cm^3, it is the primary source of asphalt. A further characteristic of asphalt is its dark brown or black hue [1-4]. It is mostly composed of condensed hydrocarbons that are paraffinic, naphthenic, and aromatic; these hydrocarbons are soluble in carbon disulphide liquid (CS_2) to varying degrees (70–98%). In addition to modest amounts of iron, aluminum, and silicon [5, 6], the asphalt composition also includes considerable amounts of nitrogen, oxygen, and sulfur-containing cyclic and noncyclic molecules. Road paving often uses asphalt. Some flaws, such cracks and deformation of roads, occur over time due to the severe climate and the maximum stresses on the original unaltered asphalt utilized in the paving process. In order to create asphalt pavement that is more resistant to the aforementioned elements, effort must be done to enhance the rheological qualities of asphalt. This is because asphalt has a strong adhesion with different minerals and adequate viscosity properties in addition to being inexpensive [7, 8]. Research on improving the specifications of asphalt through different chemical and physical treatments and the use of multiple additives, particularly polymeric ones, was spurred by the realization that additives have a major impact on improving the rheological properties of asphalt and making it more suitable for a variety of uses, particularly in the process of paving. Polyethylene terephthalate (PET) waste materials were added to asphalt in a study by [9, 10]. Huge volumes of plastic waste pose a serious disposal problem. Plastic items in solid trash may leak dangerous chemicals into the soil, which can subsequently seep into nearby rivers and lakes or groundwater, endangering the lives of any species that would consume tainted water [11-13]. Then, a number of measurements were performed using an infrared analysis, beam bending meter, dynamic shear meter, and moisture susceptibility meter. According to the findings, samples including additives made from PET and RAP performed better overall than ones using traditional binders [14-15]. The rheological properties of asphalt primer, which is commonly used in Egypt, were investigated after it was modified using different polymer products and waxes. Superplastic, Showax, used plastic bags, and polypropylene polymers are the additions. Scanning electron microscopy (SEM), rotating viscometer, penetration, and softening point tests [16-18] were conducted on the original and modified varieties of asphalt. The adjusted combinations showed improved mechanical properties and resistance to moisture damage, according to the findings. Overall, the findings demonstrate that improving pavement performance may be achieved by using polymer or wax materials as modifiers. Low density polyethylene (LDPE) and ethylene vinyl acetate (EVA), a polymeric combination, were introduced by [19, 20]. The findings showed that the right combination reduced both the resistance to stress and the persistent deformations. In addition, the researchers studied the rheological properties of the modified asphalt using a commercial adhesive that mainly consisted of EVA and also using an air-blowing procedure in the presence of anhydrous aluminum chloride. The findings demonstrated that, in addition to the most crucial use - paving, the samples met the requirements for use in mastic and flattening, among other materials [21-22]. The chemical and physical characteristics of asphalt treated with waste cooking oil (WCO) and polymeric mixes (styrene-butadiene-styrene (SBS) and EVA) were investigated by [23, 24]. The findings showed a discernible improvement in the amended asphalt's characteristics, but the storage stability was weakened [25, 26]. Using a variety of experimental laboratory test techniques, authors of [27] investigated the use of waste plastic materials, such as plastic bottles (PET) and gas pipes (PE), in asphalt materials as a bitumen modifier. The Marshall technique of asphalt mix design is commonly used in construction materials labs to choose and proportion aggregate and asphalt components for pavement construction. Marshall samples were generated using this methodology [28, 29]. By applying pressure to the test sample and monitoring its deformation, the stability and crawling measurements are simultaneously recorded using the reading recorder in the device [30, 31], which included five different rates (0%, 5%, 10%, 15%, and 20%) of the plastic content by the total weight of bitumen. This test provides an indication of the suitability of the asphalt for paving. After one and four weeks, the samples were confirmed, and the findings showed that, at the four-week mark, the stability of the asphalt concrete amended with plastic had risen. Furthermore, adding plastic waste is a successful way to reuse plastic waste when building roads and provides social and economic benefits as a sustainable method for roads [32]. The impact of using domestic waste polyethylene terephthalate (PET) plastic in the C320 certified category was investigated by [33, 34]. In Australia, this kind of bitumen is often used for local road paving. There are two phases to the evaluation of the various PET-modified bitumen components: one for aged conditions and the other for non-aged situations. The ageing and wear of the bitumen binder were confirmed using the Rolling Thin Film Ovens (RTFOT), Pressure Ageing Vessel (PAV), and Dynamic Shear Scale (DSR) tests. The asphalt's resistance to ageing conditions and other qualities significantly improved at certain polymer percentages, according to the [35] investigated the impact of moisture damage and ageing on the mechanical properties of asphalt mixes treated with waste polymers, such as waste plastic and crumb rubber. Granite aggregate, 60/70 PEN asphalt, and grout (slaked lime) were used to make asphalt mixtures for asphalt concrete (AC14). Calculations were used to determine the modulus of elasticity, dynamic flow, and indirect tensile strength. After regulating the moisture, the modulus and rupture strength of asphalt mixes treated with plastic and rubber crumbs decreased to 9% and 17%, respectively.

Low density polyethylene, a plastic waste, was investigated by [36, 37] as a modified material for asphalt. Through this research, it was discovered that using low density polyethylene waste—whose ideal value is close to 4% of the weight of asphalt—improved the performance of asphalt mixes at a range of high temperatures. It also developed Marshall stability by using this rate of polymer (D113, ASTM) [38, 39].

In this study, the effect of ethylene vinyl acetate and Noflake polymer mixture on asphalt material modification in the presence of a catalyst was investigated.

Experimental part

The asphalt material treatment apparatus was loaded with 100 grams of asphalt. A catalyst of anhydrous aluminum chloride (AlCl3) was added to a combination of EVA and Noflake in varying quantities (0.5%) by weight. The reactants were thoroughly mixed at a temperature of 150 °C while being continuously shaken for 60 minutes. For some samples, the following tests were conducted: ductility (ASTM D113-99), penetration (ASTM D 36-95), softening point (ASTM D5-97), de-asphalting (ASTM D 6560-17), chemical immersion (ASTM D 1559-89), and Marshal measurement.

Results and discussion

By adding several types of additives that strengthen the asphalt's resilience to different environments and repetitive loads, the researchers hoped to improve the performance of asphalt used in paving. A polymeric combination comprising EVA and Noflake was employed in the current investigation. As shown in Table 1, EVA and Noflake were added to the asphalt mixture in different ratios and the reaction was catalyzed by 0.5% anhydrous aluminum chloride. Table 1 displays the results of treating the asphalt material with a mixture of 0.9 grams of EVA and 0.1 grams of Noflake in different concentrations.

Table 1. The rheological characteristics of asphalt treated with various proportions of mixture EVA+Noflake (0.9+0.1) at 150 ºc and a time of 60 minutes in the presence of 0.5% weight of anhydrous aluminum chloride

Samples	Penetration, mm (100 g, 5 sec, 25 ºC)	Softening point ºC	Ductility (cm, 25 ºC)	Mix ratio %	Asphaltene %
AS0	44.1	50	+150	0	20.2
AS1	43.2	56	+150	1	21.3
AS2	43.1	55	+150	2	23.6
AS3	42.7	57	141	3	27.1
AS4	41.8	60	+150	4	28.6
AS5	40	60	100	5	28.9
AS6	37	65	76	6	29.7

Table 1 showed that the polymeric mixture addition resulted in acceptable rheological properties to a degree of 5%. It was observed that among the requirements for using asphalt in paving were the values of ductility, penetration, and softening. In every way, they were appropriate. This study aimed to modify the rheological properties of asphalt using Noflake. Noflake by itself did not provide good percentages in many of the modifications performed during the modification process. Thus, we employed it in conjunction with EVA, yielding positive outcomes in several investigations [39, 40]. In order to capitalize on Noflake in the modification and increase it together with the EVA, it was decided to increase

Noflake in each treatment and decrease the amount of EVA. The outcomes achieved with the polymeric mixture's proportions are displayed in Table 2.

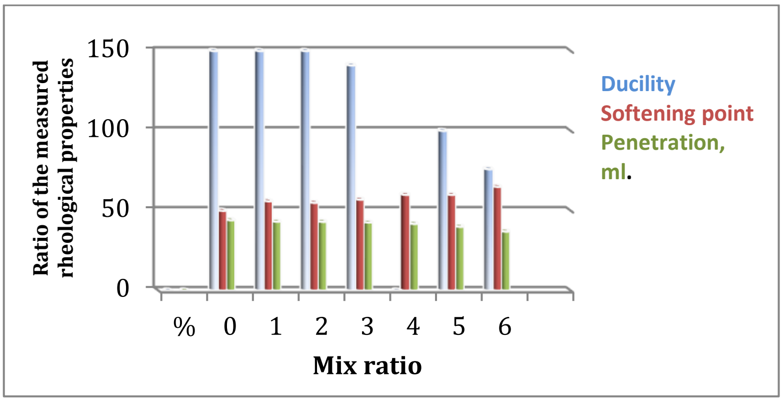

Fig. 1. The rheological characteristics of asphalt treated with the mixture (0.9g of EVA : 0.1g of Noflake)

Table 2. The rheological properties of asphalt treated with different rates of mixture EVA+NOFLAKE (0.8+0.2) at 150 °C for 60 minutes while 0.5% by weight of anhydrous aluminum chloride was present

Model	Penetration, mm (100 g, 5 sec, 25 ºC)	Softening point ºC	Ductility (cm, 25 ºC)	Mix ratio %	Asphaltene %
AS_0	44.1	50	+150	0	20.2
AS_7	43.8	55	+150	1	22.4
AS_8	42.0	65	147	2	22.7
AS_9	41.4	57	+150	3	24.6
AS_{10}	42.5	56	146	4	25.5
AS_{11}	41.6	60	131	5	27.1
AS_{12}	36	65	66	6	30.4

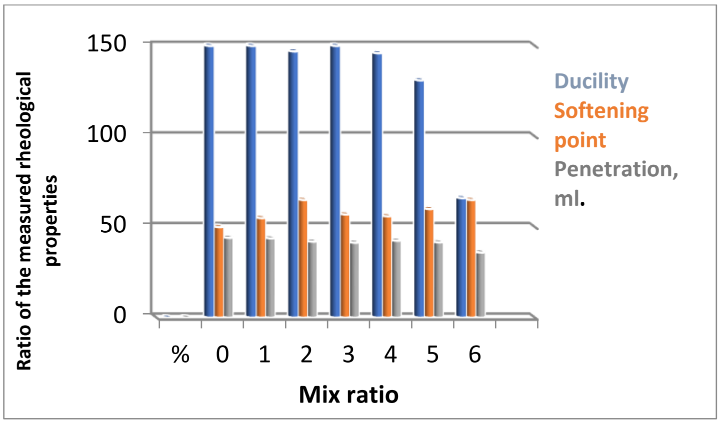

Fig. 2. The rheological characteristics of asphalt treated with a mixture (0.8 g of EVA : 0.2 g of Noflake)

It is evident from Table 2 that up to 5% of the polymeric mixture exhibited good rheological properties when EVA and Noflake were added to the asphalt at a ratio of 0.8 and 0.2. All of the ductility values were more than 100 at these percentages. Furthermore, in accordance with the Iraqi standard criteria shown in Table 10, the ductility and penetration values were within and among the required values for asphalt pavement. The graph also demonstrates that, on average, the rate of asphaltenes increases in tandem with the rate of the polymeric additive.

This is because a higher percentage of polymeric additives results in a higher molecular weight, which raises the amount of separated asphaltens. The following tables show the outcomes of our attempts to decrease the proportion of EVA and raise the percentage of the additive (Noflake) in each treatment: The results of asphalt treated with a polymeric mixture under the same previous conditions, but with various ratio of EVA:Noflake are shown in Tables 3-9.

Table 3. Asphalt treated with varying ratios of EVA + NOFLAKE (0.7+0.3) at 150 °C for 60 minutes with 0.5% by weight of anhydrous aluminum chloride was evaluated for rheological characteristics

Model	Penetration, mm (100 g, 5 sec, 25 ºC)	Softening point ºC	Ductility (cm, 25 ºC)	Mix ratio %	Asphaltene %
AS0	44.1	50	+150	0	20.2
AS13	42.7	55	+150	1	22.5
AS14	43.6	57	+150	2	24.7
AS15	42.3	60	+150	3	25.2
AS16	41.1	58	+150	4	26.7
AS17	38	58	76	5	28.4
AS20	36	61	80	6	29.7

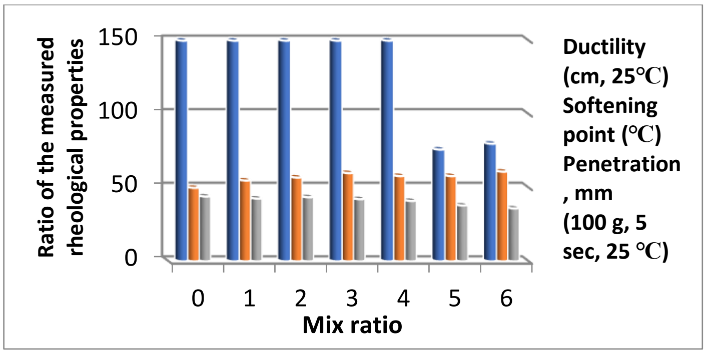

Fig. 3. Rheological characteristics of asphalt treated with a mixture (0.7 g of EVA: 0.3 g of Noflake)

Table 4. Asphalt treated with various ratios of EVA+Noflake combination (0.6+0.4) at 150 °C for 60 minutes with 0.5% by weight of anhydrous aluminum chloride was evaluated for rheological characteristics

Model	Penetration, mm (100 g, 5 sec, 25 ºC)	Softening point ºC	Ductility (cm, 25 ºC)	Mix ratio %	Asphaltene %
AS_0	44.1	50	+150	0	20.2
AS_{21}	44.5	55	+150	1	21.4
AS_{22}	43.8	56	148	2	22.6
AS_{23}	42.5	56	+150	3	24.3
AS_{24}	41.7	58	+150	4	25.8
AS_{25}	37.6	57	95	5	27.7

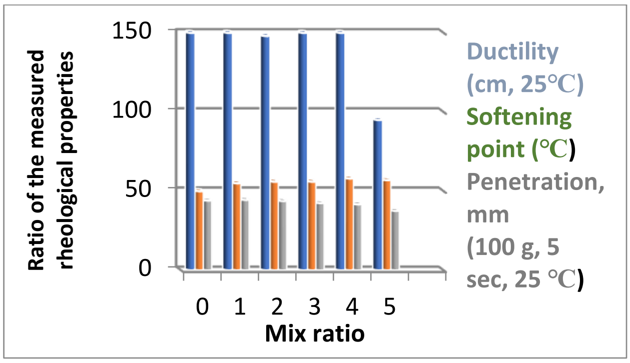

Fig. 4. Rheological characteristics of asphalt treated with a mixture (0.6 g of EVA: 0.4 g of Noflake)

Table 5. Asphalt treated with varying proportions of the EVA+Noflake mix (0.5+0.5) at 150 °C for 60 minutes with 0.5% by weight of anhydrous aluminum chloride was evaluated for rheological characteristics

Model	Penetration, mm (100 g, 5 sec, 25 ºC)	Softening point ºC	Ductility (cm, 25 ºC)	Mix ratio %	Asphaltene %
AS_0	44.1	50	+150	0	20.2
AS_{26}	44.1	54	+150	1	20.7
AS_{27}	41.9	57	144	2	25.3
AS_{28}	44.1	55	+150	3	20.2
AS_{29}	40.5	59	140	4	27.5
AS_{30}	40.7	61	74	5	30.6

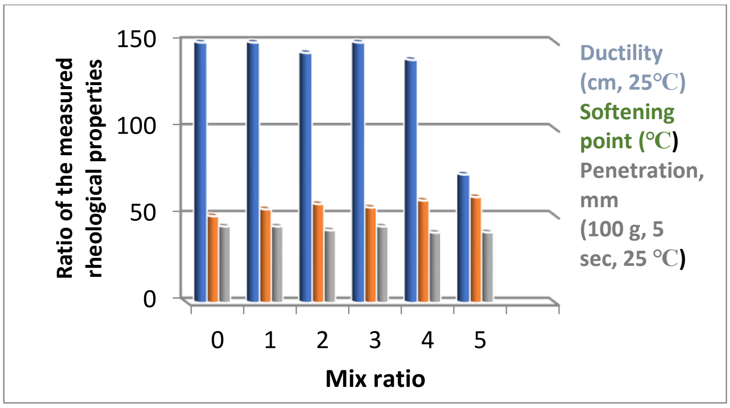

Fig. 5. Rheological characteristics of asphalt treated with a mixture (0.5 g of EVA: 0.5 g of Noflake)

Table 6. Asphalt's rheological properties after being treated with different ratios of the EVA+Noflake combination (0.1+0.9) at 150 °C for 60 minutes while 0.5% by weight of anhydrous aluminum chloride was present

Model	Penetration, mm (100 g, 5 sec, 25 ºC)	Softening point ºC	Ductility (cm, 25 ºC)	Mix ratio %	Asphaltene %
AS_0	44.1	50	+150	0	20.2
AS_{31}	43.4	59	141	1	24.3
AS_{32}	40.8	63	92	2	25.7
AS_{33}	40.1	65	77	3	26.1
AS_{34}	37.6	64	65	4	28.3
AS_{35}	29	71	35	5	38

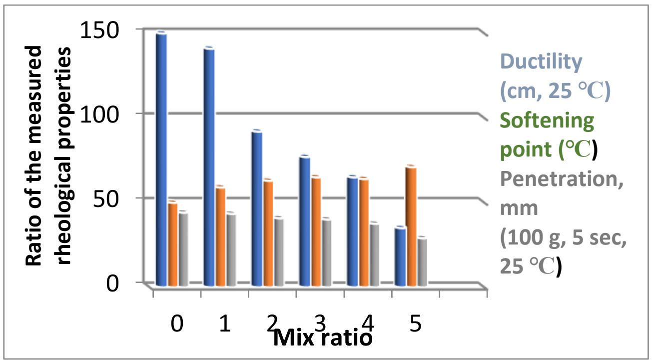

Fig. 6. Rheological characteristics of asphalt treated with a mixture (0.4g of EVA: 0.6g of Noflake)

Table 7. Asphalt treated with various ratios of the EVA+Noflake combination (0.2+0.8) at 150 °C for 60 minutes with 0.5% by weight of anhydrous aluminum chloride was evaluated for rheological characteristics

Model	Penetration, mm (100 g, 5 sec, 25 ºC)	Softening point ºC	Ductility (cm, 25 ºC)	Mix ratio %	Asphaltene %
AS_0	44.1	50	+150	0	20.2
AS_{36}	41.6	59	+150	1	22.3
AS_{37}	40.7	59	+150	2	23.5
AS_{38}	40.6	60	140	3	25.8
AS_{39}	38.7	64	80	4	28.4
AS_{40}	37.2	65	56	5	31.3

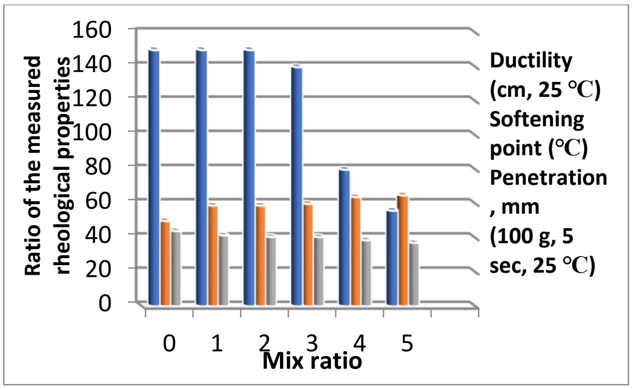

Fig. 7. Rheological characteristics of asphalt treated with a mixture (0.3 g of EVA: 0.7 g of Noflake)

Table 8. Asphalt treated with varying ratios of EVA+Noflake (0.3+0.7) at 150 °C for 60 minutes with 0.5% by weight of anhydrous aluminum chloride was evaluated for rheological characteristics

Model	Penetration, mm (100 g, 5 sec, 25 ºC)	Softening point ºC	Ductility (cm, 25 ºC)	Mix ratio %	Asphaltene %
AS_0	44.1	50	+150	0	20.2
AS_{41}	41.3	58	+150	1	21.5
AS_{42}	40.7	59	+150	2	22.6
AS_{43}	40.2	61	120	3	25.8
AS_{44}	39.2	62	100	4	26.2
AS_{45}	38.5	65	77	5	27.3

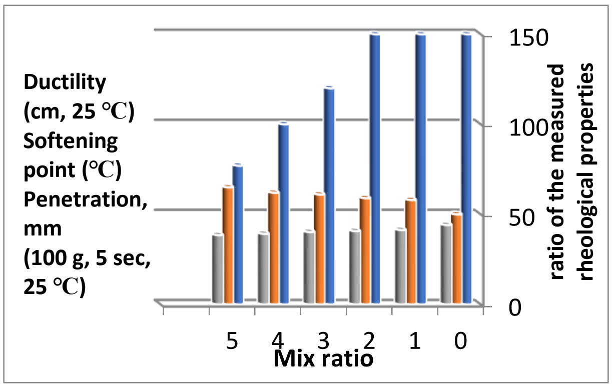

Fig. 8. The rheological characteristics of asphalt treated with a mixture (0.2 g of EVA: 0.8 g of Noflake)

Table 9. Asphalt treated with varying ratios of the EVA+Noflake combination (0.4+0.6) at 150 °C for 60 minutes with 0.5% by weight of anhydrous aluminum chloride was evaluated for rheological characteristics

Model	Penetration, mm (100 g, 5 sec, 25 ºC)	Softening point ºC	Ductility (cm, 25 ºC)	Mix ratio %	Asphaltene %
AS_0	44.1	50	+150	0	20.2
AS_{46}	40.7	58	140	1	23.1
AS_{47}	40.9	61	131	2	23.5
AS_{48}	42.1	59	>150	3	24.2
AS_{49}	40.3	60	120	4	25.3
AS_{50}	38.7	63	77	5	26.2

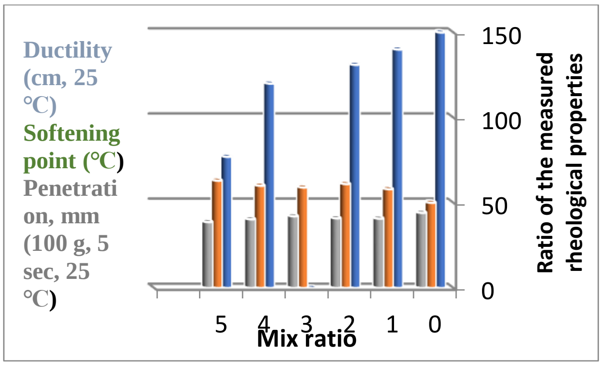

Fig. 9. The rheological characteristics of asphalt treated with the mixture (0.9 g of EVA: 0.1 g of Noflake)

The standard specifications for asphalt utilized in paving, asphalt used to produce mastic, a waterproofing material, and asphalt used for surfacing are displayed in Tables 10–12 and were the basis for the earlier modification process results [41].

Table 10. Iraqi paving asphalt

Rheological characteristics	Min	Max
Softening – point, ºC	54	60
Penetration (100 gm, 5sec, at 25ºC)	40	50
Ductility (cm, at 25 ºC)	100	-

Table 11. American mastic properties

Rheological characteristics	Min	Max

Softening – point, ^oC	54	65
Penetration (100 gm, 5sec, at 25 ^oC)	20	40
Ductility (cm, at 25 ^oC)	15	-

Table 12. Properties of Flattening-asphalt

Rheological characteristics	Min	Max
Softening-point, ^oC	57	66
Penetration (100 gm, 5sec, at 25 ^oC)	18	40
Ductility (cm, at 25 ^oC)	10	-

Nine high-quality samples (AS31, AS36, AS41, AS46, AS28, AS23, AS1, AS9, and AS14) were chosen, and chemical immersion and the Marshall test were performed on them. Furtheore, these parameters were tested for the original asphalt to determine how much the updated models had improved and, thus, if these models were appropriate for the tiling job. Through this study, stability and flow are measured, and the Marshall Quotient is computed by dividing the result of Marshall stability by the flow value [39-41]. The Iraqi Authority of Standards for Constructing Roads and Bridges' Table 13 displays the Marshal values of stability and creep of the chosen samples that were acquired and compared with the original model [28].

Table 13. Stability and flow values of the original and modified asphalt and the Iraqi Authority of Standards of Constructing Roads and Bridges (S.C.R.B)

Model	Modified asphalt (best models)			Percentage of asphalt added to aggregate (%)
AS_0	MQ	Flow (mm)	Stability (KN)	
AS_1	2.28	4.5	10.3	4.5
AS_9	4.76	3.4	16.2	
AS_{14}	5.29	2.7	14.3	
AS_{23}	5.17	3.4	17.6	
AS_{28}	5.8	3.0	17.4	
AS_{31}	4.75	2.9	13.8	
AS_{36}	4.66	3.3	15.4	
AS_{41}	4.26	3.4	14.5	
AS_{46}	5.68	2.9	16.5	
AS*	5.68	3.4	17.5	
	3.5 Min	2-4	7 Min	

It is evident from the accompanying table that when utilized as asphalt for paving, all of the modified models outperform the original model. Additionally, it has been observed that the MQ value of the updated models is greater than that of the original asphalt. This suggests that compared to untreated asphalt, modified asphalt is more resistant to long-term deformation. After the asphalt was mixed with aggregates for models AS_{31}, AS_{36}, AS_{41}, AS_{46}, AS_{28}, AS_{23}, AS_1, AS_9, and AS_{14}) and the original model, a chemical immersion test was performed to assess the asphalt's durability to acid rain and high temperatures [42-44]. The outcomes of the asphalt's separation from the debris of the original asphalt and the modified models are displayed in Table 14.

Table 14. Chemical Immersion results

No. of modified sample	Percentage of Na_2CO_3 gm	R and WNO	R&WNO For the original asphalt	R&WNO for the modified samples
AS_0	0.025	1		

	0.041	2		
	0.082	3	3	
AS_{46}	0.164	4		
AS_{28}, AS_{31}, AS_{41}	0.328	5		5
AS_{23}, AS_{36}	0.656	6		
AS_{1}, AS_{9}	1.312	7		7
AS_{14}	2.624	8		

Table 15 and the separation values show the quantity of sodium carbonate at which the asphalt began to separate from the aggregate. The modified versions began to separate a higher quantity of sodium carbonate than the original asphalt. The Riedel and Weber number, abbreviated as (R&W) is represented by the integers 0 through 8. The amount of sodium carbonate, or (0.025) grams in (50) milliliters of distilled water, is indicated by number (1). With 2.624 grams, number (8) displays the maximum quantity of sodium carbonate. The modified asphalt is more resistant to acid rain and rising temperatures because it sticks with the aggregate longer than the original asphalt does since it has higher separation values than the original asphalt. As a result, the polymeric combination enhanced the rheological characteristics of asphalt and increased its resistance to variations in temperature [43-44], repetitive loads, and environmental factors. Through this investigation, it was also discovered that Noflake material may be useful in the rheological adjustment of asphalt by being combined with EVA to modify the asphalt. When combined with EVA in the amounts that were made clear in the preceding tables, Noflakes performed well in the rheological modification but poorly when employed alone.

Conclusion

The advantages of using asphalt additives include increased pavement life, improved durability, fracture resistance, and flexibility of the asphalt binder. In comparison to commercial polymers, adding EVA to asphalt improves its flexibility, lessens cracking, and has some positive effects on the environment and the economy. When bitumen is blended with additives, it becomes more temperature sensitive and has a wider usable temperature range. The rheological characteristics and fatigue resistance of modified bitumen binder and mixes are enhanced by the use of EVA.

1. Based on this study, we can say that the combination of ethylene vinyl acetate (EVA) and Noflake in a polymeric mixture was used to affect the rheological properties of asphalt, resulting in a dual action.
2. While Noflake by itself performed poorly in the rheological modification, it performed well in combination with EVA.
3. The investigation's findings indicate that the modified samples outperformed the original asphalt, indicating that this additive may be applied to tiling projects.

Acknowledgement

The authors recognize the University of Kirkuk, the Department of Chemistry, the College of Education for Pure Science, and the University of Mosul.

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