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Research Article

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# Experimental investigation of usability of modified asphalt binder with waste expanded polystyrene (EPS)

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## Highlights

- Investigated the potential use of waste EPS in asphalt binder modification
- EPS modified binder provide better rutting resistance
- EPS modified binder lowers fatigue
- EPS modified binder lowers low temperature cracking resistance
- Using waste EPS in the binder has many economic and environmental benefits

## Abstract

EPS (Expanded polystyrene) is widely used in construction and packaging industries. Unfortunately, EPS is not mostly recyclable because it might not be economically feasible to store, transport, and process EPS so that it becomes recyclable. Therefore, alternative waste management strategies are urgently needed. The main objective of this study is to experimentally investigate the potential use of waste EPS in asphalt binder modification. As part of this study, a base asphalt binder with a Penetration grade of 70/100 was modified with waste EPS in four different ratios (0%, 1.5%, 3% and 4.5%, by weight of asphalt binder). The modified asphalt binder specimens were tested based on several physical (penetration, ductility, softening points and viscosity) as well as rheological/Superpave binder grading system tests including Rolling Thin Film Oven (RTFO), Pressure Aging Vessel (PAV), Dynamic Shear Rheometer (DSR), and Bending Beam Rheometer (BBR). Based on the tests results, rheological and engineering properties of the modified asphalt binders were compared with the ones of the base binder so that effect of waste EPS addition on the properties of the base binder was evaluated; and how the Superpave binder grades of the modified binders were changed with the additions of waste EPS was discussed. It was found out that modifying asphalt binders with EPS provided them with a better rutting resistance at the expense of lowering their fatigue and low temperature cracking resistance. Overall, it could be concluded that waste EPS can be potentially used in the asphalt binder modification by paying special attention to these factors, and it should be noted that using waste EPS in the asphalt binder modification has many economic and environmental benefits.

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## 1. Introduction

Asphalt binder is one of the two ingredients of an asphalt concrete, binding aggregates in the asphalt concrete together, a hydro carbonic, sticky, and waterproof material, produced as a result of the refinement of petroleum. Although it is mostly used as a pavement material, it has some other uses such as waterproofing, pipe and corrosion protection etc. The transportation industry has developed rapidly all over the world in recent years. With the increase in numbers and weights of vehicles, pavements tend to deteriorate faster. Being a viscoelastic material, rheological properties of the asphalt binder depend on the level of load frequency applied to it as well as its temperature and age conditions. Because of the rheological behavior of the asphalt binder, it might need to be modified to improve some of its properties such as fatigue and low-temperature cracking and rutting resistance so that its service life could be extended [1]. Asphalt binder modification could be defined as the mixing asphalt binder with various additives in certain percentages and under certain conditions to increase the performance of the mixture to be used in flexible pavements. Modified asphalt binder is mostly obtained by proportionally mixing various petrochemical products such as polymers in different proportions. The purpose of asphalt binder modification with polymers is to reduce their thermal and fatigue cracking potential at low temperatures, and to increase its resistance against rutting at high temperatures by changing their

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viscoelastic behavior [2]. The positive effects of the modification of asphalt binder can be summarized as follows [3]:

- Extending the temperature range of the asphalt binder, increasing the softening point, and reducing the penetration value
- Reducing its sensitivity to temperature
- Increasing resistance against aging and oxidation
- Reducing cracks and maintaining flexibility at low service temperatures
- Reducing rutting by acting as a hard and solid material at high temperatures
- Enhancing the fatigue resistance
- Increasing the tensile strength
- Improving its workability
- Reducing periodic maintenance costs
- Preventing flushing

Polymers are the most common additives used for asphalt binder modification. Currently, only very few percentages of polymers are recycled worldwide, meaning that they accumulate rapidly in landfills. They take not only a very long time to decompose but also cause environmental issues as they pollute sediments, soils, and water. As new products emerge on the market and rapidly replace old ones, landfilling these polymer-based materials not only has a negative environmental impact, but also hinders the opportunity for reusing them. One potential area to reuse these polymers might be in asphalt mixes. Some materials such as polyethylene (PE) [4, 5], synthetic polymers including styrene-butadiene-styrene (SBS) [6], ethylene vinyl acetate (EVA) [7] were tested to be used in the modification of asphalt binder to improve the properties of asphalt binders [8].

Expanded polystyrene (commonly called styrofoam or EPS foam) is commonly used in the construction and packaging industries. EPS has been much used in these industries due to its comparably better thermal, mechanical, and physical properties. Because EPS is mainly a byproduct of the petroleum industry, which is an unsustainable and non-renewable material source, it is an increasingly scarce and highly polluting commodity. Unfortunately, polystyrene food service packages are not mostly recyclable because it might not simply be economically feasible to store, transport, and process them before they become recyclable. Therefore, alternative ways to reuse and recycle waste EPS should be found out [8]. Some studies have investigated using waste EPS in asphalt modification. Nciri et al. [8] aimed to use EPS as performance improving additives for road pavement applications by testing asphalt binders modified with various percentages of EPS (0%, 3%, 6%, 9%) based on their chemical, thermal and morphological properties. They stated based on the morphological, chemical, and thermal test results that waste EPS could be potentially used in the modification of asphalt binder, but further mechanical and rheological tests are needed to be conducted to support this claim. Ramadan et al. [9] investigated the physical and rheological properties of the asphalt binder specimens by modifying a base binder with waste EPS in different percentages (between 0.2% and 2% with 0.2% increments, by weight of the asphalt binder specimens). Ductility, penetration, and softening point tests as well as Rotational Viscometer (RV), and Dynamic Shear Rheometer (DSR) tests were applied to the modified asphalt binder samples. They found that penetration and ductility test results decreased and softening, and flash points test results increased as waste EPS contents increase. Based on DSR test results, EPS addition enhanced the high temperature performance of the tested binder samples. Akter and Raja [10] evaluated the effect of shredded EPS on the performance of asphalt concrete and asphalt binder. They applied some physical tests (ductility, penetration, softening point, flash point, fire point and specific gravity) on the EPS-modified asphalt binder samples (in different proportions, namely between 0.25% and 1%, with 0.25% increments, by weight of total aggregate). They found that the addition of 1% EPS, by weight of total aggregate, the maximum EPS content they used, produced the maximum flash, fire, and softening points but the minimum ductility values. Yıldız et al. [11] investigated modifying a base asphalt binder with waste EPS with ratios of 2%, 4%, 6%, and 8% (by weight of asphalt binder) and the following tests were applied to the modified binders: ductility, softening point, flash point, penetration and specific gravity. They found that increase in EPS content decreased the penetration and ductility values. They recommended that modified binders with higher EPS contents might be used in comparably hotter regions where low penetration binder is needed.

Based on the studies reviewed regarding on the potential use of waste EPS on asphalt binder modification, most of the studies concluded that waste EPS could be potentially used in the modification of asphalt binder to some extent by mostly considering limited number of types of physical tests such as ductility, penetration, softening point, flash point, fire point and specific gravity etc. To better evaluate the effect of waste EPS on the performance of asphalt binders in wide-range of aging and temperature conditions, more comprehensive rheological tests including Superpave binder grading system tests including DSR, Bending Beam Rheometer (BBR), Pressure Aging Vessel (PAV) and Rolling Thin Film Oven (RTFO) should also be conducted.

The main purpose of this study is to experimentally investigate the potential use of EPS waste in asphalt binder modification. As part of this study, a base asphalt binder with a Penetration grade of 70/100 was modified with waste EPS in four different ratios (between 0% and 4.5%, with 1.5% increments, by weight of asphalt binder). The modified asphalt binder specimens were tested based on several physical (penetration, ductility, softening points, and viscosity) as well as rheological/Superpave binder grading system tests (DSR, BBR, RTFO and PAV). Based on the tests results, rheological and engineering properties of the modified asphalt binders were compared with the ones of the base binder so that effect of EPS waste modification on the properties of the base binder was evaluated; how the Superpave binder grades of the modified binders were changed with the additions of waste EPS was discussed.

## 2. Materials and Methods

As part of this study, an asphalt binder specimen with a 70/100 Penetration grade, provided by a local refinery, has been used as the base binder. The EPS used in the current study has been provided by a local packaging factory, the factory is using EPS as a packaging material and wasting some of the EPS during production. As stated earlier, as part of this study, the base asphalt binder specimens were modified with EPS waste in four different ratios (between 0% and 4.5%, with 1.5% increments, by weight of asphalt binder). Modified asphalt binder samples were prepared based on the following steps, determined based on the previous studies [12, 13] (Figure 1):

- The base asphalt binder specimen was heated in an oven for 30 minutes at 180±5°C so that it becomes fluid.
- 500 grams of fluidized asphalt binder were added to the mixer's metal chamber.
- To create a homogenous thermal source, the binder added to the metal chamber was allowed to remain inside the thermal jacket of the heater source (at 180±5°C) until it reaches equilibrium.
- EPS was poured into the hot asphalt binder at a rate determined by the weight of the asphalt binder, adding a percentage to each. EPS-binder blend was mixed with a mechanical shear mixer operating at 3000 rpm/min for 90 minutes to prepare the modified asphalt binder containing EPS.

The modified asphalt binder samples were tested based on penetration, ductility, softening points, and viscosity as well as rheology/Superpave binder grading tests (DSR, BBR, RTFO and PAV).

### 2.1. Physical test methods

Physical test methods including Penetration, softening point, ductility, and Rotational Viscometer (RV) tests were used to evaluate basic engineering properties of the modified asphalt binders.

#### 2.1.1. Penetration test

Penetration value of an asphalt binder is determined based on the depth a standard needle penetrates an asphalt specimen conditioned at a specified temperature (25° C), load (100 gr) and time (5 sec) conditions, following TS EN 1426 standard. Measurements of penetration depth are reported in penetration units of 0.1 mm. The penetration value is inversely proportional to the consistency of an asphalt binder, and the higher the penetration, the softer the asphalt binder is.

(a) (b) (c)

Figure 1. Preparation steps of the modified asphalt binder: a) weighing EPS based on percentages determined by the weight of the bitumen, b) adding EPS to base binder and c) mixing modified binder using a shear mixer

#### 2.1.2. Softening point test

The Softening point test, also called as Ring and Ball test, determines the temperature at which asphalt binder is soft enough not to be able to support the weight of a 3.5-g steel ball. This test method is used to measure the sensitivity of asphalt binder to temperature changes. Following TS EN 1427 standard, the experiment is conducted by heating an asphalt binder sample in a standard ring, with a ball on it, at a certain constant rate. The temperature when the binder sample, which softens as a result of heating, touches the bottom of the metal frame (falling a distance of 25 mm.) is recorded as the softening point.

2.1.3. Ductility test

Ductility is one of the important features of asphalt binders. It shows how many cm a standard-sized briquette of asphalt binder could stretch without breaking. The test samples are drawn at a speed of 55±0.25 cm/min at 25̊ C and the length of the samples at breaking points are reported in cm as ductility values. The test is conducted based on the ASTM D 113 standard.

2.1.4. RV test

RV test, following ASTM D 4402 standard, is used to determine viscosity of asphalt binders typically at 135°C and 165°C. It basically measures the torque value of a cylindrical spindle submerged in an asphalt binder sample while it maintains a constant rotational speed at a certain temperature. This test provides information about optimum temperature ranges for pumpability, workability and mixability properties of the tested asphalt binder specimens.

**2.2. Rheological test methods**

Rheological test methods, being used as part of the Superpave binder grading standard [14], were used to evaluate the rheological behavior of asphalt binders at various temperatures, aging and load frequency cases. These tests include RTFO, PAV, DSR and BBR tests.

2.2.1. RTFO test

RTFO test, following ASTM D 2872 standard, is carried out to simulate short-term-aging (during manufacturing and placement) properties of asphalt binders and to determine the percentages of volatiles in the asphalt binders lost throughout the short-term-aging process. As part of the test, first, unaged asphalt binder specimens are placed in cylindrical glasses and the cylindrical glasses are then placed in a rotating carriage within an oven. The carriage then rotates for 85 mins at 163°C while air with a certain pressure flows into the bottles throughout the test. Then, the glass bottles are weighed to quantify mass loss of the asphalt binder specimens and the short-time-aged asphalt binder specimens are then kept to be used in DSR and PAV tests.

2.2.2. PAV test

PAV test, following AASHTO R 28 standard, is used to simulate long-term-aging (aging over a 7-to-10-year period) properties of asphalt binders. As part of the test, first, RTFO-aged samples are placed in stainless steel pans and the pans are then put in a heated vessel for 20 hours, with a constant pressure of 2.1 MPa.

2.2.3. DSR test

Using DSR test, viscoelastic behaviors of asphalt binders are characterized at medium and high temperatures, following AASHTO T 315 standard. The test is applied to unaged, RTFO-aged or PAV-aged test specimens. The test specimens are exposed to oscillation movement during the test at 10 rad/sec (1.59 Hz) in a sinusoidal waveform, simulating a traffic speed of about 90 km/h. Using DSR test results, the complex modulus (G*) and phase angle (δ) results at different temperatures are reported. This test is used to evaluate fatigue cracking and rutting resistances of asphalt binders at medium and high temperatures, respectively.

2.2.4. BBR test

Using BBR test, low temperature stiffness and relaxation properties of asphalt binders are characterized, following AASHTO T 313 standard. Using a small asphalt beam, creep stiffness and the slope of the master stiffness curve at 60 seconds (m-value) values of the asphalt binder specimens are determined at low temperatures to evaluate low temperature cracking resistance of specimens.

**3. Results and Discussion**

In this part of the paper, test results for both physical and rheological properties of modified asphalt binder specimens with waste EPS in four different ratios (0%, 1.5%, 3% and 4.5%, by weight of asphalt binder) were presented.

**3.1. Physical test results**

Figures 2(a), 2(b) and 2(c) show penetration, softening point, and ductility test results of the modified asphalt binder specimens with the changes in waste EPS contents, respectively. As can be seen in Figure 2, as waste EPS contents in the modified asphalt specimens increased, their Penetration and ductility values decreased, and softening point values increased. Figure 3 shows viscosity test results of the modified asphalt binder specimens with the changes in waste EPS contents at 135°C and 165°C. As can be seen in Figure 3, as waste EPS contents in the modified asphalt specimens increased, their viscosity test results also increased in both temperature cases, the viscosity test results were consistently higher at 135°C compared to that at 165°C. For the case of using 4.5% EPS in the modified asphalt binders, viscosity values increased by 96% and 140% at 135°C and 165°C, respectively. Based on the viscosity test results, Superpave binder grading standard [14] provides viscosity ranges for asphalt binders to have for mixing and compaction. Temperatures corresponding to 0.17±0.02 Pa.s (170±20 cP) and

0.28±0.03 Pa.s (280±30 cP) viscosity values are specified as mixing and compaction temperature ranges, respectively. Considering the viscosity test results presented in Figure 3 and the viscosity ranges for mixing and compaction, mixing and compaction temperature ranges for the modified asphalt binder specimens are calculated and presented in Table 1. As can be seen in Table 1, as EPS contents in the modified asphalt binders increased, corresponding mixing and compaction temperatures also increased. Increase in mixing and compaction temperatures might require higher fuel cost to bring asphalt binders to mixing and compaction temperatures as well as causing additional environmental problems due to higher levels of greenhouse gases emitted during additional heating.

Based on all of the physical test results, it could be concluded that as waste EPS contents in the modified asphalt binder specimens increase, they become stiffer and their resistance against rutting tend to increase. On the other hand, having stiffer and less-flexible asphalt binders tend to have lower resistance against fatigue cracking.

Table 1. Recommended mixing and compaction temperature ranges of the modified asphalt binder with four different EPS contents

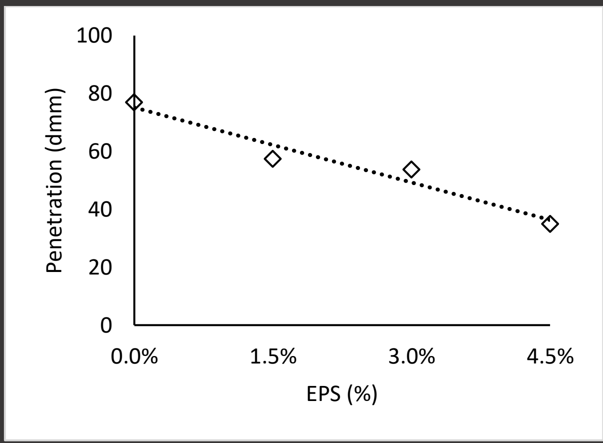
| EPS Content | Mixing Temperature (°C) | | Compaction Temperature (°C) | |
|---|---|---|---|---|
| | Lower | Higher | Lower | Higher |
| 0% | 149.7 | 154.5 | 135.3 | 142.5 |
| 1.5% | 157.0 | 160.6 | 146.3 | 151.7 |
| 3% | 158.6 | 161.9 | 148.6 | 153.6 |
| 4.5% | 162.4 | 165.0 | 154.6 | 158.5 |

### 3.2. Rheological test results

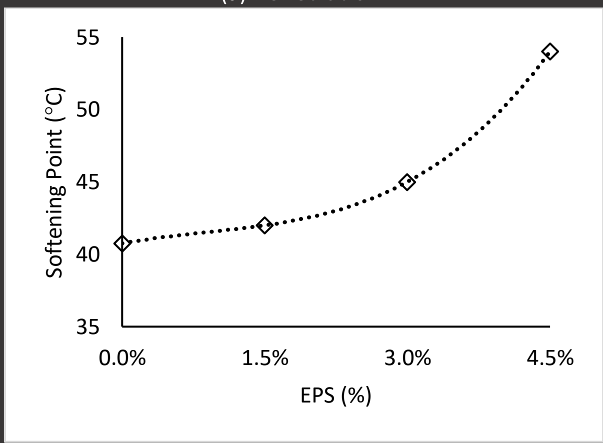
3.2.1. DSR test results

DSR tests were applied to unaged, short-term-aged (RTFO-aged) and long-term-aged (PAV-aged) asphalt specimens modified with different ratios of waste EPS. According to the Superpave binder grading standard [14], DSR test results include "G*/sinδ" (the complex shear modulus elastic portion) values at various high temperatures which indicate the rutting resistance of the tested unaged and RTFO-aged modified asphalt binder specimens; moreover, they also include "G*sinδ" (the complex shear modulus viscous portion) values at various medium temperatures which indicate the fatigue resistance of the tested PAV-aged asphalt binder specimens. Among these values, "G*/sinδ" determines the failure temperature value of the modified binder samples at high temperatures, while "G*sinδ" determines the failure temperature value at medium temperature.

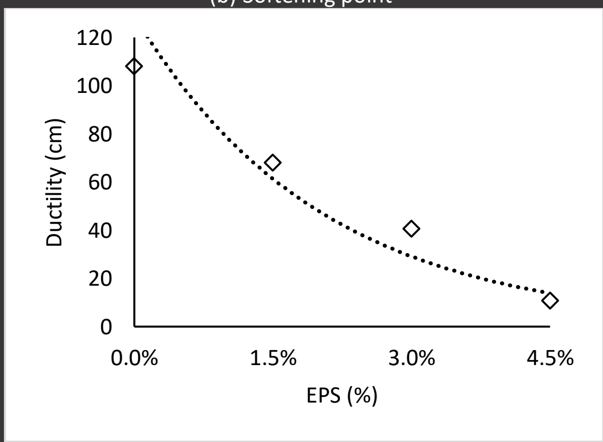
Figures 4 and 5 show DSR test results of the unaged and RTFO-aged modified asphalt binder specimens with four different EPS contents at various high temperatures, respectively. At high temperatures, asphalt binders should be stiff enough to be able to not deform too much to resist rutting and elastic enough to return to their original shape after loading. Therefore, Superpave binder specification defines a minimum value for G*/sinδ (1 kPa for unaged, and 2.2 kPa for RTFO-aged specimens, respectively). As asphalt binders age, they become stiffer and less elastic. Therefore, for the same test temperature, RTFO-aged asphalt binder samples resulted in higher G*/sinδ values compared to the unaged samples, as expected (Figures 4 and 5).




(a) Penetration




(b) Softening point




(c) Ductility

Figure 2. (a) Penetration, (b) Softening point and (c) Ductility test results of the asphalt binder specimens modified with four different ratios of waste EPS

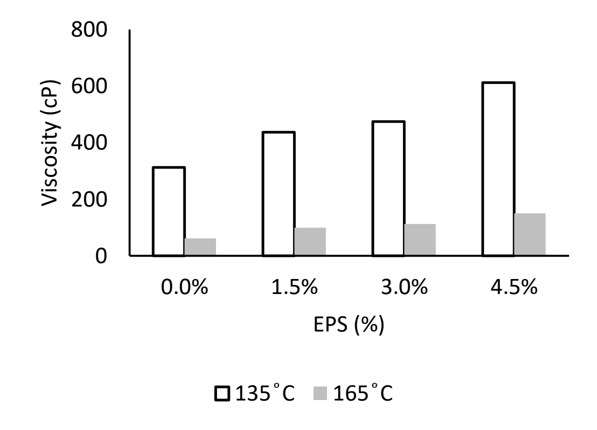


Figure 3. Viscosity test results of the asphalt binder specimens modified with four different ratios of waste EPS

Based on the DSR test results of the unaged modified asphalt binder specimens (Figure 4), it was determined that the allowable temperature values against rutting resistance increased in the unaged samples as the EPS additive ratio increased. Accordingly, while the allowable temperature of the base binder was found to be 58 °C, the allowable temperature value of the modified asphalt binder with 1.5% EPS content was found to be 64 °C. Furthermore, allowable temperature for the modified the asphalt binder with 3% EPS content was found to be 70 °C while allowable temperature for the modified the asphalt binder with 4.5% EPS content was found to be 76 °C. Accordingly, performance grade (PG) of the modified asphalt binders in terms of high temperatures increased as their EPS contents increased. Having higher allowable temperatures with the increase in EPS contents means that addition of EPS positively affected rutting performance of modified asphalt binders, making them stiffer at high temperatures.

Similar to the unaged binder cases, as EPS contents of the RTFO-aged modified asphalt binders increase, their allowable high temperature values also mostly increased (Figure 5). As can be seen in Figure 5, the allowable temperatures of the base and the modified binder with 1.5% EPS were found to be 64 °C whereas the allowable temperature values of the modified asphalt binder with 3% and 4.5% EPS contents were found to be 70 °C and 76 °C, respectively.

Based on the DSR results of PAV-aged modified asphalt binders, as EPS contents increased, G*sinδ values also increased for the same middle temperatures cases, implying that adding EPS to the asphalt binder specimens negatively affected fatigue resistance of the modified asphalt binder specimens at middle temperatures, making them more brittle and less flexible (Figure 6).

Overall, based on the DSR test results of the unaged, RTFO-aged and PAV-aged modified asphalt binder specimens, it was observed that as EPS contents of the modified asphalt binders increased, their rutting resistance increased but their fatigue resistance decreased.

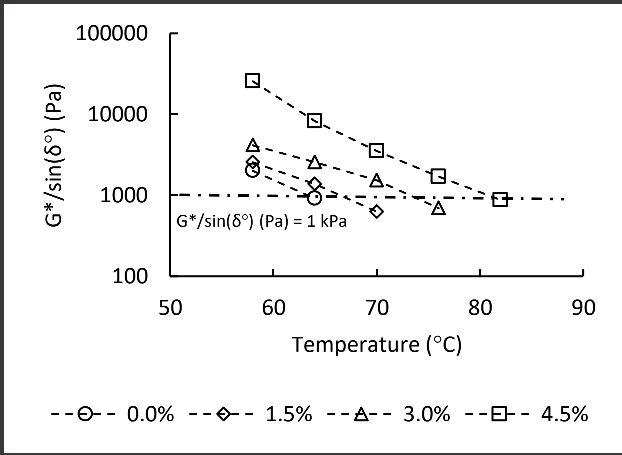


Figure 4. DSR test results of the unaged asphalt binder specimens modified with four different ratios of waste EPS

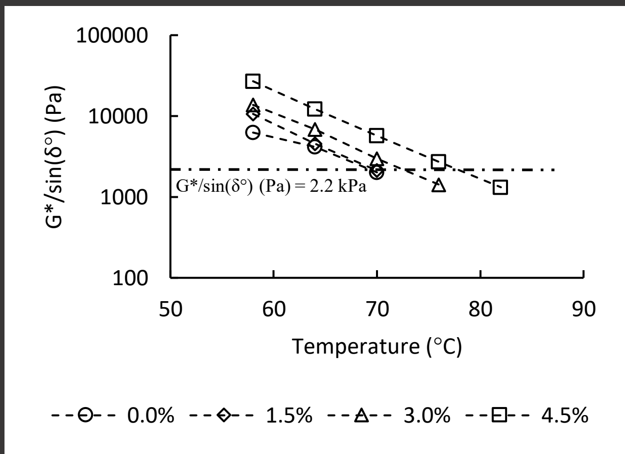


Figure 5. DSR test results of the RTFO-aged asphalt binder specimens modified with four different ratios of waste EPS

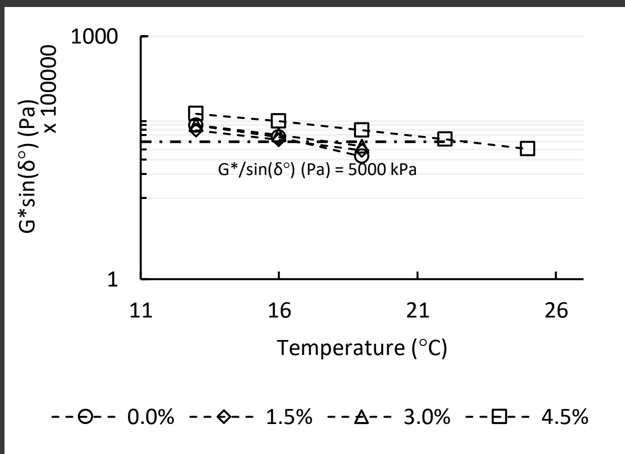


Figure 6. DSR test results of the PAV-aged asphalt binder specimens modified with four different ratios of waste EPS

### 3.2.2. BBR test results

BBR tests were applied to long-term-aged (PAV-aged) asphalt specimens modified with different ratios of waste EPS. According to the Superpave binder grading standard [14], BBR test results include creep stiffness (S) and the slope of the master stiffness curve at 60 seconds (m-value) values of the modified asphalt binders at various low temperatures which indicate their thermal cracking resistance at those low temperatures. Since a higher creep stiffness value means higher thermal stresses, Superpave binder grading standard [14] specifies a maximum creep stiffness value of 300 MPa. Moreover, a lower m-value means a lesser ability to relax stresses; therefore, a minimum m-value of 0.300 is specified.

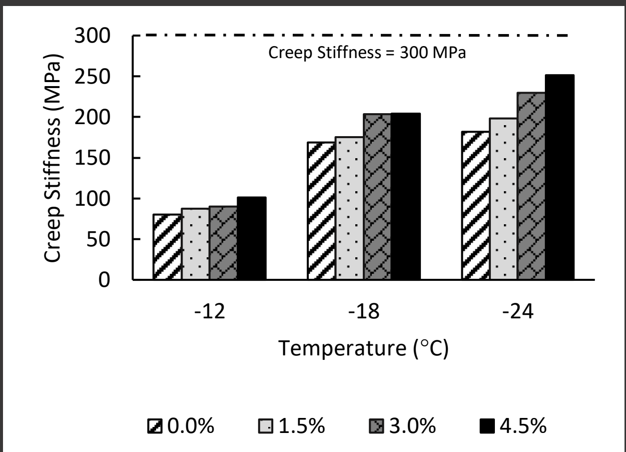



Figure 7. Creep stiffness results of the PAV-aged asphalt binder specimens modified with four different ratios of waste EPS

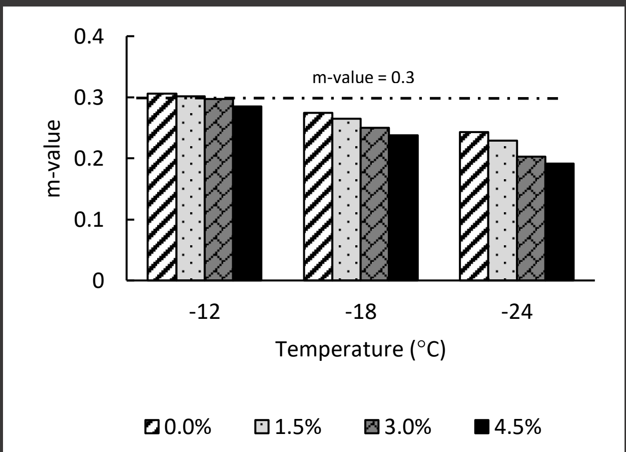



Figure 8. M-value results of the PAV-aged asphalt binder specimens modified with four different ratios of waste EPS

Figures 7 and 8 show creep stiffness and m-value results of the modified asphalt binder specimens with four different EPS contents at -12 °C, -18 °C and -24 °C test temperatures. Based on the test results, as EPS contents of the modified asphalt binder specimens increase their creep stiffness values increase and their m-values decrease. The increase in creep stiffness and decrease in m-values indicate that the modified asphalt binders exhibit a more brittle behavior at low temperatures and their resistance to thermal cracking decreases. Based on Figures 7 and 8, it was observed that all tested modified binder samples were below the creep stiffness limit of 300 MPa whereas only the base asphalt binder and the modified asphalt binder with 1.5% EPS content at -12 °C satisfied the minimum m-value limit of 0.3. Accordingly, while the allowable temperature values of the base binder and the modified asphalt binder with 1.5% EPS content were found to be -22 °C, the allowable temperature values of the modified asphalt binders with 3% and 4.5% EPS contents were found to be higher than -22 °C.

## 4. Conclusions

This study experimentally investigated the potential use of EPS waste in asphalt binder modification. As part of this study, a base asphalt binder with a Penetration grade of 70/100 was modified with waste EPS in four different ratios (0%, 1.5%, 3% and 4.5%, by weight of asphalt binder). The modified asphalt binder specimens were tested based on several physical (penetration, ductility, softening points and viscosity) as well as rheological/Superpave binder grading system tests (DSR, BBR, RTFO and PAV). The following conclusions could be drawn from this study:

- As waste EPS contents in the modified asphalt specimens increased, their penetration and ductility values decreased, and softening point values increased.
- As waste EPS contents in the modified asphalt specimens increased, their viscosity test results also increased in both temperature cases (135°C and 165°C). For the case of using 4.5% EPS in the modified asphalt binders, viscosity values increased by 96% and 140% at 135°C and 165°C, respectively. Furthermore, as EPS contents in the modified asphalt binders increased, corresponding mixing and compaction temperatures also increased. Increase in mixing and compaction temperatures might require higher fuel cost to bring asphalt binders to mixing and compaction temperatures as well as causing additional environmental problems due to higher levels of greenhouse gases emitted during additional heating.
- Based on all of the physical test results, it could be concluded that as waste EPS contents in the modified asphalt binder specimens increased, they became stiffer and their resistance against rutting tended to increase. On the other hand, having stiffer and less-flexible asphalt binders tend to have lower resistance against fatigue cracking.
- Based on the DSR test results of the unaged, RTFO-aged and PAV-aged modified asphalt binder specimens, it was observed that as EPS contents of the modified asphalt binders increased, their rutting resistance increased but their fatigue resistance decreased.
- Based on the BBR test results, as EPS contents of the modified asphalt binder specimens increased, their creep stiffness values also increased but their m-values decreased. The increase in creep stiffness and decrease in m-values indicate that the asphalt binders exhibit a more brittle behavior at low

temperatures and their resistance to thermal crack decrease.

- In terms of PG of the base and modified asphalt binders, PG of the base binder was found to be 58-22, while PG of the modified asphalt binder with 1.5% EPS content was found to be 64-22. Furthermore, allowable high temperature for the modified the asphalt binder with 3% EPS content was found to be 70 °C while allowable temperature for the modified the asphalt binder with 4.5% EPS content was found to be 76 °C. PG of the modified asphalt binders in terms of high temperatures increased as their EPS contents increased. On the other hand, the allowable low temperature values of the modified asphalt binders with 3% and 4.5% EPS contents were found to be higher than -22 °C. Accordingly, as EPS contents of the modified asphalt binder specimens increase, the modified asphalt binders exhibit a more brittle behavior at low temperatures and their resistance to thermal cracking decreases and their low temperature PG values increase.

Considering the findings of this study, it could be stated that modifying asphalt binders with EPS provides better rutting resistance at the expense of lowering low temperature and fatigue cracking resistance. Overall, it was concluded that waste EPS can be potentially used in the asphalt binder modification by paying special attention to these factors, and it should be noted that using waste EPS in the asphalt binder modification has many economic and environmental benefits.

## Declaration of Interest Statement

The author declares that he has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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