Utilization of Pyrolytic Carbon Black Derived from Scrap Tyres for Enhance Bitumen Properties

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Abstract: Increasing disposal of tyres poses significant environmental challenges, prompting the exploration of effective reuse methods such as pyrolysis, which produces Fuel oil and carbon black (CB) as a byproduct. This research investigates the utilization of CB, a waste byproduct of pyrolytic plants, as a bitumen modifier to enhance the performance of asphalt binders. Using penetration grade 60/70 bitumen from Sri Lanka, CB was incorporated at varying concentrations (2%, 6%, 10%, and 14%) to evaluate its impact on physical and rheological properties. Conventional tests revealed that CB modification increased the bitumen softening point while decreasing penetration and ductility. Rheological assessments using a Dynamic Shear Rheometer (DSR) indicated that CB-modified bitumen exhibited improved resistance to rutting and enhanced elastic behavior, particularly at lower temperatures. The rutting factor $G^*/\sin\delta$ also increased with CB content, confirming enhanced deformation resistance. However, beyond 6% CB content, the benefits plateaued or diminished, highlighting the importance of optimal concentration. This study underscores the potential of CB modification to improve bituminous materials for road construction, offering a sustainable solution to waste disposal and enhanced road performance
Keywords: Pyrolytic carbon black, Bitumen modification, Environmental impact mitigation, Scrap tyres.

Introduction

The increasing usage of automobiles and trucks has led to a corresponding increase in the disposal of tyres, posing significant environmental challenges. Effective tire disposal solutions have become imperative with the high volume of discarded tires. Various technologies and processes have been developed to address this issue, among which pyrolytic plants play a prominent role. Pyrolytic plants employ a method wherein tyres are subjected to high temperatures without oxygen, producing fuel oil as the main product and Carbon Black (CB) as a waste by-product [1].

Through the pyrolysis process, tyres are broken down into their constituent components, and oil is extracted, which resembles diesel in its properties. This oil can be refined and used for various purposes, reducing reliance on conventional fossil fuels. CB [1], one of the byproducts of the pyrolysis process, is generated in significant quantities.

CB is generated as a byproduct of the pyrolytic process, comprising approximately 25 % of the total output [2]. While some of this carbon black finds applications in various industries, a significant portion often remains unused and is stored in storage yards. Despite its potential utility, disposing of unused carbon black poses challenges, particularly in managing storage and disposal logistics.

Only a few pyrolysis plants in Sri Lanka are operating, utilizing scrap tyres to produce fuel oil. However, the carbon black made in the process remains unprocessed in most of the pyrolytic plants in Sri Lanka. It is stored in the dumping yards, leading to significant environmental issues and ultimately resulting in the suspension of plant operations [3].

In recent years, numerous studies have been conducted worldwide to explore the utilization of CB as a modifier for asphalt binders [4][5][6]. A few research projects have also been undertaken in Sri Lanka, with one trial section laid. Notably, studies in Sri Lanka primarily focus on incorporating carbon black into asphalt concrete rather than directly into bitumen.

This study evaluated the utilization of CB as a bitumen modifier, using CB taken from Sri Lankan plants to modify bitumen properties.

The primary objectives of this research were threefold: first, to determine the optimal percentage of CB that can be effectively utilized for bitumen modification; second, to investigate the impact of incorporating CB on bitumen's mechanical and rheological properties; and third, to assess the effects of using this modified bitumen in asphalt concrete production, particularly on the properties and performance of the asphalt concrete mixture.

Material & Sample Preparation

Asphalt Binder

The asphalt binder utilized for this study was penetration grade 60/70 penetration grade bitumen, which Sri Lanka typically prefers for Road construction works.

Pyrolysis Carbon Black

The CB obtained from the Global Macro Industries (Pvt) Ltd pyrolysis plant (Figure 1), which operates in Wagawatta / Sri Lanka, was taken for bitumen modification for this research project. This carbon black is typically disposed of as waste material in its unprocessed state. Due to its unprocessed nature, it retained steel components in the tyres from which it was derived. Therefore, these steel components are removed before being used in testing procedures to prevent any potential damage to the complex testing equipment.

Initially, attempts were made to sieve the carbon black using a 65μm sieve. However, this method proved challenging due to its powdery consistency and nanoparticle size. The carbon black dispersed throughout the laboratory due to its lightweight nature, impeding the sieving process.

Subsequently, an alternative method, utilizing a magnet, was employed to remove steel wires. This approach successfully removed all steel components from the carbon black, facilitating its suitability for laboratory testing.

Figure 1: Pyrolytic plant at Wagawatta / Sri Lanka

Sample Preparation

The preparation of bitumen samples involved several sequential steps. Firstly, the bitumen was heated to 150°C in an oven to ensure it reached the desired temperature for subsequent processing. Following heating, the heated bitumen was carefully transferred into a mixing bowl, where the temperature was adjusted to 160°C to facilitate proper blending. CB, used to enhance the properties of bitumen, was then incorporated into the bitumen at varying percentages ranging from 2%, 6%, 10%, and 14% by weight of the Bitumen. Once the appropriate amount of CB was added, the mixing process commenced in the laboratory mixer, operating at a rate of 1000 rpm to 1200 rpm, and continued for one hour. This thorough mixing ensured uniform dispersion of the CB throughout the bitumen, thereby achieving consistent and reproducible samples for subsequent testing and analysis.

Bitumen blended with CB at different percentages is henceforth denoted as CBMB-2%, CBMB-6%, CBMB-10%, and CBMB-14% for clarity and ease of reference in subsequent discussions.

Methodology

The methodology employed in this research begins with examining both 60/70 penetration grade bitumen and Carbon Black modified bitumen, focusing initially on conventional properties such as penetration, softening point, and ductility. Subsequently, rheological properties are assessed for all samples, utilizing the Dynamic Shear Rheometer (DSR) to determine complex shear modulus and phase angle at different temperatures. The optimal maximum percentage of CB blended with bitumen is determined through a comprehensive analysis of these results.

After that, mixing and compaction temperatures are determined according to the prescribed test method outlined in the Asphalt Institute guidelines.

Finally, asphalt concrete samples were prepared according to the marshal method for evaluation. Samples for wearing and binder courses were fabricated for the Marshal test method analysis, which aimed to comprehensively measure the properties of the asphalt.

This methodological approach (summarized in Figure 2) ensures a thorough understanding of the effects of Carbon Black modification on bitumen properties, encompassing both laboratory testing and performance evaluation of asphalt concrete samples.

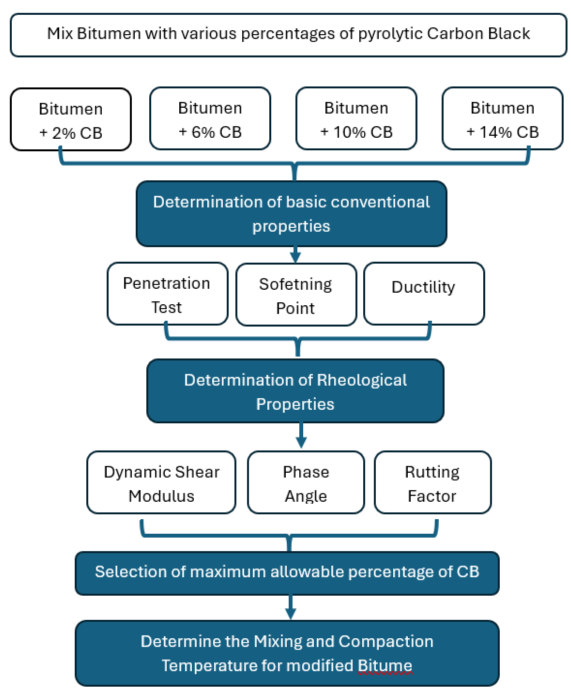

Figure 2: Flow Diagram of the Study Methodology

Result & Discussion

Physical Property Testing

Conventional physical tests, essential for understanding the behaviors of the critical material, were initially conducted on 60/70 penetration grade bitumen and modified bitumen mixtures. These tests provide crucial insights into the fundamental characteristics of bitumen, both in its virgin form and when modified with carbon black. Understanding these properties is paramount for ensuring bituminous materials' optimal performance and longevity of the road pavement [6].

Table 1 presents the tested physical properties of virgin 60/70 bitumen and the specification limits prevailing in Si Lanka for road construction works.

Table 1: Properties of 60/70 Penetration Grade Bitumen

Tested Parameter	**Test Standard**	**Test result**	**Specification Limit/s**
Penetration	ASTM D5	64	60–70
Softening Point	ASTM D36	49.3	48–56
Ductility	ASTM D113	107	> 100

Figure 3(a) to Figure 3(c) illustrate the variation of Penetration, softening point, and Ductility of modified bitumen with CB percentage.

The penetration test provides valuable insights into a material's capacity to withstand various stresses, including traffic loads, environmental factors, and temperature fluctuations [7]. According to the analysis shown in Figure 3(a), an increase in the percentage of CB results in a decrease in bitumen penetration.

The softening point of bitumen is a crucial indicator of its behavior during the transition from solid to liquid states, particularly in the context of road pavement [7]. As Figure 3(b) demonstrates, adding CB increases the bitumen's softening point. This suggests that the modified bitumen can withstand high temperatures, improving its resistance to deformation under traffic loads and long-term structural integrity.

Moreover, the ductility of the bitumen significantly decreases after reaching a CB content of 6%, as shown in Figure 3(c). This observation aligns with previous research, corroborating the findings of Wang, H., et al. [8]. The consistency across studies reinforces the validity of our results and supports the notion that incorporating CB enhances the high-temperature properties of bitumen. This enhancement can be attributed to CB's increased capacity to absorb heat and its high surface area.

Based on these findings, further investigation was conducted to study the behavior of this modified bitumen. Complex testing on bitumen modified with 2%, 6%, and 10% CB was carried out to identify its rheological properties.

Rheological Properties

The rheological properties of modified bitumen play a pivotal role in determining its performance within road pavement applications, particularly under varying temperatures and loading conditions. To assess these properties, a Dynamic Shear Rheometer (DSR) was utilized to evaluate the dynamic shear modulus (G^*), phase angle (δ), and rutting factor ($G^*/\sin\delta$) of the modified asphalt samples.

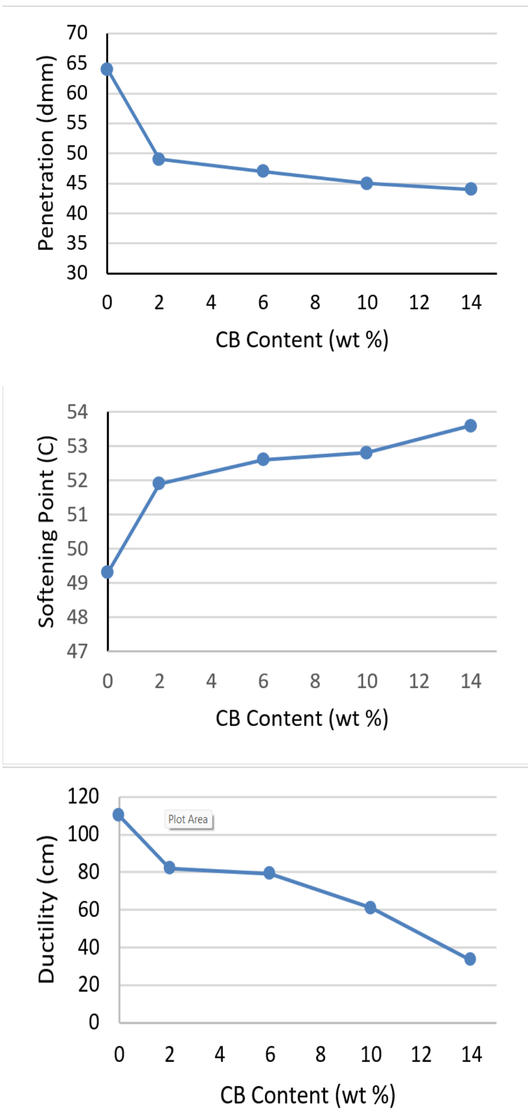

Figure 3: Conventional physical properties of bitumen with different contents of CB: (a) softening point; (b) penetration; and (c) ductility.

This experimental approach offers invaluable insights into the behavior of bitumen, enabling a deeper understanding of its mechanical response and durability in the field [9].

The dynamic shear modulus and phase angle was determined across a temperature range from 52°C to 70°C, capturing critical thermal conditions relevant to asphalt pavement environments.

Figure 4 illustrates the variation of failure temperature, defined as the temperature at which $G^*/\sin(\delta)$ equals 1.

The data in the figure indicates that bitumen failures occur at higher temperatures as the CB content increases to 6%. However, when the CB content exceeds 6%, failures appear at lower temperatures, even lower than the failure temperature of the 60/70 bitumen. This suggests that increasing the CB content beyond 6% does not continue to enhance, and may even detract from, the rheological properties of the bitumen.

A detailed evaluation was conducted over a range of temperatures to understand further the impact of CB content on the rheological properties of bitumen. Figure 5 illustrates the complex relationship between the CB content and the rheological behavior of the modified bitumen.

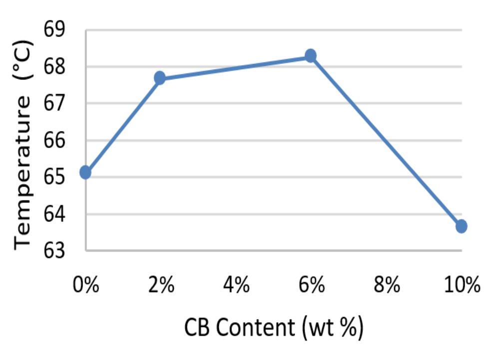

Figure 4: Failure Temperature for different CBMB samples

Specifically, it shows variations in the dynamic shear modulus (G^*) and phase angle (δ) across different temperatures for each level of CB content. The complex modulus indicates the stiffness of the bitumen, while the phase angle provides insight into the viscoelastic balance between the elastic and viscous behavior of the material. At lower CB contents, up to 6%, the increased stiffness and improved temperature susceptibility suggest enhanced performance characteristics, such as more excellent resistance to permanent deformation under load. However, beyond 6% CB content, the bitumen begins to show reduced performance, likely due to the over-saturation of CB particles, which can interfere with the bitumen matrix and negatively impact its viscoelastic properties. These findings are critical for developing and optimizing bitumen formulations, as they highlight the importance of balancing CB content to achieve desired performance characteristics. Additionally, the behavior of the rheological properties over a range of temperatures was evaluated for further analysis. Fig. 5 illustrates this intricate relationship, showing variations in complex modulus and phase angle across different temperatures for each CB content.

Integrating the observations from dynamic shear modulus and phase angle test results shown in Figure 5 provides a comprehensive understanding of how Carbon Black modification influences the rheological properties of modified bitumen.

Firstly, the analysis of the complex shear modulus (Figure 5(a)) reveals a consistent trend across temperatures, indicating a decrease in modulus with increasing temperature for all samples.

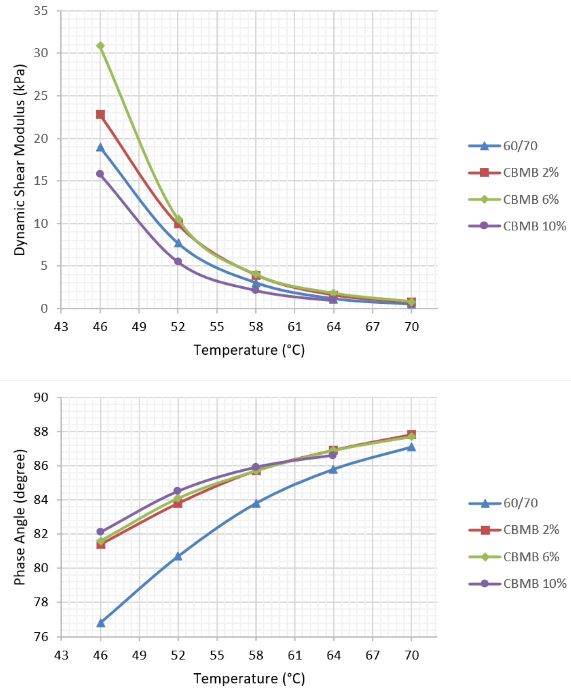

Figure 5: Rheological parameters of bitumen with different contents of CB at different temperatures: (a) complex shear modulus and (b) phase angle.

This behavior aligns with the expected response of bitumen, which tends to become less stiff and more flowable at higher temperatures. Furthermore, introducing Carbon Black modification increases stiffness in the bitumen matrix, as evidenced by higher dynamic shear modulus values than the original 60/70 bitumen for the CBMB 2% and CBMB 6% samples. However, the trend changes for the CBMB 10% sample, and the dynamic modulus value decreases below that of the original 60/70 bitumen. The increase in modulus values indicates that Carbon Black reinforces the bitumen, enhancing its rheological properties.

Similarly, the phase angle variations highlight the impact of temperature and Carbon Black modification on the viscoelastic behavior of bitumen. As temperatures rise, all samples consistently decline in phase angle, indicating a transition towards more fluid-like behavior. However, modified bitumen with Carbon Black consistently exhibits lower phase angle values than the original bitumen, indicating an enhancement in stiffness and elastic behavior due to Carbon Black reinforcement. Moreover, the concentration of Carbon Black influences the degree of improvement in stiffness, with higher concentrations resulting in lower phase angle values, indicative of enhanced elastic behavior. Unlike the variation in modulus values, the phase angle increases continuously with the increase in Carbon Black content. However, this increase in phase angle is significant at lower temperatures (up to 58°C) and less pronounced at higher temperatures for high Carbon Black percentages.

Like the dynamic modulus and phase angle, the rutting factor $G^*/\sin(\delta)$ is also an effective parameter for evaluating bitumen samples. The rutting factor reflects the ability of the asphalt binder to resist deformation, which inherently decreases with an increase in temperature. Generally, asphalt binders with a higher rutting factor have more potential to resist deformation and rutting at high temperatures. The $G^*/\sin(\delta)$ test results for the bitumen samples evaluated in this study are graphically shown in Figure 6 as a function of temperature.

With an increase in the CB content, the rutting factors correspondingly increased, indicating an improvement in the rutting resistance of the modified bitumen. This is similar to the complex modulus, which also shows a nonlinear relationship. According to the SHRP specification [10], the value of $G^*/\sin(\delta)$ must be more than 1.0 kPa for asphalt binders to mitigate the rutting phenomenon.

As shown in Figure 6, the rutting factor ($G^*/\sin(\delta) > 1.0kPa$) for the 60/70 penetration grade bitumen should not exceed 64°C and 70°C for the and the CBMB, respectively. For the 10% CB content, it is 64°C. This illustrates the potential of CBMB to reduce the temperature sensitivity of the asphalt binder and enhance its high-temperature rutting resistance potential up to a CB content of 6%.

Comparing these results reveals additional insights. The diminishing return observed with higher concentrations of Carbon Black in the dynamic shear modulus data is mirrored in the phase angle results, where there is a plateauing effect in stiffness improvement beyond a specific concentration. This suggests that while higher concentrations of Carbon Black lead to greater stiffness enhancement, there is an optimal concentration beyond 6%. Further increases do not yield significant additional benefits in terms of viscoelastic behavior.

In summary, integrating dynamic shear modulus and phase angle data highlights the multifaceted influence of Carbon Black modification on bitumen's physical and viscoelastic properties. It underscores the importance of Carbon Black concentration in achieving desired stiffness improvements while considering the balance between performance enhancement and practical application considerations across various temperatures.

Rotational Viscosity

Viscosity plays a crucial role in assessing the flow characteristics of bitumen binders due to their viscoelastic nature. This property is influenced by temperature and the loading rate, impacting mixability and workability. Rotational viscosity tests were conducted using a Brookfield viscometer at temperatures of 135°C and 165°C, yielding results plotted in Figure 3 for both original 60/70 bitumen and CBMB samples.

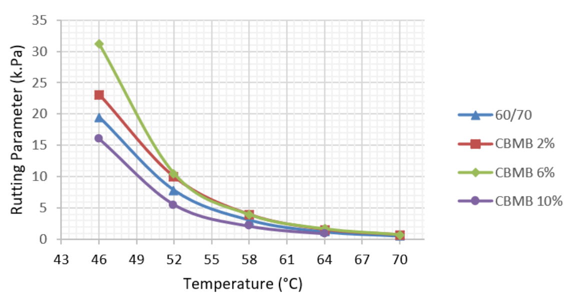

Figure 6: Variation of rutting factor with different contents of CB at different temperatures

The findings reveal a consistent trend: as the temperature increased from 135°C to 165°C, the viscosity of all binders decreased proportionally. Additionally, incorporating CB had a notable influence on viscosity at 135°C, whereas its effect was diminished at the higher temperature of 165°C. This temperature versus viscosity variation graph can be used to determine the mixing and compaction temperatures for asphalt concrete. According to the recommendations given in NCHRP project 09-39 [11], the recommended viscosity ranges for mixing and compaction temperatures are 0.15 Pa.s to 0.19 Pa.s and 0.32 Pa.s to 0.38 Pa.s respectively.

Based on the test results, the mixing and compaction temperatures for CBMB were analyzed for all the CB-modified bitumen samples and are presented in Table 3.

Conclusion

The comprehensive analysis of the physical and rheological properties of bitumen modified with CB presents valuable insights into the potential enhancements in bituminous materials for road construction applications. The physical property testing revealed a nuanced relationship between CB content and fundamental bitumen characteristics. As the CB percentage increased, penetration decreased, indicating potential improvements in bitumen's ability to withstand stresses and environmental factors. Additionally, the softening point increased with CB modification, suggesting enhanced resistance to high temperatures and deformation under traffic loads. However, ductility decreased beyond a certain CB content, aligning with previous research and emphasizing the complex interplay between CB incorporation and bitumen properties. Further investigation into rheological properties through rotational viscosity and Dynamic Shear Rheometer (DSR) testing provided deeper insights into the viscoelastic behavior of CB-modified bitumen. The findings demonstrated that CB modification led to increased stiffness in bitumen, improving its resistance to rutting and enhancing elastic recovery. Moreover, the concentration of CB influenced the degree of improvement in stiffness, with diminishing returns observed at higher concentrations.

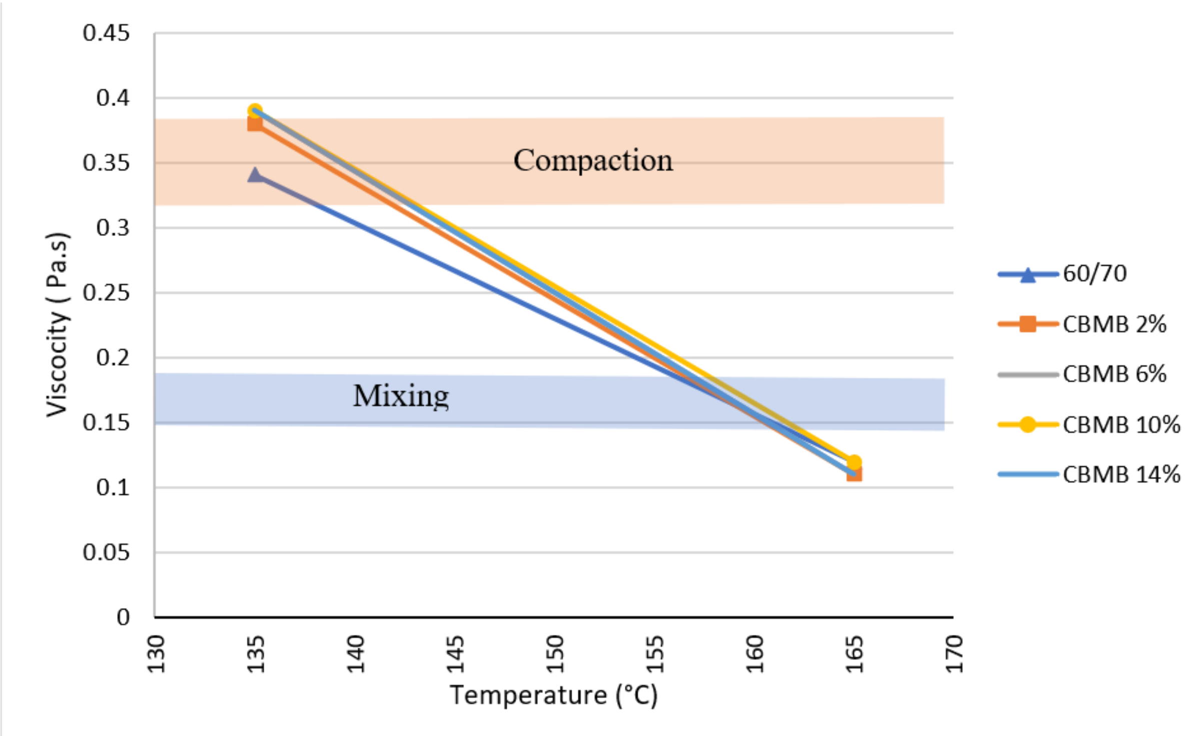

Figure 7: Rotational viscosity of bitumen with different contents of CB

Table 2: Mixing/Compaction temperatures of CBMB

Binder	Rotational Viscosity (Pa.s)		Temperature Range (° C)	
	at 135 ° C)	at 165 ° C)	Mixing	Compaction
60/70	0.34	0.12	155.0-162.0	139.0-147.0
CBMB-2%	0.38	0.11	156.0-160.5	135.0-142.0
CBMB-6%	0.39	0.11	156.5-161.0	135.0-142.0
CBMB-10%	0.39	0.12	157.0-162.0	136.0-143.0
CBMB-14%	0.39	0.11	157.5-162.0	136.0-143.0

The integration of complex shear modulus and phase angle data highlights the influence of CB modification on bitumen properties across varying temperatures. While higher concentrations of CB enhanced stiffness, there is an optimal concentration, identified as 6%, beyond which further increases did not yield significant benefits.

In conclusion, this research underscores the potential of CB modification to enhance the performance and longevity of bituminous materials in road construction. By understanding the complex relationship between CB content and bitumen properties, engineers and policymakers can make informed decisions regarding the formulation and application of modified bitumen, ensuring sustainable and resilient road infrastructure. Further studies could explore additional aspects of CB-modified bitumen and its practical implications in real-world road construction scenarios.

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