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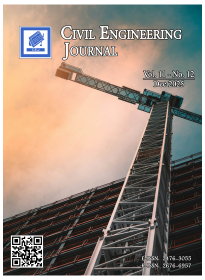
Civil Engineering Journal

(E-ISSN: 2476-3055; ISSN: 2676-6957)

Vol. 11, No. 12, December, 2025



Performance Characterization for Polymer Modified Bitumen Contained Newly Used Terpolymer

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Received 20 April 2025; Revised 27 October 2025; Accepted 11 November 2025; Published 01 December 2025

Abstract

Polymer-modified bitumen (PMB) plays a vital role in extending the service life of hot mix asphalt (HMA) used in flexible pavement construction. Several types of polymers have been used to produce PMB, among which styrene–butadiene–styrene (SBS) is the most widely used. However, the use of SBS in PMB production presents several limitations, including storage stability issues, high mixing temperatures, and the requirement for a relatively high modifier content. The present research investigated the use of a new terpolymer, EVA-GMA (LOTADER® AX8670T), for PMB production and compared the resulting PMB with PMB produced using 4% SBS polymer. Rheological, performance, and chemical composition tests were conducted on neat bitumen as well as PMB modified with EVA-GMA and SBS. The results indicated that the optimal LOTADER® AX8670T content required to produce PMB was 2.5%. In addition, storage stability increased by 11% compared to 4% SBS-modified PMB. The viscosity was found to be 50% higher than that of asphalt modified with 4% SBS-PMB and 100% higher than that of unmodified asphalt. The performance grade (PG) was determined to be PG 82-10 for both PMB types, while unmodified bitumen exhibited a PG of 76-10. Based on these results, it can be concluded that PMB produced with LOTADER® AX8670T can perform comparably to SBS-modified PMB while requiring a lower modifier content, lower mixing temperatures, and offering improved storage stability, thereby enhancing economic, production, and environmental aspects.

Keywords: Dynamic Shear Rheometer (DSR); Polymer Modified Bitumen (PMB); Performance Grade (PG); Rheological Properties; Thermoplastic Elastomer (TPE).

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1. Introduction

Bitumen is a traditional construction material that is derived from crude oil by distillation and has exceptional durability and flexibility. Road construction accounts for the majority of the world's asphalt usage [1]. The refining processes and the origins of crude oil closely relate to the properties of asphalt. However, the limited availability of raw materials and the technological limitations of refineries have impeded the production of high-quality asphalt, impacting both industries. They've shifted their focus to bitumen modification. Moreover, conventional pavements have proven inadequate over the past four decades due to a significant increase in traffic volume and stress, which has led to decreased durability and higher maintenance costs. The objective is to enhance the adhesion, durability, and efficiency of asphalt materials. Polymer modification has been thoroughly investigated as one of the most extensively studied approaches to modification [1-4]. The variable weather conditions and heavy traffic offer significant challenges for conventional asphalt surfaces. These pavements may be prone to rutting, cracking, potholes, and several other forms of damage. As a result, they often experience permanent deformation and fail to resist the demands of heavy traffic [5, 6]. To

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http://dx.doi.org/10.28991/CEJ-2025-011-12-022

successfully increase the durability of asphalt pavement and reduce the occurrence of the listed problems, it has become necessary to enhance the neat asphalt binder by making modifications that result in better performance [7, 8].

Plastomers, elastomers, and rubbers are the three main categories of polymers used in the modification of bitumen [9]. Every category of polymers has unique advantages and disadvantages when used as modifying agents for asphalt binders. Numerous instances demonstrate that plastomers provide substantial improvements in high-temperature properties, aging resistance, storage stability, and rutting resistance. They exhibit compatibility with asphalt matrices, show little changes in viscosity, possess thermal stability, and are easily accessible at a comparatively cheap price [7, 10]. The modification of elastomers, however, often affects the durability of asphalt pavement [11]. Conversely, the use of crumb rubber enhances the tensile strength of asphalt pavement and its resistance to rutting. This modifier is prone to oxidation and degradation [12-14]. Many factors, such as chemical structure, atomic weight distribution, and crystallization, influence the modification process [15]. The original asphalt grade used in binders is a crucial factor influencing the effectiveness of PMBs. The initial bitumen grade significantly influences the mechanical performance of the combinations [16].

The characteristics of PMB vary significantly based on the quantity of polymer included in the asphalt matrix. PMB is categorized into three kinds based on polymer content: low (less than 4 wt.%), medium (4–7 wt.%), and high (more than 7 wt.%). Incorporating small amounts of polymer into asphalt ingredients may expand the operational temperature range [17]. Excessive polymer quantities adversely affect asphalt, reducing the binder's ability to penetrate, soften, and elongate [18]. For PMB to perform effectively in the field, the compatibility between the polymer and asphalt is essential. The compatibility prevents the polymer from aggregating within the asphalt matrix [19]. The polymers in the asphalt matrix exhibit three levels of compatibility: low (incompatible), moderate, and high. The polymer and asphalt must exhibit sufficient compatibility to prevent separation within the asphalt, ensuring that the pavement meets acceptable quality standards. Asphalt may experience separation during storage, pumping, and application processes [17]. PMBs exhibit the susceptibility to property of material degradation when subjected to storage or transportation at temperatures that exceed 160°C. The issue of storage stability in SBS-modified asphalt is attributed to the low compatibility between asphalt and the high amount of polymer characteristic of SBS modification [20]. During the preparation and transportation of asphalt, it undergoes thermo-oxidative aging. The process affects the molecular bond of the SBS polymer, resulting in decreased stability when exposed to heat and oxygen [1]. Research indicates that extended storage durations at high temperatures result in the development of isolated agglomerates from uniformly dispersed SBS [21]. The observed morphological changes of SBS-modified asphalt during one hour of storage at 180°C confirm this assertion [22].

A summary of the literature demonstrates significant research focused on PMB aimed at improving the durability of hot mix asphalt. SBS is the most common type of modifier employed. Recent studies have identified several issues with SBS, including problems related to storage stability, the necessity for higher mixing temperatures, and the requirement for a substantial quantity of the modifier to accomplish optimal performance. Additional studies investigate different materials, including bio-based polymers, and assess the effects of additives, such as softening agents and fibers, on aging and performance. The existing literature shows a significant gap in research regarding the effectiveness of the terpolymer ethylene-vinyl acetate-glycidyl methacrylate (EVA-GMA) in modifying bitumen. LOTADER® AX8670T refers to this specific terpolymer [23-26].

The current research tries to address and identify the limitations of fatigue assessment methods in literature. Studies on high-viscosity modified binders suggest that conventional fatigue tests, including the Linear Amplitude Sweep (LAS) test, may inadequately represent these materials, as the pseudo-strain energy peak remains undetectable even at higher shear strain levels. This finding highlights the need for new or updated failure criteria to predict the fatigue life of significantly different binders accurately [27]. This study employs a thorough methodology to evaluate the effectiveness of the novel terpolymer EVA-GMA in enhancing bitumen performance. The methodology utilizes advanced performance testing together with traditional rheological assessments, such as the Dynamic Shear Rheometer (DSR) and the Bending Beam Rheometer (BBR). This approach aims to enhance the evaluation of the terpolymer's fatigue properties, exceeding conventional testing methods that may not adequately assess significantly modified binders. The research will evaluate the binder's performance and its capacity for recovering elasticity following both short- and long-term aging. Aging significantly influences material performance and the effectiveness of polymer modification. This comprehensive approach offers a complete understanding of the effectiveness of the EVA-GMA terpolymer in bitumen. The results likely provide useful information to facilitate the production of stronger and more efficient paving materials [25, 28].

EVA-GMA offers benefits over traditional EVA-modified bitumen due to its incorporation of glycidyl methacrylate (GMA). This material with this chemical composition is very limited investigated in literature, which has superior performance in improving storage stability. Traditional EVA and bitumen exhibit restricted compatibility, leading to poor storage stability. GMA in EVA-GMA acts as a reactive monomer, attractive in chemical interactions with polar elements of bitumen. This results in a more uniform and stable network, reducing phase separation during storage and promoting longer-lasting and strong modification [29, 30]. EVA-modified bitumen is susceptible to aging, particularly under high temperatures, due to chemical modifications. The GMA component creates a more resilient polymer network, exhibiting greater resistance to thermal degradation and chemical modifications. This network maintains the asphalt's superior properties, such as heightened stiffness and cracking resistance, for extended periods. However, EVA-PMB performance in elastic recovery and fatigue cracking resistance is miner to other modifiers like SBS. The permanent chemical network established by the GMA group significantly improves the bitumen's elasticity and recovery characteristics, enhancing its resilience to repeated loading and unloading. This chemical modification offers a more durable solution than physical modification [31].

The study aimed to investigate the rheological properties of asphalt binder modified with a terpolymer called EVA-GMA, also known as LOTADER® AX8670T, which is thermoplastic elastomer (TPE). The research involved using five different percentages (1.5, 2, 2.5, 3, and 3.5%) of LOTADER® AX8670T by weight of asphalt binder to produce polymer-modified bitumen (PMB). The study involved three phases: sample preparation, aging considerations for normal asphalt and PMB, and determining the performance grade (PG) for bitumen with LOTADER® AX8670T terpolymer. The study procedure was designed to evaluate the performance and characteristics of a modified asphalt binder that included various amounts of LOTADER® AX8670T and SBS polymers. The main goals of the research are to examine the chemistry by applying Fourier transform infrared spectroscopy (FTIR), measure the viscosity according to ASTM D4402, and ascertain the storage stability by means of the polymer's tendency to separate from PMB. Both short-term and long-term aging factors are addressed by experiments that simulate in-service aging conditions: rolling thin film oven (RTFO) and pressurized aging vessel (PAV), respectively. The research also aims to examine the elastic recovery skills, low-temperature performance, and dynamic rheological characteristics utilizing the BBR test, AASHTO T301 test, and DSR test, respectively. To ensure the modified bitumen is successful and suitable for real-world applications in road building and maintenance, the study aims to analyze these features in order to define its performance grade.

2. Materials and Methods

Figure 1 shows the flowchart of the research methodology through which the objectives of this study were achieved.

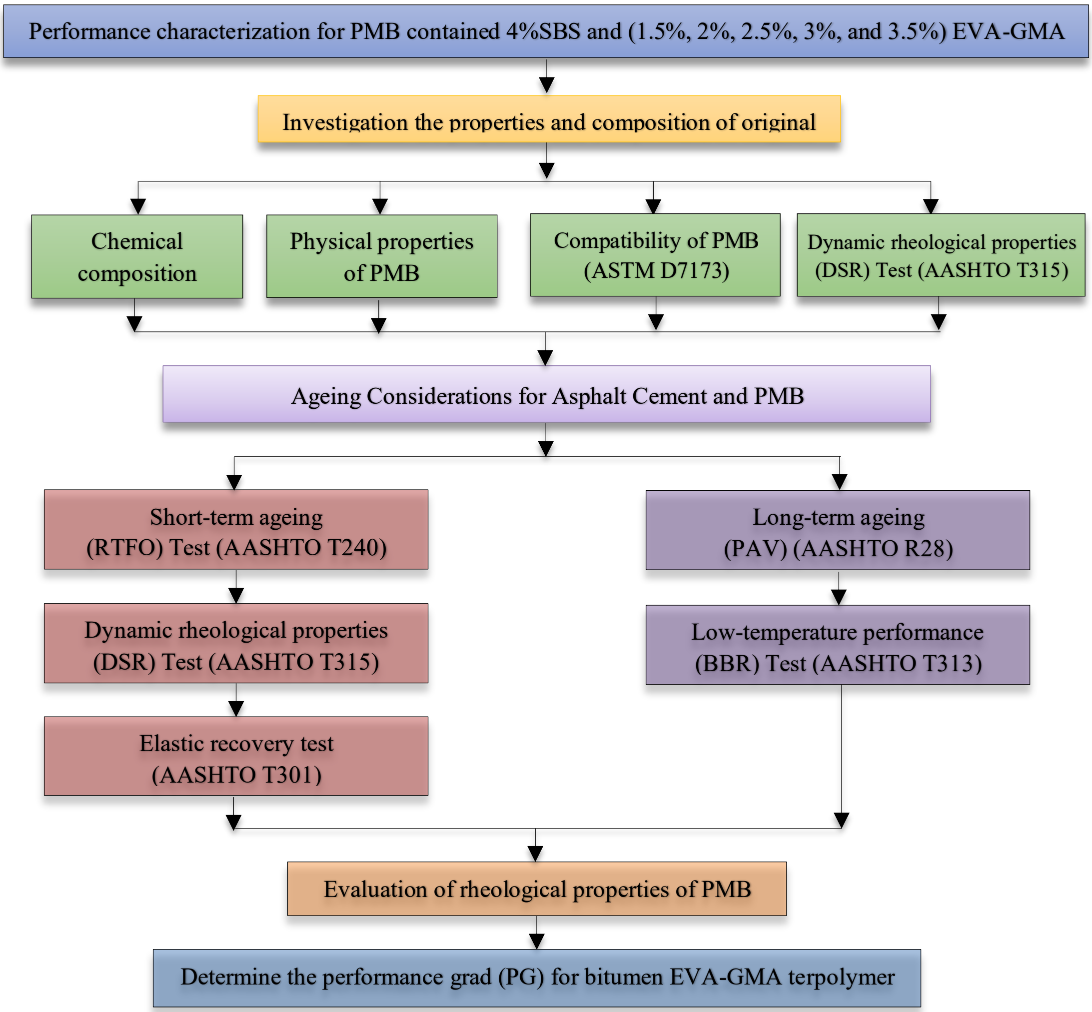


Figure 1. Research Flowchart

2.1. Asphalt Cement

The asphalt cement type used with penetration grade (40-50). The physical properties and tests of the used asphalt cement are shown in Table 1.

Table 1. physical properties and tests of the used asphalt cement

Property	Test method	Test Results	SCRB Specifications	AASHTO Specifications
Penetration (0.1 mm)	ASTM D 5	42	40-50	--------
Ductility (cm)	ASTMD 113	104	>100	--------
Flash point (^{O}C)	AASHTOT 48	315	Min.232	Min.230
Softening point (^{O}C)	AASHTOT 53	53	Min.65	
RV @ 135 °C, Pa. Sec	AASHTOT 316	0.762	---------	Max.3
RV @ 165 °C, Pa. Sec	AASHTOT 316	0.175	---------	Max.3
%Solubility in trichloroethylene	AASHTOT 44	99.69	Min. 99%	--------
Separation tendency difference between softening points, C	ASTMD 7173	1.21	Max. 4	--------

2.2. Polymers

This research used two types of polymers to improve the properties and performance of bitumen binder under different traffic loads and environmental conditions. The first type, LOTADER® AX8670T, shown in Figure 2-a, is an ethylene vinyl acetate-glycidyl methacrylate terpolymer (EVA-GMA), which increases tensile strength and flexibility while reducing HMA's temperature sensitivity. It provides elasticity and good properties at low temperatures. The second type, KUMHO KTR®103, shown in Figure 2-b, is a linear block copolymer (S-B-S) composed of styrene and butadiene, with a 31.5% styrene content by mass. It enhances flexibility, impact characteristics, and wear resistance and is easily processed. The chemical composition of each type was determined through an AT-FTIR test. The research provides a beneficial understanding of the potential of these polymers to improve asphalt binder performance.

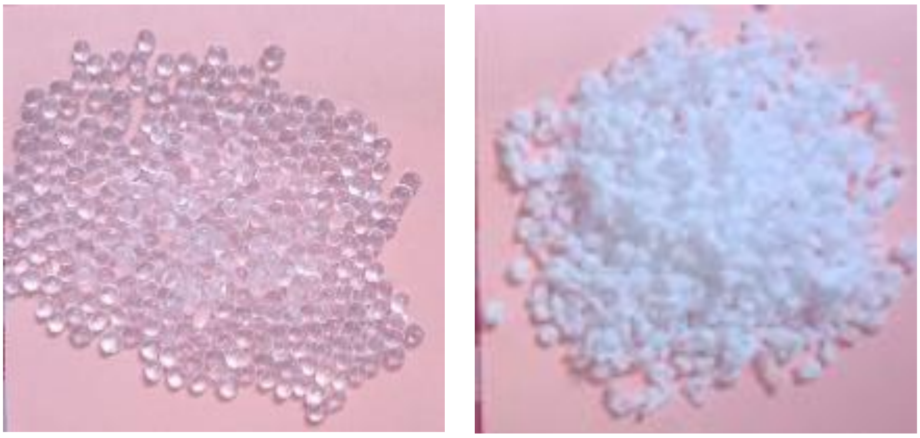

(a) (b)

Figure 2. Polymers used in asphalt modification: (a) EVA-GMA, (b) SBS

2.3. Preparation of Polymer Modified Bitumen (PMB)

The research focuses on the production of PMB using SBS and EVA-GMA terpolymer-modified bitumen. The mixture consists of 4% SBS [32] and varying amounts of EVA-GMA, specifically 1.5%, 2%, 2.5%, 3%, and 3.5%. The mixing process for PMB involves two techniques, influenced by the chemical composition of the polymers used, specifically SBS and EVA-GMA.

To mix SBS with pure bitumen, it should be heated in an oven at 160°C for an hour, then added to bitumen gradually in a high-shear mixer (5000 r/min.), raising the asphalt temperature up to 180°C. The mixture is blended with a high-shear mixer for one hour. After a four-hour curing process at 160°C in an oven, the mixture is returned to the mixer and mixed for an hour while a 180°C temperature is kept. The second type of polymer (EVA-GMA) was mixed with the neat bitumen by heating the bitumen in an oven at 160°C for one hour. Then, the neat bitumen is mixed in a high-shear mixer with the slow addition of EVA-GMA polymer to avoid agglomerations. This polymer is added with 1.5%, 2%, 2.5%, 3%, and 3.5% of the weight of neat bitumen. The mixing temperature of asphalt and polymer is gradually raised up to 165°C. Ultimately, the mixture was blended using a high-shear mixer operating at a velocity of 5000 r/min at a temperature of 165°C for a duration of two hours. Figure 3 shows sample preparation for the mixing process.

Figure 3. Sample preparation and mixing of PMB

The experimental mixing operations revealed that the EVA-GMA polymer had better mixability than the SBS polymer. The mixing process took less than two hours and required a temperature lower than 165°C. Using EVA-GMA in asphalt mixture production could lead to increased energy efficiency and reduced environmental pollution, as well as accelerate the road paving process.

2.4. Ageing Considerations for Asphalt Cement and PMB

The standard was developed to evaluate asphalt binders' typical qualities at the point in service where specific types of distress are most likely to occur. So, the original binder and RTFOT residue are used to measure the quality characteristics related to rutting. The pressure egging vessel (PAV) residue is then used to measure the quality characteristics related to fatigue cracking and thermal cracking [33]. RTFO test is a standard method for assessing the impact of heat and air on asphalt binders (ASTM D2872 and AASHTO T240). It simulates the aging process for HMA by conditioning asphalt binders, subjecting the binder to high temperatures and circulating air. The test measures mass change and provides a numerical evaluation of volatile substances depleted during the aging process. Figure 4 shows sample preparation for RTFO test.

Figure 4. Sample preparation for RTFO test

The Pressure Aging Vessel (PAV) test can simulate the rheological changes that occur in asphalt binders during in-service oxidative aging, but it might not be a perfect representation of the relative aging rates. This test is described in ASTM D6521 and AASHTO M320, "Standard Practice for Accelerated Aging of Asphalt Binder Using a Pressurized Aging Vessel (PAV)." For the purpose of assessing physical properties, PAV offers a long-term aged asphalt binder simulation. Figure 5 shows the PAV test sample preparation.

Figure 5. Sample preparation for PAV test

2.5. Test Methods

The bitumen's functional groups were determined using an attenuated total reflection (ATR) Bruker Alpha II FTIR spectrometer. Droplets of hot bitumen were placed on pieces of butter paper and left to sit in a dark, sealed container for 10 minutes. The samples were tested no later than one hour after their preparation. Room temperature was used for the spectra recording. Using OPUS software, the spectra were examined with a resolution of 4 cm^{-1} and 32 scans, covering an absorption wavenumber range of 400-4000 cm^{-1}. We scanned each sample three times, and we scanned three samples per binder. In all, sixty-three spectra were captured. The min-max approach was used to standardize the spectra. Figure 6 shows the (ATR) Bruker Alpha II FTIR spectrometer test samples.

Figure 6. Preparation for FTIR spectrometer test samples

The ASTM D7173 test measures the separation tendency of polymers from PMB under static heated storage conditions. The test reveals significant differences between the upper and lower sections of the tube, suggesting incompatibility between the polymer and the underlying asphalt. Asphalt binder is a complex material; therefore, it requires a detailed investigation to define its features under relevant conditions. Conventional tests, such as consistency tests, are used to simplify this process. These tests are essential to define the different grades of asphalt cement. They serve as the foundation for asphalt cement standards, providing information on hardness or consistency but not details about viscoelastic responses. The evaluation of asphalt binder's workability and high-temperature performance depended on the measurement of viscosity. This is because asphalt flow behavior during pumping and mixing is of practical importance for ensuring the quality control of its manufacture, storage, transportation, and construction [34]. The Brookfield test is an essential method for evaluating the workability and performance of PMB during its entire service life. The test is conducted according to ASTM D4402 to compare between the two types of polymers used in this research (SBS and EVA-GMA) in terms of viscosity values. Figure 7 shows the Conventional tests of bitumen.

(a) (b) (c)

Figure 7. Conventional tests of bitumen: (a) rotational viscosity test, (b) separation tendency test, (c) elastic recovery test

The rheological properties of bitumen and PMB are crucial for asphalt pavement production and quality. Penetration, softening point, and viscosity have been mainly used to evaluate the rheological properties of bitumen since the last century. Recently, a new procedure is used to do that (see Figure 8). The procedure is mostly included within the steps of binder selection in the Superpave method. The dynamic shear rheometer (DSR) is a tool used to measure the viscous and elastic behavior of asphalt binders at medium to high temperatures according to AASHTO T315. It measures the complex shear modulus |G*| and phase angle (δ) of asphalt binders under dynamic shear using parallel plate geometry. Satisfactory elastic recovery can reduce high-temperature rutting and low-temperature cracking in asphalt pavement.

The AASHTO T301 standard test method evaluates asphalt material's elastic recovery by measuring recoverable strain. Low-temperature cracking in asphalt leads to fractures and collapses, resulting in significant costs for transportation departments and organizations worldwide. This defect occurs due to thermal susceptibility and increased thermal tensile stresses, potentially causing microcracks on pavement surfaces. The Bending Beam Rheometer (BBR) test is a standard method used to determine the flexural creep stiffness of asphalt binder according to AASHTO T313. This test offers information about the low-temperature stiffness and relaxation properties of asphalt binders. These characteristics provide an indication about asphalt binder's capacity to resist low-temperature cracking.

(a) (b) (c)

Figure 8. Rheological properties tests of bitumen: (a) DSR and BBR test samples, (b) DSR test device, (c) BBR test device

3. Results and Discussion

3.1. FTIR-ATR Results

SBS and EVA-GMA are two polymers that are used for modifying bitumen. Figure 9 shows an FTIR spectral examination of these two materials. Important factors in determining the polymers' performance and appropriateness for various asphalt applications are their chemical structures, which the spectral analysis shows to have substantial variances. The blue SBS spectrum mainly shows the absorption peaks that are associated with butadiene and styrene. The aromatic components' bending vibrations are matched by the sharp peak at 699 cm-1 and the butadiene units' double bonds by the larger peak around 1500 cm^{-1} [35]. These characteristics give validity to a structure that increases the material's resistance to deformation and fatigue by adding the high elasticity and resilience characteristic of SBS-PMB. The EVA-GMA spectrum, on the other hand, shows clear peaks at around 1730 cm^{-1}, which are carbonyl groups from vinyl acetate, shown in green [36]. The peaks at around 910 cm^{-1}, which are linked to epoxy groups, indicate that the polymer contains glycidyl methacrylate [37]. These categories are well-known for their capacity to increase asphalt's thermal stability and resistance to moisture by increasing adhesion and cross-linking within the polymer matrix. These distinctions serve to confirm the unique characteristics that each polymer has. When it comes to the pavement's structural stability and flexibility, SBS improves its material characteristics. At the same time, EVA-GMA changes the chemical relationships, which may make the asphalt more resistant to conditions like temperature changes and water penetration. By utilizing the chemical functionalities uncovered by FTIR analysis, asphalt formulations can be optimized to improve road performance and longevity.

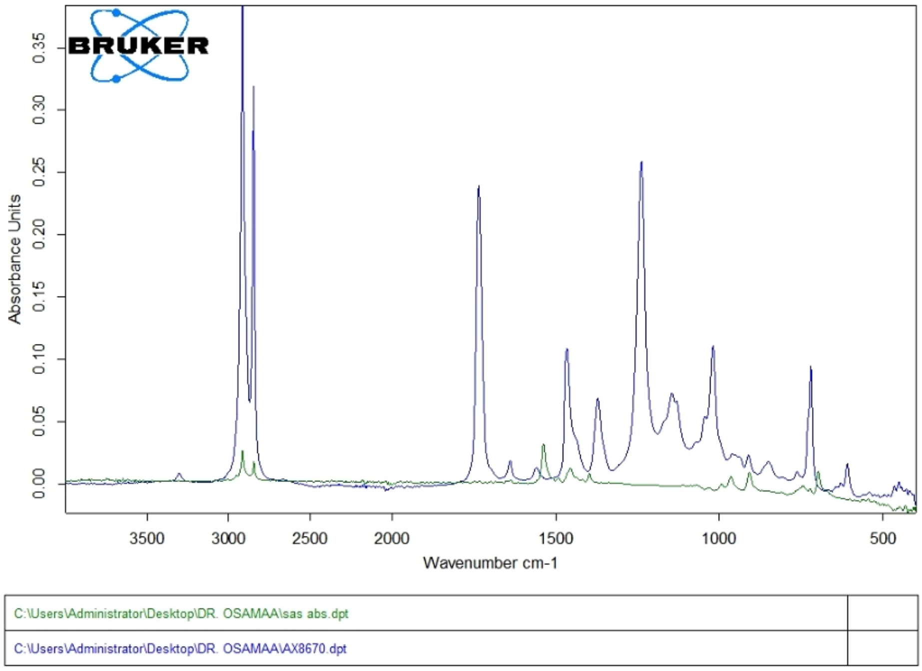


Figure 9. FTIR spectroscopic results for EVA-GMA and SBS

Asphalt is a complex organic material mostly composed of aliphatic and aromatic hydrocarbons. The control spectrum of unmodified asphalt is anticipated to have distinctive peaks corresponding to these components. The peaks at 2840 to 3000 cm^{-1} correspond to C-H stretching vibrations of aliphatic compounds, whereas the peaks at 1465 to 1380 cm^{-1} represent C-H bending vibrations of alkane and methylene groups [38]. This spectrum serves as a crucial baseline, since any new peaks or significant changes in the strength of existing peaks in the modified asphalt spectra are directly attributable to the additional polymer.

The red curve in Figure 10 illustrates the Fourier Transform Infrared (FTIR) spectrum of the unmodified asphalt binder (control). This spectrum serves as an essential basis for analysis, since its peaks are distinctive to the particular substance and act as a chemical fingerprint. The spectrum has broad absorption bands, indicating that asphalt comprises a variety of hydrocarbons. Key characteristics involve 2800–3000 cm^{-1}: an extensive absorption band resulting from the C–H stretching vibrations of aliphatic groups.1465 cm^{-1} and 1380 cm^{-1}: Peaks indicative of C–H bending vibrations of methylene and methyl groups within alkane chains.1000–1100 cm^{-1}: Peaks indicating the presence of sulfoxide groups [39]. This distinctive fingerprint is necessary for any investigation of modifications. Analysis of the spectra of modified asphalt compared to the control shows any new peaks or significant changes in the intensity of existing peaks, indicating the incorporation of a polymer. Furthermore, natural asphalt peaks, such as the one at around 1450 cm^{-1}, can act as internal reference points for determining polymer content. This is a fundamental concept in quantitative spectroscopic analysis. In the absence of a reliable baseline, accurately identifying the polymer's spectral fingerprint and quantifying its concentration becomes challenging.

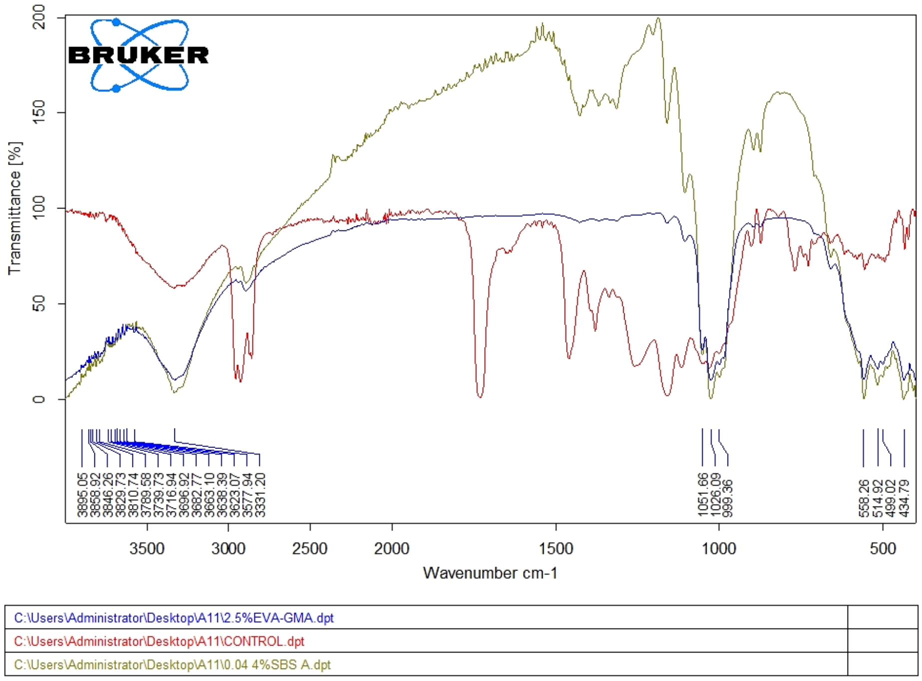


Figure 10. FTIR spectroscopic results for Net bitumen+4%SBS+Net bitumen+2.5%EVA-GMA+Net bitumen

The yellow curve displays the FTIR spectrum of SBS-PMB. In comparison to the control spectrum (red curve), two new and significant peak groups occur. These groups are significant indicators of the presence of the SBS polymer. A strong peak occurs at around 699 cm^{-1}, accompanied by a secondary, less significant peak at 757 cm^{-1}.The two peaks result from the out-of-plane bending vibrations of the monosubstituted benzene ring characteristic of the polystyrene blocks. A distinct, strong peak occurs at around 966 cm^{-1}.The bending vibrations of the C=C double bond in the trans-alkene structure of the polybutadiene blocks is responsible for this peak. The existence of these distinct SBS peaks presents qualitative evidence of the modification and facilitates quantitative analysis. Research indicates a significant linear correlation between the intensity ratio of the primary polymer peak, demonstrated by the polybutadiene peak at 966 cm^{-1}, and the concentration of the polymer in asphalt. The weight percentage of SBS can be accurately determined by utilizing an internal asphalt reference peak, specifically the C-H bending peak at 1375 cm^{-1}.Several studies have reported correlation coefficients (R^2) greater than 0.96, validating the effectiveness of this method and establishing FTIR as an effective and quick quality control instrument [40]. The presence of both SBS peaks and original asphalt peaks, along with the absence of new peaks indicative of a chemical reaction, reinforces the concept that the modification primarily results from a physical blending process. The polymer chains elongate and interact with the lighter components of asphalt (maltenes), resulting in a polymeric network that enhances the material's strength. The "physical blending" model explains the inadequate performance of SBS-PMB, regardless of temperature or age. The physical network may ultimately fail, resulting in a decreased intensity of the peak at 966 cm^{-1}.

The blue curve in Figure 10 illustrates the FTIR spectra of asphalt modified with EVA-GMA polymer. This spectrum significantly differs from the SBS spectrum, indicating the chemical differences of this modifier. The spectrum contains numerous significant peaks indicating the functional groups present in the terpolymer. A significant, strong peak is seen at about 1724 cm^{-1}. This peak is important for the C=O stretching vibration (carbonyl group) and is present in both the vinyl acetate and glycidyl methacrylate (GMA) segments of the terpolymer. The critical peak for identifying this polymer occurs at around 905 cm^{-1}. The unique absorption is completely due to the epoxy ring, an essential functional element of the GMA polymer. Supplementary Peaks: notable peaks include a C=C stretching vibration approximately at 1637 cm^{-1} and a C-O-C stretching vibration about 1160 cm^{-1}. Both phenomena are attributable to the GMA polymer.

The comparison of the three spectra shows obvious differences between the unmodified and modified asphalts. Control Spectrum (Red): This spectrum represents the chemical baseline of the pure material. The data exhibits distinct, characteristic peaks associated with hydrocarbons. SBS Spectrum (Yellow), exhibits sharp, distinct peaks in the lower wavenumber range, specifically at 699 cm^{-1} and 966 cm^{-1}, which serve as clear indicators of the polystyrene and polybutadiene blocks, respectively. The EVA-GMA spectrum (Blue) reveals a significant peak at 1724 cm^{-1}, indicative of a carbonyl group, with an important peak for the epoxy ring at 905 cm^{-1}. This spectral data allows for a direct conclusion of a fundamental difference in the modification mechanisms. The distinct peaks of styrene and butadiene in the SBS spectrum suggest a physical blending mechanism, wherein the polymer chains intertwine with the asphalt components to create a network that improves flexibility and recovery. The unique peak at 905 cm^{-1} in the EVA-GMA spectrum indicates a highly reactive functional group, suggesting that its modification require both physical mixing and a chemical reaction. The chemical interaction can result in a more stable, cross-linked network, thereby improving the asphalt binder's resistance to thermal aging and long-term moisture damage. This comparison demonstrates the importance of the spectral signature region (1500–600 cm^{-1}) in differentiating subtle differences among polymers and understanding the underlying modification mechanisms.

The spectrum analysis of the three asphalt samples indicates that FTIR spectroscopy effectively characterizes modified asphalt binders. The control spectrum provides a fundamental fingerprint for the material, whereas the modified spectra provide a distinct fingerprint for each polymer. The SBS-PMB spectra exhibit significant peaks at 699 cm^{-1} and 966 cm^{-1}, indicating the presence of styrene and butadiene groups. This suggests that the modification process mainly involves physical mixing. The EVA-GMA-PMB spectrum exhibits an important carbonyl peak at 1724 cm^{-1} and a notable epoxy ring peak at 905 cm^{-1}.This indicates the presence of a terpolymer and suggests a potential chemical interaction with the asphalt components.

3.2. Compatibility Test Results

The separation tendency data shown in Figure 11 demonstrates significant variations among the different PMBs, highlighting the impact of polymer types and percentages on blend compatibility and stability. The 2.5% EVA-GMA-PMB has a perfect separation tendency value of 0.8, which is lesser than the SBS-PMB (0.9) and significantly higher compared to both the unmodified bitumen (control) (1.2) and the excessively modified 3% EVA-GMA (1.4). The improved phase stability is due to the grafting effectiveness and molecular interactions between the EVA-GMA polymer and the bitumen matrix. The results of the dynamic rheological properties for neat bitumen and different types and ratios of polymers used to produce PMBs provide further support for these results: The 2.5% EVA-GMA-PMB exhibits a significant enhanced complex modulus and phase angle characteristics during the DSR test, indicating a strong and uniformly dispersed polymer network. The enhanced homogeneity is essential for field performance, as it provides uniform mechanical behavior throughout the storage, mixing, and paving processes. Moreover, it enhances long-term durability by reducing oxidative age gradients and specific binding sensitivities. The 2.5% EVA-GMA binder provides a well-balanced formulation that improves thermal storage stability while maintaining workability and performance characteristics. In contrast, higher EVA-GMA percentages (e.g., 3.5%) exhibit excessive stiffening and increased separation (separation tendency = 0.9) due to polymer overloading and potential agglomeration effects.

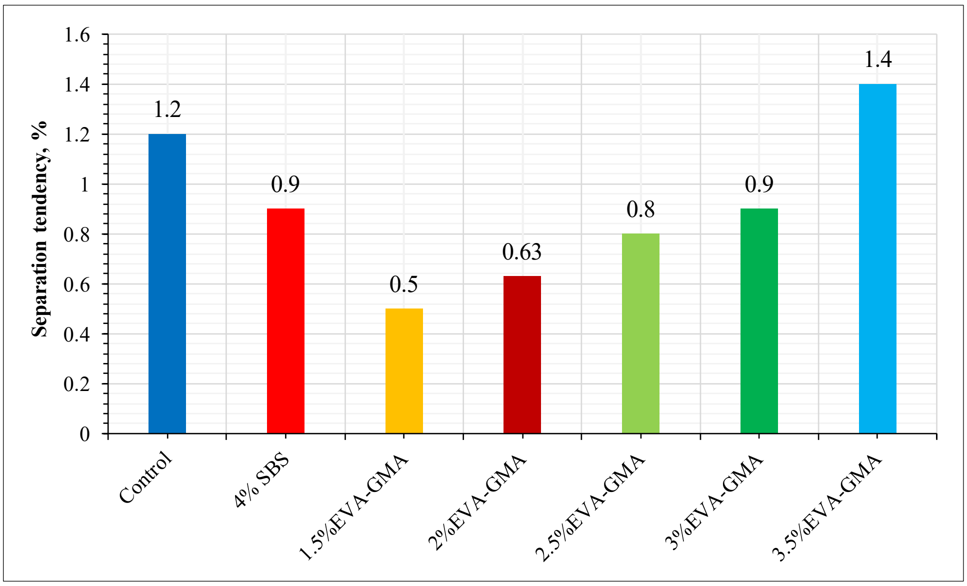


Figure 11. Separation tendency test results of all types of bitumen

Figure 12 shows the percentage solubility in trichloroethylene for several kinds of bitumen, including neat bitumen (control) and many PMBs. An increased solubility percentage signifies enhanced compatibility and dispersion of the polymer inside the bitumen matrix. The control bitumen has a solubility of 99.69%. Among the PMBs, 2.5% EVA-GMA-PMB demonstrates superior solubility in trichloroethylene (99.91%) relative to 4% SBS-PMB (99.81%) and other percentages of EVA-GMA. The increased solubility of 2.5% EVA-GMA indicates a more uniform and stable mixture, essential for improved field performance and increased pavement durability. The enhanced solubility of 2.5% EVA-GMA-PMB is due to the unique chemical characteristics of EVA-GMA and its interaction with bitumen components. EVA-GMA is a terpolymer that allows GMA to add epoxy functional groups. The epoxy groups interact with the polar parts of bitumen, including asphaltenes and resins, resulting in chemical bonding. This interaction improves the compatibility of the polymer with bitumen, avoiding phase separation and resulting in a more stable and soluble system. Conversely, SBS is a block copolymer that mostly links with bitumen by physical expansion and tangles, resulting in a network structure. Although effective, this physical association could fail to achieve the same degree of molecular-scale compatibility as the chemical interactions provided by EVA-GMA, resulting in the comparatively reduced solubility noticed for 4% SBS-PMB.

The Asphalt Performance Grade (PG) test results shown in Table 2 confirm the enhanced performance of EVA-GMA-PMBs, with 2.5% EVA-GMA-PMB attaining a PG of 82-10, similar to 4% SBS-PMB, which similarly is at 82-10, and 3% EVA-GMA-PMB achieving 88-10. This study demonstrates that both polymers significantly enhance the high-temperature performance of bitumen. The increased solubility, especially for 2.5% EVA-GMA, results in higher homogeneity and stability of the binder, which is essential for long-term pavement durability. A thoroughly solubilized polymer-bitumen mixture exhibits a decreased probability of phase separation during storage, handling, and the operational period, resulting in uniform pavement characteristics, enhanced resistance to aging, and reduced susceptibility to various distresses, such as rutting and cracking, thereby enhancing overall field performance and increasing the service life of asphalt pavements.

Table 2. Asphalt performance grade (PG) test Results

Property	Test method	Net bitumen (N)	N+4%SBS	N+1.5% EVA-GMA	N+2% EVA-GMA	N+2.5% EVA-GMA	N+3% EVA-GMA	N+3.5% EVA-GMA
Asphalt performance grade (PG)	AASHTO M 320	76-10	82-10	76-10	76-10	82-10	88-10	82-10

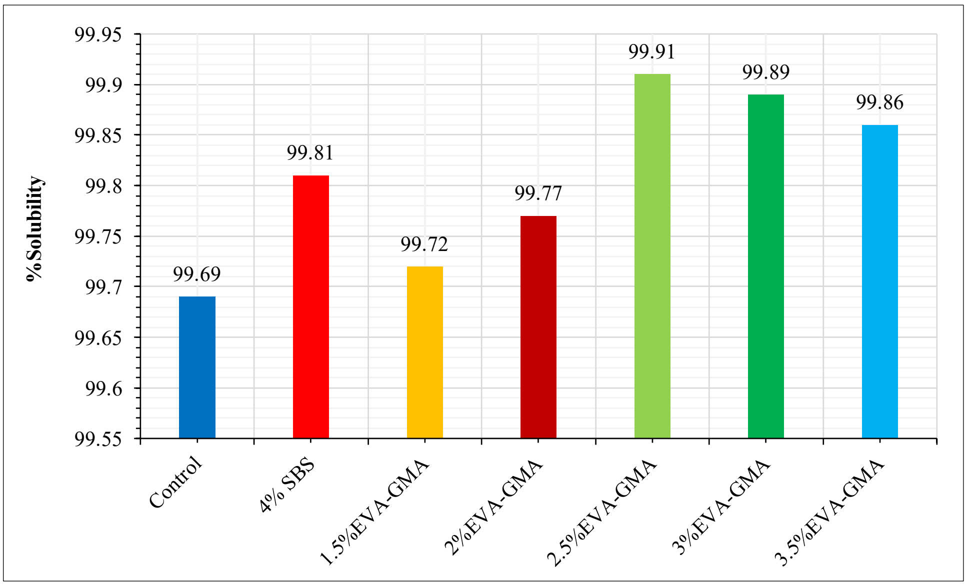


Figure 12. Relationship between %Solubility in trichloroethylene for all types of bitumen

The FTIR data indicate that the polymers and bitumen exhibit distinct separation and dissolution behaviors due to their chemical and physical interactions. The FTIR spectra of SBS-PMB exhibit distinct peaks at 965 cm^{-1} corresponding to polybutadiene and at 700 cm^{-1} associated with polystyrene. This indicates the presence of the polymer. The spectra of EVA-GMA-PMB exhibit a distinct C=O stretching peak at approximately 1740 cm^{-1}. This indicates that EVA-GMA has been appropriately applied. Changes in the O-H region at 3300 cm^{-1} indicate that chemical reactions are occurring. The epoxy group in GMA enhances the compatibility of components. The enhanced chemical bonding facilitates improved network functionality and cross-linking in bitumen, thereby reducing of phase separation. The EVA-GMA samples exhibit reduced separation, especially at lower concentrations, primarily establishing a physical network. The modified samples exhibit enhanced performance in conjunction, resulting in improved solubility in trichloroethylene.

The polymer components may exhibit improved solubility in the solvent due to their uniform distribution and chemical bonding. The increased tendency to separate and reduced solubility at high EVA-GMA concentrations (3.5%) indicate that polymers may agglomerate, resulting in reduced stability and solubility.

3.3. Results of Physical Properties of PMB

The use of EVA-GMA in asphalt binder mixing results in a significant change in its physical characteristics. The polymer enhances the binder's stiffness and flexibility, also increasing its viscosity. This improves the asphalt's resistance to flow under high temperatures. Figure 13 shows the results of the penetration test, providing a direct enhancement of binder consistency. The neat bitumen, which serves as the control, has the maximum penetration value of 42 (0.1 mm), indicating that it is the softest binder among all evaluated samples. Conversely, all PMBs consistently exhibit much lower penetration values than neat bitumen, indicating a distinct enhancement in hardness and stiffness due to polymer integration. The 4% SBS-PMB has a penetration of 36 (0.1 mm). The EVA-GMA-PMBs have a distinct pattern of reducing penetration as polymer content increases: 1.5% EVA-GMA (38), 2% EVA-GMA (35), 2.5% EVA-GMA (30), 3% EVA-GMA (26), and 3.5% EVA-GMA (23). The 2.5% EVA-GMA-PMB, with a penetration of 30 (0.1 mm), is much stiffer than the 4% SBS-PMB, which has a penetration of 36 (0.1 mm). Increased concentrations of EVA-GMA (3% and 3.5%) further enhance this stiffening action. All PMBs, except for 1.5% EVA-GMA, conform to or are below the standard SCRB requirement range of 40-50 (0.1 mm) for penetration, with 2.5% EVA-GMA exhibiting exceptional performance in this aspect.

The constant decrease in penetration values across all PMBs, particularly with increased EVA-GMA percentages, reflects an enhancement in binder hardness and stiffness. A reduced penetration value directly reflects a harder bitumen. In pavement engineering, a stiffer bitumen exhibits greater resistance to permanent deformation, referred to as rutting, especially at high temperatures and heavy traffic loads. This observed trend demonstrates that polymer modification successfully reduces one of the main failure mechanisms associated with conventional asphalt pavements. The content-dependent stiffening influence observed in EVA-GMA indicates that polymer concentration is an essential consideration for optimizing the binder's performance at higher temperatures.

The important result is that the 2.5% EVA-GMA-PMB achieves a lesser penetration (30) compared to the 4% SBS-PMB (36). The result demonstrates increased stiffness with reduced polymer content. Achieving enhanced stiffness with a decreased amount of polymer indicates that the reactive characteristics of the GMA component in EVA-GMA enhance more durable, probably chemical, interactions within the bitumen matrix. This chemical modification enables a more significant and effective stiffening per polymer unit, impacting both performance and sustainability. These results provide support to the idea that the chemical reactivity and strength of polymers have a significant impact on the final product of bitumen change [41].

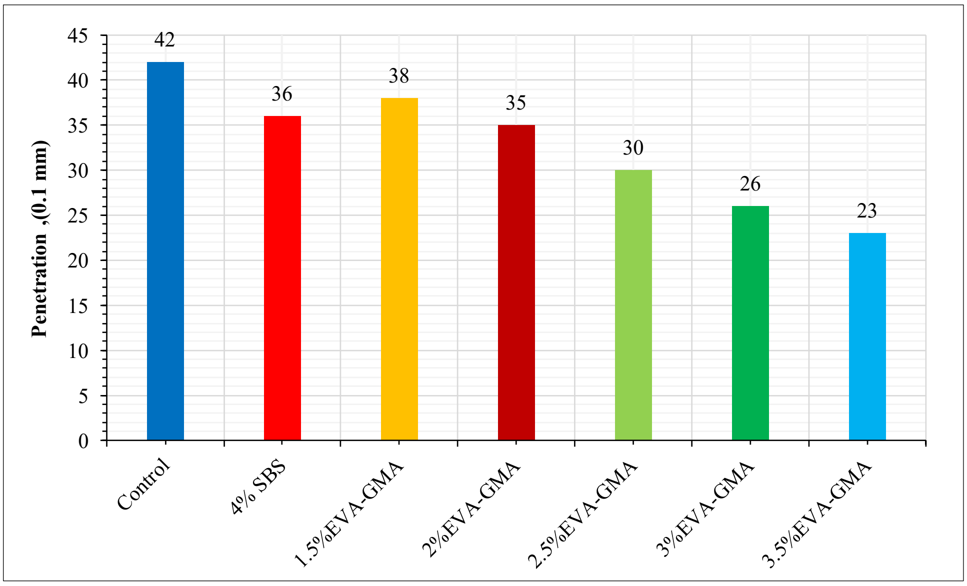


Figure 13. Penetration test results for all types of bitumen

Figure 14 shows the results of the softening point (SP) test, which indicates the temperature at which bitumen reaches a certain level of softness. The SP of the neat bitumen (control) is the lowest, measured at 56°C. All PMBs exhibit a much higher SP than neat bitumen, indicating superior performance at high temperatures. The 4% SBS-PMB has an SP of 70°C. The SP increases with the addition of polymer content in EVA-GMA-PMBs: 1.5% EVA-GMA (68), 2% EVA-GMA (68), 2.5% EVA-GMA (73), 3% EVA-GMA (77), and 3.5% EVA-GMA (75). The 2.5% EVA-GMA-PMB has a higher SP (73°C) compared to the 4% SBS-PMB (70°C). The SP significantly increases with higher amounts of EVA-GMA (3% and 3.5%). The results specify the SP requirements for SCRB; nonetheless, it is clear that all PMBs exceed neat bitumen. Polymer modifiers significantly increase the softening point (SP) of bitumen, making it less prone to deformation upon heating. This makes the material more effective in resisting rutting under hot climates and heavy traffic. The 2.5% EVA-GMA-PMB formulation has a higher SP (73°C) compared to the 4% SBS-PMB (70°C), indicating superior performance at high temperatures. The EVA-GMA polymer reduces the fundamental temperature sensitivity of bitumen, ensuring it remains stiff and structurally stable. The reduced penetration rates for 2.5% EVA-GMA consistently support this observation, demonstrating that the reactive polymer enhances the internal structure's strength and thermal resistance.

From the FTIR spectra shown in Figure 10, it can be observed that there are specific absorbance peaks that signify the presence and changes in functional groups due to the addition of EVA-GMA to the bitumen. Key peaks typically associated with EVA-GMA modifications include those for ester groups (1735 cm^{-1}), vinyl groups (around 1600 cm^{-1}), and the ether groups from the GMA component (1100 cm^{-1}). These absorbance changes are indicative of the chemical modifications within the bitumen structure. The GMA component in EVA-GMA offers glycidyl groups that can undergo reactions with the carboxylic acid groups in the bitumen or hydroxyl groups from other EVA chains, leading to cross-linking. This network formation significantly increases the molecular weight and, consequently, the viscosity. The increased peak intensities at regions associated with these functional groups corroborate the presence of more extensive cross-linking with higher EVA-GMA content.

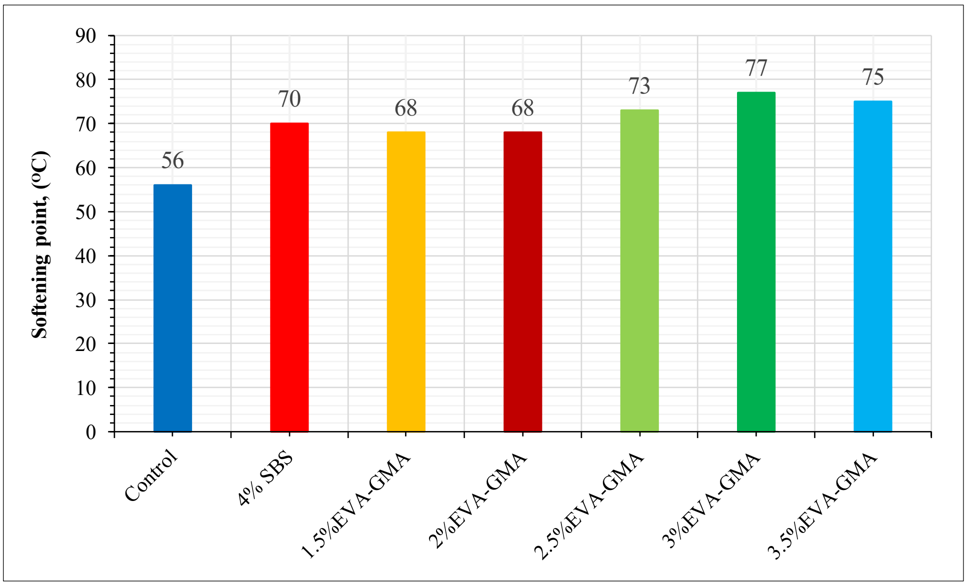


Figure 14. Softening Point test results for all types of bitumen

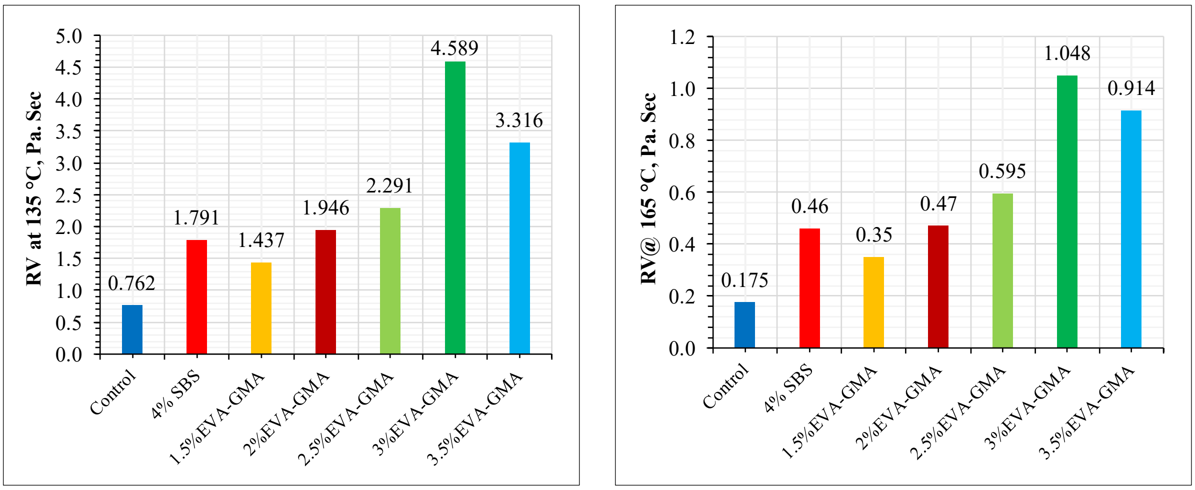
The results of rotational viscosity (RV), shown in Figure 15, measure the binder's flow resistance at high temperatures, which is essential for workability during mixing and compaction. At 135°C, neat bitumen exhibits the minimum RV of 0.762 Pa.sec. All PMBs exhibit a significant increase in viscosity relative to the unmodified bitumen (control). The 4% SBS-PMB has a viscosity of 1.79 Pa·sec, and EVA-GMA-PMBs exhibit a progressive increase in viscosity corresponding to increased polymer percentages: 1.5% EVA-GMA (1.437), 2% EVA-GMA (1.946), 2.5% EVA-GMA (2.291), 3% EVA-GMA (4.589), and 3.5% EVA-GMA (3.316). The 2.5% EVA-GMA-PMB, with a viscosity of 2.291 Pa.sec, demonstrates a greater viscosity compared to the 4% SBS-PMB (1.79 Pa.sec). All binders conform with the AASHTO M 320 standard, which sets a maximum viscosity of 3 Pa.sec, except for the 3% EVA-GMA binder, which measures 4.589 Pa.sec and exceeds this limit. At 165°C, neat bitumen has a viscosity of 0.175 Pa.sec. All PMBs exhibit enhanced viscosity relative to the unmodified bitumen. The 4% SBS-PMB has a viscosity of 0.46 Pa.sec; EVA-GMA-PMBs exhibit an increase in viscosity with increasing polymer content up to 3% (1.048 Pa.sec), followed by a slight reduction at 3.5% (0.914 Pa.sec). The 2.5% EVA-GMA-PMB, exhibiting a viscosity of 0.595 Pa.sec, has a greater viscosity than the 4% SBS-PMB, which has a viscosity of 0.46 Pa.sec. All binders conform with the AASHTO M 320 standard, which requires a maximum viscosity of 3 Pa.sec at high temperatures.

Polymer modification consistently results in an important improvement in RV values at both 135°C and 165°C. Higher viscosity is directly associated with improved resistance to permanent deformation (rutting) at higher service temperatures. Viscosity influences the workability of the asphalt mixture during manufacture and paving; too high a viscosity requires higher mixing and compaction temperatures, possibly resulting in greater energy consumption and emissions or inadequate compaction in the field. The 3% EVA-GMA concentration, which exceeds the 3 Pa.sec limit at

135°C, suggests a potential workability issue, highlighting an important balance in PMB design where performance improvements must be weighed against practical application limitations. The 2.5% EVA-GMA-PMB has a greater viscosity than 4% SBS-PMB at both testing temperatures, although the 3% EVA-GMA is above the viscosity criterion at 135°C, and the 3.5% EVA-GMA shows an approximate reduction in viscosity relative to the 3% at both temperatures. Such behavior further supports the enhanced high-temperature performance effectiveness of 2.5% EVA-GMA, since it reaches higher flow resistance with reduced polymer content. The effectiveness is likely due to the chemical cross-linking enabled by the GMA component, which forms a more resilient and stable network structure within the bitumen. The exceedance of the viscosity criterion at 3% EVA-GMA indicates that this concentration may represent an "over-modification" point, when the advantages of enhanced stiffness are exceeded by workability challenges. The little reduction in viscosity from 3% to 3.5% EVA-GMA suggests intricate interactions at high polymer concentrations, resulting from polymer accumulation, saturation of reactive sites, or minor breakdown during mixing, necessitating additional examination.

The observed increase in viscosity of the bitumen samples modified with different doses of EVA-GMA may be ascribed to many variables evident from the FTIR measurements shown in Figure 10. The GMA component of EVA-GMA provides glycidyl groups that may react with carboxylic acid groups in bitumen or hydroxyl groups from other EVA chains, resulting in cross-linking. The creation of this network significantly raises the molecular weight and, as a result, the viscosity. The elevated peak intensities in areas linked to these functional groups confirm the existence of more extended cross-linking with more EVA-GMA content. This polarity enhances the interaction of bitumen molecules, resulting in improved flow resistance, which is evident as increased viscosity. EVA gives thermoplastic characteristics to the bitumen, seen at various temperatures. At 135°C, the enhanced softening and flow properties attributed to EVA presence are associated with increased viscosity, since the polymer phase transitions influence the total matrix hardness. At 165°C, while the viscosity remains greater than the control, it decreases across all samples owing to decreased intermolecular interactions at rising temperatures. The FTIR test confirms that the chemical modifications from EVA-GMA improve the structural integrity and flow resistance of bitumen, as shown by the viscosity measurements. The key causes for the observed increase in viscosity are the augmented cross-linking and improved molecular interactions generated by the additives. These adjustments enhance the physical qualities designed for particular performance criteria and may also prolong the service life of bitumen in paving.

A direct comparison of 4% SBS-PMB and different concentrations of EVA-GMA-PMB demonstrates significant performance properties. Both SBS and EVA-GMA polymers significantly enhance the characteristics of bitumen in comparison to the unmodified (control) binder. A comprehensive study demonstrates that EVA-GMA, especially at a 2.5% concentration, frequently demonstrates enhanced performance in critical high-temperature parameters (softening point and viscosity) relative to 4% SBS-PMB. The above information indicates that 2.5% EVA-GMA-PMB exhibits improved high-temperature performance (reduced penetration, increased softening point) and flow resistance (increased viscosity) relative to 4% SBS-PMB, which is very significant. This performance advantage is accomplished despite the 2.5% EVA-GMA mix having a reduced polymer concentration compared to the 4% SBS. This effectiveness clearly indicates a fundamentally more effective modification process for EVA-GMA. Although both polymers improve the binder, the extent and specific characteristics of this improvement vary, indicating different basic interaction processes with the bitumen matrix. The result strongly indicates the enhanced effectiveness of the EVA-GMA modification. The main difference in interaction mechanisms—chemical bonding in EVA-GMA compared to mainly physical interactions in SBS—offers an important reason for the identified performance differences and provides important advice for the development of next-generation polymer-modified asphalt binders.



(a) (b)

Figure 15. Rotational viscosity test results for all types of bitumen: (a) at 135 C^{o}, (b) at 165 C^{o}

3.4. Dynamic Rheological Properties Tests Results

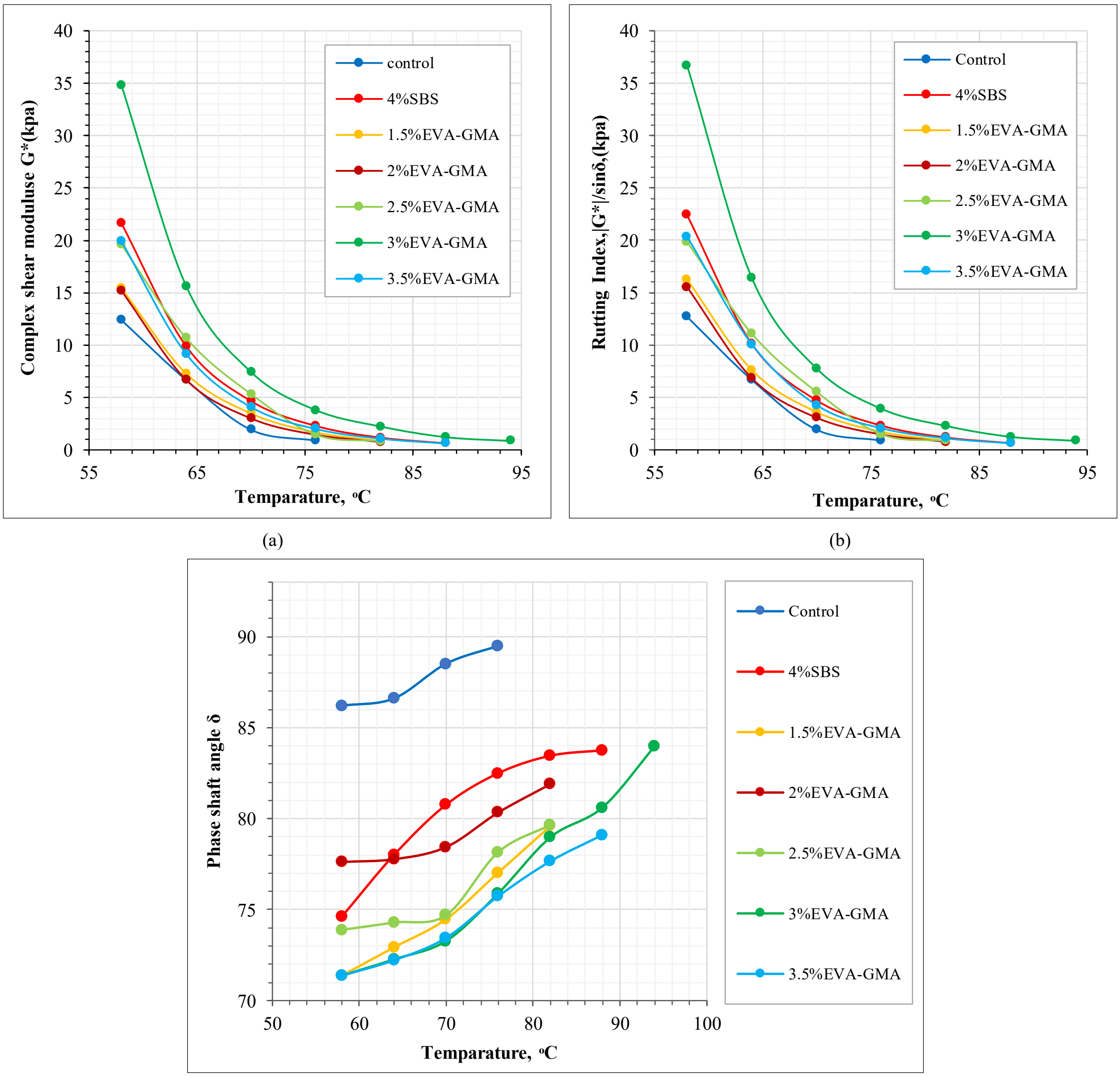
Figure 16 displays the results of testing different types of bitumen using the Dynamic Shear Rheometer (DSR). To understand bitumen's performance in paving applications, these tests measure its viscoelastic qualities, which include its resistance to deformation under load and temperature variations. The figures display the results for control bitumen, bitumen containing 4% SBS, and bitumen modified with different concentrations of EVA-GMA. The annual statistics from the Iraqi Central Bureau of Statistics show that the selected test temperature consistently stays above 58°C throughout the summer.

Figure 16a provides the values of the Complex Shear Modulus (G)*. The data shows that the control (neat) bitumen exhibits the lowest stiffness across all temperatures. At 58°C, the G* of the control binder is 12.3764 kPa, but it rapidly decreases to 0.8752 kPa at 76°C. Conversely, all PMBs have significantly enhanced G* values, indicating increased stiffness. The G* of the 4% SBS-PMB decreases from 21.6068 kPa at 58°C to 0.6289 kPa at 88°C. At 58°C, the G* of the 2.5% EVA-GMA-PMB is 19.5789 kPa, but at 88°C, it is 0.8303 kPa. For the EVA-GMA modifications, G* often exhibits a larger increase as the polymer percentage rises from 1.5% to 2.5% EVA-GMA over the whole test temperature. At lower temperatures, 2.5% EVA-GMA consistently exhibits the highest G* value. However, when the percentage exceeds 2.5% (such as 3% or 3.5% EVA-GMA), the G* values often stay constant or even decrease slightly, particularly at high temperatures. Such behavior indicates using the appropriate amount of polymer to enhance stiffness. As expected for viscoelastic materials, G* significantly decreases for all binders with increasing temperature. The decrease indicates that the materials exhibit a decrease in stiffness as the temperature increases.

Figure 16b demonstrates that the rutting resistance of all the PMBs significantly exceeds that of the control binder within the temperature range of 58°C to 94°C. At 58°C, the control binder has the lowest G*/sinδ value, at 12.6985 kPa. At 76°C, the G*/sinδ is 0.8752 kPa. At 58°C, the 4% SBS-PMB reaches 22.4094 kPa, whereas at 88°C, it decreases to 0.6326 kPa. The 4% SBS-PMB consistently exhibits the highest G*/sinδ values, exceeding those of the 2.5% EVA-GMA-PMB (19.8567 kPa at 58°C, decreasing to 0.8440 kPa at 88°C). The 2.5% EVA-GMA adjustment is more effective in reducing rutting in its original condition. As the temperature increases, G*/sinδ rapidly decreases for all binders. Figure 16c illustrates that the phase angle (δ) indicates the response of the binders to high temperature within the range of 58°C to 94°C. The control bitumen consistently exhibits the highest phase angle. For example, it is 86.21° at 58°C and 89.50° at 76°C. This result indicates that it is mostly viscous and not very elastic. The control exhibits larger phase angles than any of the PMBs, indicating more flexibility. Because of the elastomeric nature, the 4% SBS-PMB typically exhibits the lowest phase angles, namely 74.62° at 58°C and 83.76° at 88°C. EVA-GMA PMBs significantly reduce δ. For example, 2.5% EVA-GMA reduces the temperature to 73.89° at 58°C and then increases it to 79.66° at 88°C, exceeding SBS overall. As the temperature increases, the phase angle for all binders also increases, which meets with the expectations that the viscosity of binders decreases with increasing temperature.

The FTIR spectra, as shown in Figure 10, explain the chemical mechanism of action of the modified EVA-GMA bitumen's improved rheological qualities. When EVA-GMA is added, the FTIR spectra revealed the presence of ester (around 1735 cm-1) and ether (about 1100 cm-1) groups. These functional groups enhance the material's stiffness and resistance to deformation by strengthening intermolecular connections. The presence of epoxy group signals in the FTIR spectrum indicates that the glycidyl methacrylate component of EVA-GMA adds cross-linking capabilities. The cross-linking improves the bitumen's stiffness and elastic characteristics by creating a more linked molecular network. In addition to increasing bitumen's stiffness and elasticity, EVA-GMA changes considerably improve the rutting resistance of bitumen over a temperature range, according to DSR data corroborated by FTIR analysis. These enhancements are a result of the molecular changes brought about by EVA-GMA, namely the increased cross-linking and improved intermolecular forces caused by the integration of ester and ether groups. Because of this, EVA-GMA modified bitumen is a more multipurpose paving material that can withstand a wider range of temperatures and stresses.

Short-term aging, as shown by the Rolling Thin Film Oven Test (RTFOT), affects the rheological properties of all bitumen binders. Figure 17 demonstrates that all types of bitumen exhibit increased hardness and stiffness as the temperature rises from 58°C to 94°C. This improvement is evident in the original binder data presented in Figure 17. After RTFOT, the Complex Shear Modulus (G*) values for all binders significantly increase, with the control bitumen rising from 12.3764 kPa to 34.4454 kPa at 58°C and the 2.5% EVA-GMA-PMB rising from 34.7776 kPa to 37.157 kPa at 70°C. The Rutting Index (G*/sinδ) also increases for all binders following RTFOT across the 58°C to 94°C range, indicating enhanced rutting resistance as the binder hardens over time. The phase angle (δ) generally decreases for all binders across the test temperature range after RTFOT, indicating that the hardening process causes them to exhibit properties similar to solids or elastomers. The control bitumen's δ decreases from 86.21° to 81.21° (RTFOT) at 58°C and from 88.52° to 85.49° at 70°C. The 2.5% EVA-GMA-PMB's δ decreases from 73.89° to 72.98° at 58°C and from 74.70° to 73.54° at 70°C.Oxidation and volatile loss cause the physical hardness and changes observed during the RTFOT process. Rapid reactions between bitumen hydrocarbons and air oxygen, particularly during high-temperature mixing and compacting, lead to short-term aging. This reaction produces more polar molecules with greater molecular weights, often resulting in a bitumen matrix with more asphaltene-like parts. High temperatures cause lighter, more volatile parts of the bitumen, mainly maltenes, to evaporate and be lost, making the binder harder and stiffer. After RTFOT, all binders become harder, with the control binder showing a large rise in G*/sinδ, making it highly likely to harden quickly. Balancing short-term performance improvements with long-term durability concerns is crucial, as too much hardening during short-term aging may lead to premature pavement breaking due to fatigue.

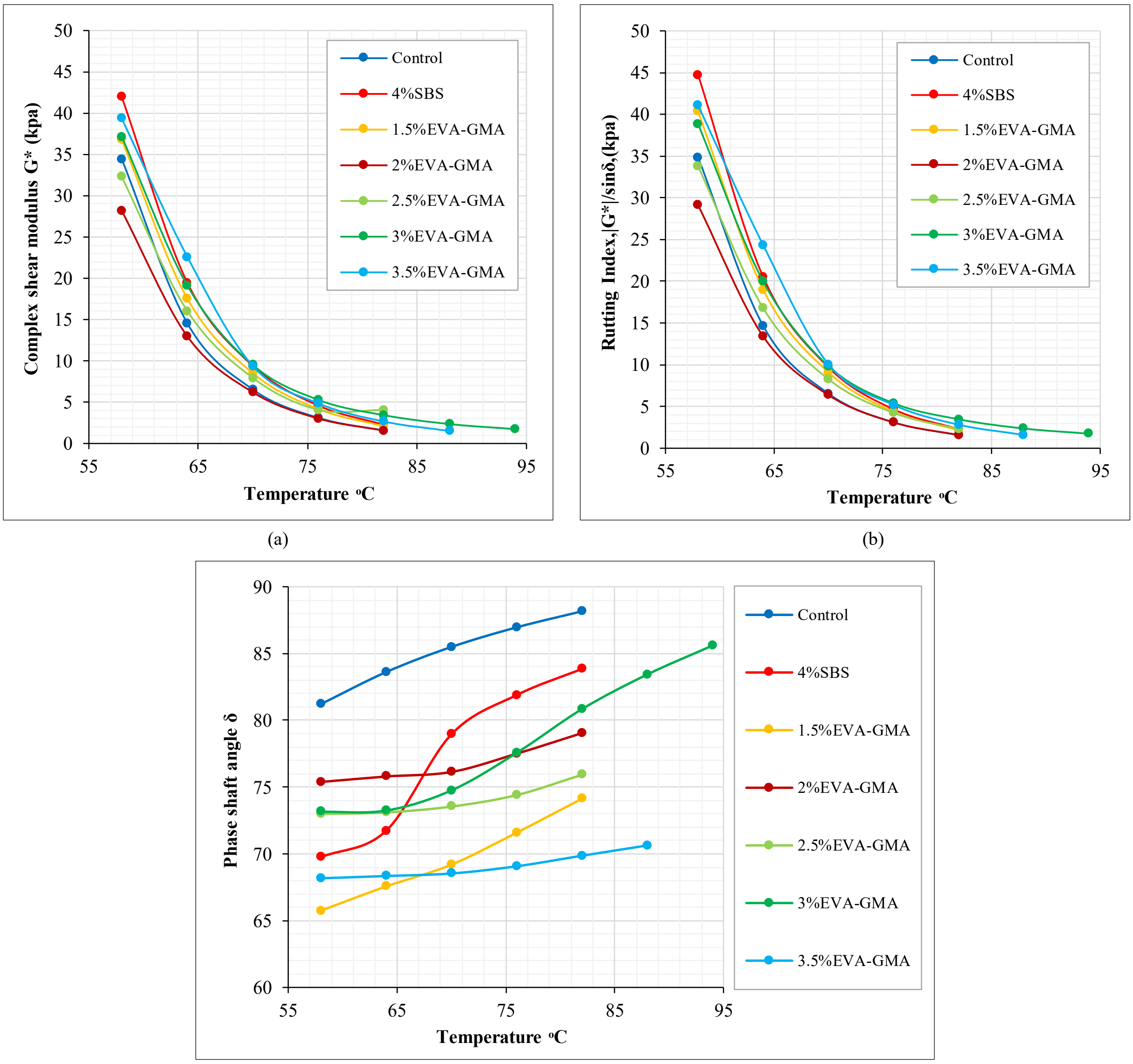


(a)

(b)

(c)

Figure 16. DSR test results for all types of bitumen (Original Binder): (a) complex shear modulus G*, (b) G*/sinδ, (c) Phase shaft angle, Temperature



(a)

(b)

(c)

Figure 17. DSR test results for all types of bitumen (After RTFOT): (a) complex shear modulus G*, (b): G*/sin, (c): Phase shaft angle, Temperature

A comprehensive analysis of 2.5% EVA-GMA-PMB and 4% SBS-PMB subject to various aging conditions demonstrates different performance properties, showing the advantages and disadvantages of each polymer modification. The 2.5% EVA-GMA-PMB consistently demonstrates superior rutting resistance, as shown by the rutting index $|G*|/\sin\delta$, over all temperature ranges compared to the 4% SBS-PMB. The dynamic modulus $|G*|/\sin\delta$ of the 2.5% EVA-GMA-PMB is 7.73 kPa at 70°C, significantly above the 4% SBS-PMB's 4.71 kPa. This indicates that the EVA-GMA-PMB is significantly less prone to resilient deformation when it is fresh and unused. The 4% SBS-PMB typically exhibits smaller phase angles (δ) compared to the 2.5% EVA-GMA-PMB. At 70°C, the SBS-PMB exhibits a phase angle of 80.77°, whereas the EVA-GMA-PMB displays a phase angle of 74.70°. The SBS-PMB exhibits a reduced phase angle, indicating its potential for superior strength. This characteristic is a fundamental aspect of elastomeric polymers. After Short-Term Aging (RTFOT), both binders exhibit the expected rise in rutting resistance. The 2.5% EVA-GMA-PMB (for example, 9.8434 kPa G*/sinδ at 70°C) generally exhibits superior resistance to rutting at higher temperatures compared to the 4% SBS-PMB (for example, 9.6944 kPa G*/sinδ at 70°C). Polymer chemistries and how they interact with bitumen have an effect on how well 2.5% EVA-GMA-PMB and 4% SBS-PMB work as an elastomer that makes the bitumen matrix a lot more elastic. This technique makes the bitumen more flexible, which means it won't break down as quickly over time. Styrene components form physical crosslinks with each other, while maltenes maintain the flexibility of the butadiene components and increase their size. This approach builds a network that is always flexible. EVA-GMA is a kind of plastic that makes bitumen thicker and stiffer. This modification makes it able to hold more weight and lessens the chance of irreversible deformation. The changes make it stronger and less likely to permanently distort, which makes it better at avoiding rutting.

3.5. Elastic Recovery Test Results

Figure 18 illustrates the impact of polymer modification on the flexibility of bitumen after RTFOT. The results indicate that all PMBs had superior elastic recovery compared to neat bitumen, which demonstrated an elastic recovery rate of 77%. The use of polymers enables the formation of a network of reinforcing components inside the bitumen matrix. This enhances the material's flexibility and its capacity to recover after being extended. The 2.5% EVA-GMA PMB can recover 90% of its original form, significantly exceeding the 4% SBS-PMB, which can only recover 81% of its original shape.

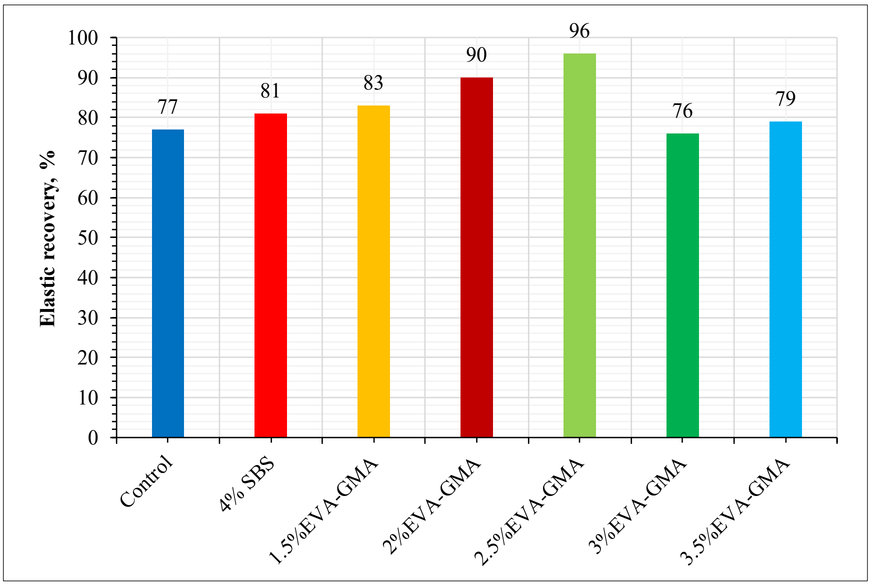


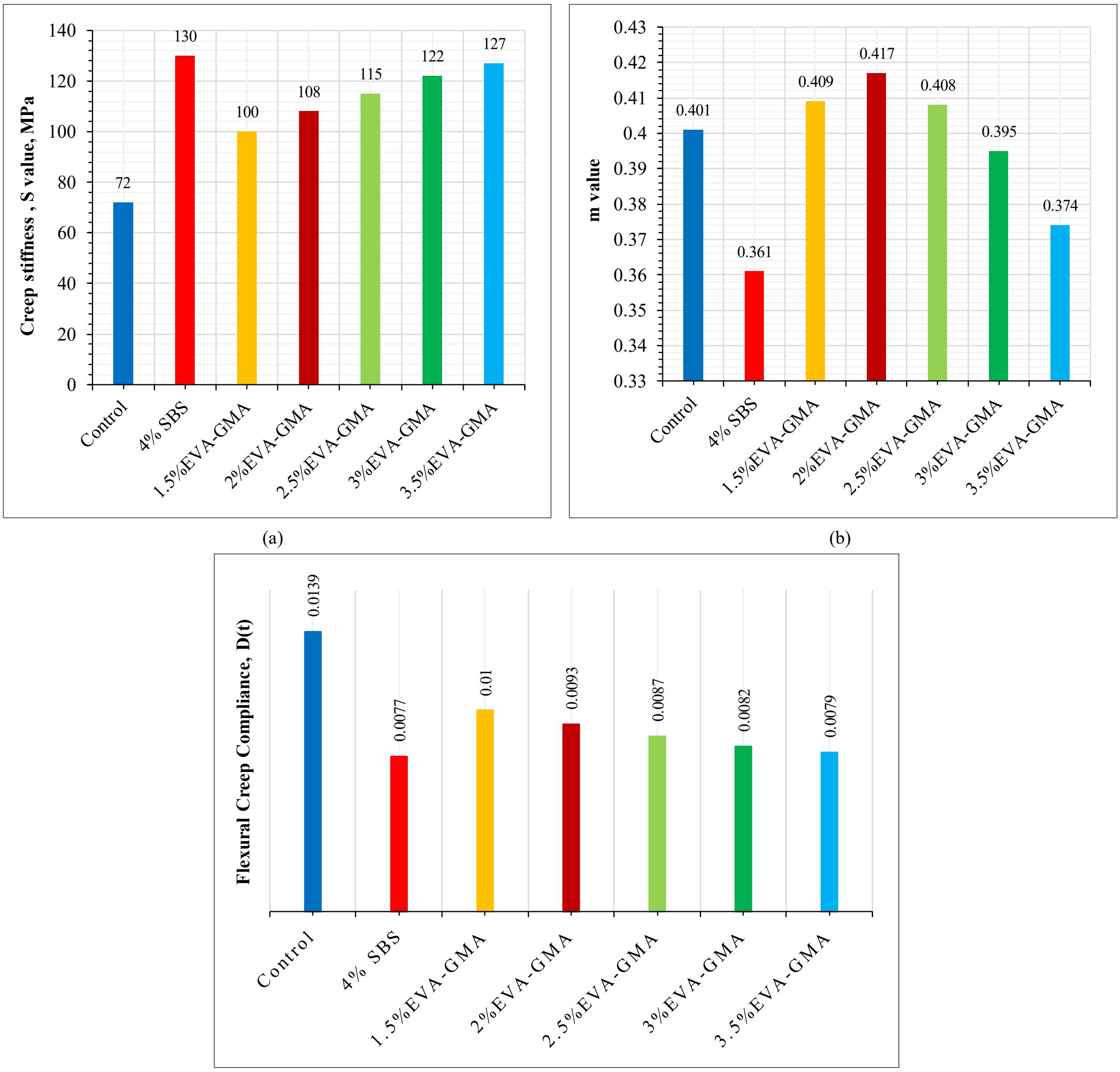
Figure 18. Relationships between the present of Elastic recovery of asphalt after RTFOT % and additives content

The EVA-GMA PMB exhibits superior performance due to its interaction with the chemical composition and structure of bitumen, enhancing the product's effectiveness. The polar functional groups of the EVA-GMA polymer enhance its compatibility with the polar components of bitumen, particularly the asphaltenes. This kind of interaction enhances the strength and interconnectivity of the polymer network. Conversely, SBS polymers usually interact with the maltene components of bitumen. The nonpolar SBS and the asphaltene-rich phases form a network, although they do not operate well in combination. Reduced contact makes the network elastic, resulting in decreased resilience compared to the optimal EVA-GMA modification. The incorporation of polymers into bitumen significantly impacts its microstructure. Polymers absorb the lighter, oily components of the maltenes. The resulting change causes the polymer to expand and form an individual, polymer-rich phase. This method generates a polymer network that remains inside the bitumen matrix.

This feature is important for maintaining system stability because it prevents the aggregation of asphaltene micelles. The 2.5% EVA-GMA-modified bitumen (PMB) offers the optimal equilibrium between the interaction of the polymer and bitumen and the development of the network over time. EVA-GMA forms a forceful polymer network that effectively interacts with asphaltenes. These are important features that influence the overall stiffness and flexibility of the material. The EVA-GMA polymer and asphaltenes function together, hence improving the material's elastic recovery. The FTIR spectra test results reveal chemical changes in asphalt's elastic recovery due to the presence and intensity of absorbance bands connected to ester, vinyl, and ether groups created by EVA-GMA. These groups influence the material's physical characteristics, such as flexibility and cross-linking in the bitumen matrix. High levels of EVA-GMA make it harder for the material to stretch and recover, causing recovery rates to drop. The thermal stability of the asphalt may also be affected by the components, with higher esters making the bitumen more likely to break down in heat. Excessive modification using EVA-GMA may reduce the asphalt's recovery capacity post-deformation, owing to hard cross-linked networks.

3.6. Low-Temperature Performance Tests Results

Figure 19 shows the results of a Bending Beam Rheometer (BBR) test, which measures the low-temperature performance of asphalt binders using three important parameters: flexural creep compliance (D(t)), m-value, and stiffness (S value). The asphalt's resistance to cracking at low temperatures may be evaluated by carefully considering these characteristics.



(a) (b)

(c)

Figure 19. Bending Beam Rheometer (BBR) test results: (a) S value (MPa), (b) m value, (c) creep compliance, D(t) and bitumen type

At low temperatures, the control (pure bitumen) exhibits distinctive rheological characteristics. The material exhibits the lowest creep stiffness (S = 72 MPa), the highest flexural creep compliance (D(t) = 0.0139), and an m-value of 0.401. These figures suggest an improvement in low-temperature flexibility; yet, it is well-known that standard bitumen has minimal ductility and resilience at low temperatures, resulting in its susceptibility to fracture. A low S-value and a high D(t) together do not indicate the effectiveness of unmodified binders in resisting cracking at low temperatures, since they fail to demonstrate the material's capacity to control or dissipate thermal stresses without fracturing. Polymers must be modified in order to achieve optimal ductility and toughness for enhanced performance at low temperatures. The use of both SBS and EVA-GMA polymers consistently results in a creep stiffness superior to that of the control bitumen. The 4% SBS-PMB exhibited the highest creep stiffness of all examined samples (S=130 MPa). An increase in polymer content in EVA-GMA-PMB results in enhanced stiffness. The stiffness increases from S=100 MPa at 1.5% EVA-GMA to S=127 MPa at 3.5% EVA-GMA. As these values increase, both the SBS-PMB and the EVA-GMA-PMB are significantly below the Superpave maximum S(60s) criterion of 300 MPa. This indicates that they have sufficient flexibility at low temperatures without becoming overly stiff to the point of fracturing.

The m-value, indicating the rate of tension relaxation, exhibits complex patterns. The control bitumen (0.401) exhibits a higher m-value than the 4% SBS-PMB (0.361), indicating that the SBS modification reduces stress at a slower rate. Conversely, with EVA-GMA-PMB, the m-value increases from 0.409 at 1.5% to a highest value of 0.417 at 2% EVA-GMA. This is the maximum m-value across all examined samples. The m-value gradually decreases to 0.374 at 3.5% EVA-GMA. The atypical behavior of EVA-GMA is significant, as it indicates an optimal range (about 2.5%) where polymer modification most effectively enhances the binder's capacity to reduce strain. Exceeding this quantity of plastomers may increase the material's stiffness, hence reduce the rate of viscous flow and decrease the m-value. All PMB, including SBS and EVA-GMA types, meet the Superpave minimum m(60s) requirement of 0. 300.

The flexural creep compliance, D(t), typically decreases as the polymer experiences changes, corresponding to the reported S-value. A reduced D(t) indicates that the material exhibits decreased flexibility and increased stiffness. The 4% SBS-PMB had the lowest D(t) value (0.0077), indicating it is the stiffest of the samples tested. As the polymer amount increases, D(t) for EVA-GMA-PMB consistently decreases, from 0.0100 at 1.5% to 0.0079 at 3.5%. This result confirms the concept that higher amounts of polymers increase stiffness.

The results are consistent with the study of Al-Nawasir & Al-Humeidawi [42] which demonstrated that the incorporation of a polymer increases creep stiffness and decreases flexural creep compliance. This suggests that altering a polymer, such as SBS, results in a harder binder that is less prone to deformation. The m-value for the EVA-GMA modifier indicates a distinct rheological behavior. The m-value reaches its maximum value at a concentration of 2.5% and then declines. This clearly shows the importance of discussing novel polymers, as their rheological properties may differ from those of established modifiers such as SBS.

The addition of EVA-GMA causes noticeable changes in the bitumen's chemical structure, as shown by the FTIR spectra results in Figure 10, which are connected with the enhanced stiffness. Peaks at 1735 cm^{-1} and 1100 cm^{-1}, respectively, show that these functional groups contribute to a more complex molecular structure, which in turn enhances the asphalt's stiffness. In the bitumen matrix, the presence of glycidyl groups in GMA causes cross-linking, as seen by shifts in the spectra around the absorbance peaks of the epoxy groups. Because it produces a more densely packed network structure inside the binder, cross-linking is a major factor in the enhanced stiffness. The bitumen is modified by EVA-GMA chemically changed, as can be seen by the appearance of new chemical groups that increase stiffness by cross-linking and molecular complexity, as shown by the BBR test results when analyzed with the FTIR spectra. These enhancements improve the bitumen's performance properties, particularly its stiffness and temperature sensitivity. The properties of obtained PMB with EVA-GMA are necessary for long-lasting pavement performance in different climates.

3.7. Asphalt Performance Grade (PG)

The results of the tests that were conducted on the types of asphalt binder used in the research according to the standard steps that conform to the international specifications and the AASHTO M320 specification, where the asphalt performance grade (PG) was chosen for each type of asphalt binder and according to the type and percentage added of each polymer used in the research, are shown in Table 2.

After analyzing the results shown in Table 2 and evaluating the indicators that were found or calculated through the standard tests above, represented by the viscosity, softening point, creep stiffness, elastic recovery, G*/sin δ, storage stability, and percent of solubility in trichloroethylene, it was noted that the modified asphalt using a concentration of 2.5% of EVA-GMA polymer is the optimal concentration that produced modified asphalt with a performance grade (PG) of (82-10) compared to the other concentrations used (1.5%, 2%, 3%, and 3.5% of EVA-GMA), although the concentration of 3% of polymer EVA-GMA produced modified asphalt with a performance grade (PG) of (88-10). The reason for not choosing concentrations of 3% and 3.5% of EVA-GMA, respectively, is that they achieved a viscosity higher than the limits required for the AASHTO M320 specification (3 pa.sec). The increase in the viscosity value of asphalt has negatively affected the value of elasticity recovered, which is necessary to resist failure due to fatigue at intermediate temperatures (25°C). Also, the creep stiffness value was high, which affects the performance of paving at low temperatures and makes the paving exposed to failure by cracking at low temperatures.

By comparing the results of the performance grade (PG) of the asphalt modified using EVA-GMA, the asphalt modified using SBS, and the normal asphalt, it was noted that the asphalt modified using EVA-GMA at a concentration of 2.5% achieved a performance grade (PG) of 82-10, which is the same as the performance grade (PG) achieved using 4% of SBS. On the other hand, using a concentration of 2.5% of the polymer EVA-GMA produced asphalt with better viscosity, softening point, creep stiffness, elastic recovery, G*/sin δ, storage stability, and percent of solubility in trichloroethylene than in the case of using 4% of SBS. According to obtained results, EVA-GMA can be considered as a superior technique for enhancing asphalt performance with less polymer content compared to SBS. This approach provides superior overall characteristics as well as reduced expenses in comparison to SBS-modified asphalt. The superior performance of EVA-GMA is attributed to its reactive functional groups, which facilitate compatibility, chemical bonding, and network formation in the asphalt. This results in enhanced viscosity, elasticity, heat stability, and overall durability, even at reduced polymer concentrations relative to SBS polymer [43].

The present investigation demonstrates significant improvements in storage stability, viscosity, and overall performance grade from the use of EVA-GMA (LOTADER® AX8670T) terpolymer in PMB. These results consistent with, and in some cases exceed, those recorded in previous research with SBS polymers. Al-Nawasir & Al-Humeidawi [42] determined that a 5% addition of SBS optimally enhanced the physical properties and performance grade (PG76-10) of local bitumen in Iraq. This study achieves a performance grade of PG82-10 with just 2.5% EVA-GMA, signifying a more effective enhancement of rheological properties at a lower concentration. Werkovits et al. [44] indicated that the storage stability of SBS-PMB remains a concern due to the potential for phase separation at high temperatures. The aging processes degraded 21–25% of SBS molecular substructures, resulting in significant polymer deterioration. The EVA-GMA modified binder used in this research exhibited an 11% greater stability during storage compared to the 4% SBS-PMB. This indicates that it resists heat more effectively and decompose in phases. Sakib et al. [45] emphasized the significance of elastic and torsional recovery in the assessment of PMB. SBS-modified binders have shown a significant correlation between recovery tests and polymer content. SBS generally enhances elastic recovery; nevertheless, it requires high mixing temperatures and is susceptible to oxidation and agglomeration. The binder modified with EVA-GMA in this investigation was 50% substantial than the binder modified with SBS and 100% thicker than neat asphalt. It exhibited superior performance, requiring less mixing time and can be mixed at significantly lower temperatures. In conclusion, the current investigation indicates that EVA-GMA terpolymer performs equally or better compare to SBS-PMB with lower quantity of additive and less mixing energy or effort. This result indicates that obtained PMB represents an ecologically sustainable and cost-effective.

4. Conclusions

The results highlight the effectiveness of EVA-GMA as a suitable asphalt modification that enhances mechanical qualities and performance while exhibiting reduced environmental implications relative to traditional SBS modifiers. This makes it an attractive option for improving asphalt's durability and functional properties under diverse operating settings. The research on using EVA-GMA as a modifier for asphalt binders reached the following main conclusions:

- The optimal amount of EVA-GMA for the modification of asphalt binders was determined to be 2.5% by weight. This concentration offered the optimal equilibrium between performance enhancement and economic viability.
- The asphalt modified with 2.5% EVA-GMA exhibited notable enhancements in viscosity and storage stability, demonstrating a 50% increase in viscosity and an 11% improvement in storage stability relative to asphalt modified with 4% SBS-PMB.
- Both EVA-GMA-modified asphalt and SBS-modified asphalt attained a performance grade of PG 82-10, exceeding the PG 76-10 of unmodified bitumen. This indicates that EVA-GMA achieve similar improvements in high-temperature performance at a reduced concentration relative to SBS.
- The incorporation of EVA-GMA resulted in enhanced mechanical characteristics, including high complex modulus and elastic recovery, which are essential for the asphalt's performance under dynamic loading conditions.
- EVA-GMA serves as a highly efficient modifier for asphalt binders, enhancing mechanical characteristics and performance while having a reduced environmental impact compared to traditional SBS modifiers.

5. Declarations

5.1. Author Contributions

Conceptualization, O.H.C., B.H.A., and A.H.A.; methodology, B.H.A. and A.H.A.; investigation, O.H.C.; data curation, O.H.C.; writing—original draft preparation, O.H.C.; writing—review and editing, O.H.C., B.H.A., and A.H.A. All authors have read and agreed to the published version of the manuscript.

5.2. Data Availability Statement

The data presented in this study are available in the article.

5.3. Funding

The authors received no financial support for the research, authorship, and/or publication of this article.

5.4. Conflicts of Interest

The authors declare no conflict of interest.

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