



# SYNERGISTIC ASSESSMENT OF CARBON BLACK AND CRUMB RUBBER ON THE PERFORMANCE OF ASPHALT BINDER: PHYSICOCHEMICAL CHARACTERIZATION AND DOSAGE OPTIMIZATION

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## Abstract

The major objective of the study was to optimize the combined dosage of carbon black (CB) and crumb rubber (CR) for maximizing the synergistic potential of CB-CR conglomerate, chiefly to produce a set of CB-CR-modified asphalt binders with superior performance than the traditional asphalt binder. Scanning Electron Microscopy revealed that CB featured a porous, reinforcing morphology, while CR showed rough, swelling-prone structures. Fourier Transform Infrared Spectroscopy further identified unsaturated hydrocarbon groups in CR that promoted enhanced elastic recovery; in contrast, CB contained aromatic and oxygenated groups that enhanced stiffness, aging resistance, and overall durability. Consistency test results showed a progressive reduction in Penetration and ductility of asphalt binder accompanied by increased softening point and viscosity with varying CB-CR percentages, indicating enhanced stiffness and thermal stability. Creep and recovery tests showed significant improvement, with higher percent recovery and lower non-recoverable creep compliance compared to the unmodified binder, demonstrating enhanced high-temperature performance through synergistic stiffness and elasticity effects. Asphalt binders with balanced CB-CR proportions, i.e., 10% each of CB and CR by weight of the base asphalt binder that was designated VG10-CB50:CR50 exhibited acceptable storage stability, whereas CR-dominant asphalt binders showed a propensity towards phase separation. Compatibility ranking identified VG10-CB50:CR50 as the optimum blend, offering the best balance of rheological performance and storage stability for high-temperature as well as heavy-traffic pavement applications.

*Keywords: pyrolysis, carbon black, crumb rubber, synergistic modification, waste valorization, compatibility ranking*

## 1 Introduction

Over the past few decades, rapid urbanization coupled with the exponential population growth have caused a significant increase in generation of waste materials [1]. Per current estimate, global waste tire generation is expected to reach approximately 5,000 million tires by 2030 [2]. Currently, the end-of-life tires (ELTs) are being dumped, buried, or disposed in open landfills in an unscientific manner, which has caused several environmental issues such as land pollution, groundwater contamination, and increased carbon emissions [3]. Furthermore, due to their non-biodegradable nature, ELTs persist within the environmental hierarchies and contribute to the release of toxic pollutants such as dioxins and furans [4]. Therefore, it has become necessary to adopt scientific approaches to effectively utilize the ELTs for diverse applications to promote the waste-to-wealth concept.

To date, several attempts have been made to pre-process and integrate ELTs for application across different end uses. For instance, mechanical recycling techniques such as shredding and thermochemical processes such as pyrolysis have emerged as viable solutions for up-cycling the ELTs [5]. However, the insufficient availability of recycling facilities, coupled with the complexities of recycling processes has negatively influenced recycling rates [6]. Consequently, researchers have strategically incorporated ELT-derived residues such as pyrolysis carbon black, also known as the carbon black (CB) and crumb rubber (CR) into asphalt pavement systems, yielding superior set of modified asphalt binders and mixtures designed to withstand extreme high- and low-temperature distresses [7].

Amongst various sustainable modification strategies, CR derived from shredding of ELTs has received significant attention due to its environmental and engineering benefits. CR-modified asphalt binders exhibit improved elasticity, enhanced rutting resistance, and superior fatigue performance, primarily due to rubber particle swelling and interaction with the asphalt matrix [8]. However, despite these notable performance enhancements, their field application remains challenging due to elevated viscosities that risk phase separation without proper blending [5]. Furthermore, these higher viscosities entail increased mixing and compaction temperatures, resulting in greater energy input and greenhouse gas emissions [9]. On the other hand, CB, a by-product of scrap tire pyrolysis has been used as a valuable additive in rubber and other compound formations [10]. Previous study has demonstrated that CB-modified asphalt binders can enhance high-temperature stiffness, improve rutting resistance, and increase aging resistance of asphalt binders by restricting molecular mobility and reinforcing the binder microstructure [11]. Elsewhere, researchers investigated the aging characteristics of CB-modified asphalt binder and concluded that incorporating CB can significantly enhance the anti-aging properties and thermal conductivity of the conglomerate even at relatively low concentration [12].

Despite their individual benefits in performance enhancement and waste mitigation, the synergistic potential of combining CB and CR in asphalt binders remains largely unexplored. While substantial volumes of CB are utilized in rubber, inks, coatings, and related industries, a significant fraction of both materials lies unused, accumulating in storage yards as massive stockpiles. Notwithstanding their potential utility, managing surplus CB and CR presents persistent challenges in safe storage, inventory handling, and end-of-life disposal logistics. Thus, the main objective of this study was to optimize the combined dosage of CB and CR for maximizing the synergistic potential of CB-CR conglomerate, chiefly to produce a set of CB-CR-modified asphalt binders with superior rheological properties and enhanced environmental sustainability. It is envisioned that incorporating CB and CR into asphalt binders will help understand the potential benefits, including improvement in pavement durability and performance across a broad spectrum of service temperatures. The incorporation of CB increases high-temperature stiffness and reduces temperature susceptibility, thereby enhancing rutting resistance, whereas CR imparts elasticity and energy dissipation. Furthermore, the use of CB and CR derived from waste tires is envisioned to promote environmentally responsible construction by repurposing a significant volume of waste that would otherwise be landfilled, thus exemplifying waste-to-wealth and circular economy principles.

## 2 Materials and methodology

### 2.1 Materials

In this study, VG-10 (internationally designated as AC-10) asphalt binder, having a specific gravity of 1.020 was adopted as the base asphalt binder and procured from a local refinery. Pyrolysis-derived CB with a particle size of 75  $\mu\text{m}$  and specific gravity of 1.800, and CR with a particle size of 300  $\mu\text{m}$  and specific gravity of 1.150 were also procured locally.

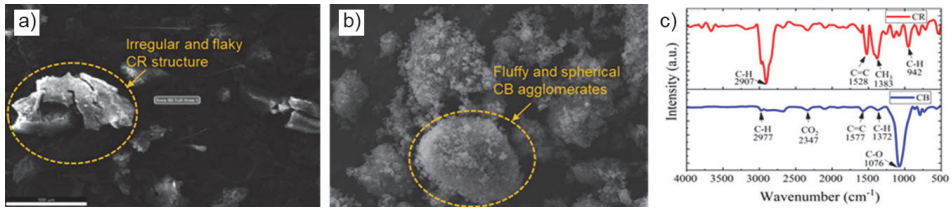
## 2.2 Methodology and experimental program

CB-CR-modified asphalt binders were prepared through the wet-process technique, which involved adding the modifier to the asphalt binder. The base asphalt binder was initially heated to 160-165°C, after which the combined blending of CB and CR in the asphalt binder was carried out in combinations of 0:100, 25:75, 50:50, 75:25, and 100:0 for the fixed additive dosage of 20% by weight of the base asphalt binder. For instance, a CB:CR ratio of 75:25 signifies that CB constitutes 75% (15% by weight of the asphalt binder) and CR constitutes 25% (5% by weight of the asphalt binder) of the total dosage of 20% modifier. The blending was conducted using a high-shear mixer operating at 1000 revolutions per minute (rpm). Following a pre-heating period of 10 min, the required quantity of CB and CR was gradually introduced into the asphalt binder, and mixing was continued for an additional 50 min to ensure uniform dispersion resulting in a total blending equaling 60 min. The resulting CB-CR-modified asphalt binders underwent fundamental consistency tests and rheological analysis to systematically evaluate their synergistic effects on consistency and performance. The physicochemical characteristics of the raw materials (CB and CR) were evaluated using Scanning Electron Microscopy (SEM) and Fourier Transform Infrared Spectroscopy (FTIR) techniques. SEM analysis was performed to examine surface morphology and dispersion characteristics of the materials. FTIR spectroscopy conducted over 4000-400  $\text{cm}^{-1}$  identified functional groups and assessed physicochemical interactions. Further, Penetration, softening point (SP) using ring-and-ball apparatus, and ductility tests were carried out following the American Society for Testing and Materials (ASTM) International standards to determine the consistency, temperature susceptibility, and elongation properties of the unmodified and modified asphalt binders. Rotational viscosity of the binder was measured using a rotational viscometer in accordance with ASTM D4402 [13]. Further,  $A_1$ -VTS<sub>i</sub> relationship following the ASTM procedure [14] was utilized to determine the mixing and compaction temperatures of all the asphalt binders. In addition, the Penetration Index (PI), which quantifies asphalt binder temperature susceptibility from Penetration and SP was determined. Subsequently, the performance grade (PG) test was carried out to assess the failure temperature of the modified asphalt binders using a Dynamic Shear Rheometer (DSR) with a parallel plate geometry and a 1 mm gap. The Multiple Stress Creep Recovery (MSCR) test was conducted in accordance with ASTM D7405 at the respective PG high temperature of each binder at stress levels of 0.1 kPa and 3.2 kPa to assess recovery and rutting resistance in terms of percent recovery (%R) and non-recoverable creep compliance ( $J_{nr}$ ). To assess phase separation behavior in CB-CR-modified asphalt binders at elevated temperatures, an aluminum tube test was conducted following ASTM D7174. Finally, rankings evaluated the CB-CR-modified binders and optimized dosages using fundamental characteristics, rheological properties, and phase separation results.

## 3 Results and discussion

### 3.1 Physicochemical characterization

The SEM micrographs of CB and CR are presented in figures 1a and 1b. CB exhibited fine, nearly spherical primary particles forming dense agglomerates with a highly porous structure and large specific surface area, which facilitated physical interactions and enhanced its effectiveness as a reinforcing filler in the asphalt matrix, thereby improving rutting resistance. In contrast, CR showed irregular, angular, and flaky particles with rough surfaces containing micro-voids generated during mechanical grinding. The larger particle size and surface texture of CR promoted binder absorption and swelling at elevated temperatures, thus contributing to improved elastic response of the modified binder.

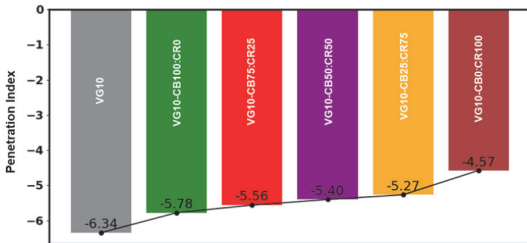


**Figure 1** Physiochemical characterization of the modifiers: a) SEM micrograph of CR, b) SEM micrograph of CB, and c) FTIR spectra of CR and CB

Figure 1c presents the FTIR spectra of CR and CB, where CR exhibited a strong absorption peak at  $2907\text{ cm}^{-1}$  corresponding to aliphatic carbon-hydrogen (C-H) stretching, along with a distinct band at  $1528\text{ cm}^{-1}$  attributed to carbon-carbon double bond (C = C) stretching of unsaturated rubber chains. Additional peak at  $1383\text{ cm}^{-1}$  was attributed to methyl (-CH<sub>3</sub>) bending vibrations, while the band at  $942\text{ cm}^{-1}$  corresponds to out-of-plane C-H vibration, further confirming its polymeric structure. The presence of unsaturated hydrocarbon chains and elastomeric functional groups explained the swelling of CR through absorption of lighter maltene fractions of asphalt binder and its contribution to elastic recovery under cyclic loading. In contrast, CB showed a prominent band at  $1076\text{ cm}^{-1}$  corresponding to carbon-oxygen (C-O) stretching, indicating oxygen-containing surface functional groups that is responsible for enhanced aging resistance. The band at  $1577\text{ cm}^{-1}$ , associated with aromatic C = C stretching, confirmed graphitic domains responsible for increased structural rigidity and stiffness in the asphalt binder matrix.

### 3.2 Fundamental consistency test results

Table 1 presents fundamental consistency test results for the asphalt binders evaluated in this study. The results demonstrated a gradual increase in SP and a progressive reduction in Penetration with the addition of CB-CR to the base asphalt binder. The mechanism was explained by alterations in the maltenes-to-asphaltenes ratio within the asphalt binder. While the base asphalt binder contained a higher proportion of maltenes that promoted molecular mobility, the incorporation of CB and CR absorbed a significant portion of these fractions, reducing fluidity and thereby resisting rutting and indicating improved high-temperature performance. Rotational viscosity increased with CB-CR modification due to CR swelling and CB's filler action, enhancing flow and deformation resistance. All asphalt binders met the Superpave limit of 3000 cP at 135°C except VG10-CB0:CR100, confirming acceptable workability. Further, PI provided a combined measure of the variation of asphalt binder consistency with temperature, which showed a progressive increase with the addition of the modifiers, as shown in figure 2, confirming lower temperature susceptibility of the CB-CR-modified asphalt binders.



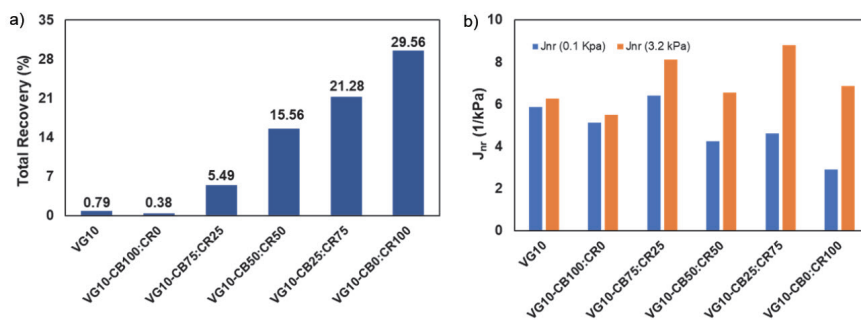
**Figure 2** Penetration index of various asphalt binders with varying CB-CR percentages

**Table 1** Fundamental consistency test results of various asphalt binders

Asphalt Binders	Penetration	SP	Ductility	Viscosity (mPa.s)		
	[0.1 mm]	[°C]	[mm]	120°C	135°C	150°C
VG10	124.7	37.6	100	1200	300	170
VG10-CB100:CR0	85.6	41.2	72.6	2200	600	360
VG10-CB75:CR25	60.1	43.5	35.7	2900	900	520
VG10-CB50:CR50	48.8	45.1	31.2	4100	1700	860
VG10-CB25:CR75	39.1	46.7	18.3	4700	2400	1100
VG10-CB0:CR100	37.2	50.8	16.2	7500	5300	2900

### 3.3 Rheological assessment

The PG results showed a notable increase in failure temperature and corresponding improvement in high-temperature performance for the modified asphalt binder with variation in CB-CR percentages. Relative to the base asphalt binder with a failure temperature of 62.7°C, synergetic CB-CR modification substantially enhanced thermal resilience resulting in the increase of the failure temperature by 9-27%, and upper PG from 58 to 76°C, thereby demonstrating superior resistance to rutting at elevated temperatures. The MSCR results for the base and CB-CR-modified binders are shown in figure 3. The base binder exhibited negligible %R and high  $J_{nr}$ , indicating viscous behavior and permanent deformation susceptibility. CR-dominant blends exhibited highest %R due to elastic recovery, while CB-dominant blends showed stiffness gains without substantial elasticity. The base binder showed high  $J_{nr}$  at both stress levels, indicating permanent deformation susceptibility. At 0.1 kPa,  $J_{nr}$  fluctuations indicated combined influence of stiffness enhancement from CB and elastic deformation introduced by CR within the modified binder conglomerate. Modified blends exhibited consistently higher  $J_{nr}$  at 3.2 kPa, with pronounced fluctuations in balanced CB-CR proportions suggesting stress-dependent elastic network rearrangement under severe loading.



**Figure 3** MSCR results for various asphalt binders: a) %R, b)  $J_{nr}$

### 3.4 Storage stability

The storage stability test using an aluminum tube was conducted, and results revealed a progressive increase in difference in SP between top and bottom sections with varying CB and CR dosages, indicating growing instability and a decline in asphalt binder uniformity. All the modified asphalt binder blends recorded a difference in SP between top and bottom sections of less than 4°C, well within the acceptable limit, confirming adequate storage stability.

However, VG10-CB25:CR75 and VG10-CB0:CR100 exhibited temperature differences of 5.7<sup>o</sup>C and 8.8<sup>o</sup>C respectively, exceeding the allowable threshold of 4<sup>o</sup>C. The poor homogeneity was attributed to excessive CR content, rendering these specific combinations unsuitable for practical use due to inhomogeneity and phase instability.

### 3.5 Optimization through compatibility rankings

Table 2 presents the compatibility ranking of modified asphalt binders using a multiplicative scoring approach that combines individual performance parameter ranks into a composite index, ensuring suboptimal performance in any critical property substantially impacts the overall CB-CR synergy assessment. The parameters used for ranking included PI, viscosity at 135<sup>o</sup>C, failure temperature, %R and J<sub>nr</sub>, and storage stability (difference in SP). Asphalt binders with higher PI, higher failure temperatures, and higher viscosities (up to a threshold value of 3000 cP) were assigned better ranks. For viscosity values beyond 3000 cP, the viscosity was considered detrimental, and a score of zero was assigned to account for inadequate workability. The MSCR results substantiated the improvement in high-temperature deformation resistance, as evidenced by increased %R and reduced J<sub>nr</sub>. Modified asphalt binders with a difference in SP between top and bottom sections exceeding 4<sup>o</sup>C were assigned zero scores, as instability is undesirable for practical applications. It is noteworthy that the VG10-CB50:CR50 achieved the highest overall ranking, as it exhibited balanced improvements in PI, failure temperature, and MSCR parameters, while maintaining viscosity within acceptable limits and satisfactory storage stability.

**Table 2** Parameters used for optimization of CB-CR percentages in the asphalt binder

Asphalt Binder	PI	Viscosity at 135 °C	Failure Temperature	MSCR		Diff. in SP	Ranking
		(mPa.s)	[°C]	%R	J <sub>nr</sub>	[°C]	
VG10	-6.34	300	62.7	0.79	5.87	-	4
VG10-CB100:CR0	-5.78	600	68.7	0.38	5.13	1.6	2
VG10-CB75:CR25	-5.56	900	72.2	5.49	6.39	2.3	3
VG10-CB50:CR50	-5.40	1700	73.6	15.56	4.24	3.4	1
VG10-CB25:CR75	-5.27	2400	76.2	21.28	4.62	5.7	5
VG10-CB0:CR100	-4.57	5300	80.1	29.56	2.89	8.8	6

## 4 Conclusion

The main objective of this study was to optimize the combined dosage of CB and CR for maximizing the synergistic potential of CB-CR conglomerate, chiefly to produce a set of CB-CR-modified asphalt binders with superior rheological properties. The findings of the study elucidated the role of CB and CR as an additive in asphalt binders aimed at advancing sustainable pavement construction. Based on the experimental results, the following are the conclusions:

- Physiochemical characterization: SEM analysis demonstrated that CB possessed a porous reinforcing morphology, whereas CR exhibited rough, swelling-prone structures. FTIR identified unsaturated hydrocarbon groups in CR responsible for improved elastic recovery, while CB displayed aromatic and oxygenated groups contributing to stiffness enhancement and improved aging resistance.

- **Fundamental consistency and rheological test results:** The incorporation of CB-CR increased the PI, indicating reduced temperature susceptibility of the modified binders. The modified asphalt binders exhibited higher viscosity and satisfied Superpave workability limits, except VG10-CB0:CR100, demonstrating improved rutting resistance with acceptable workability. PG test showed a progressive increase in failure temperature, upgrading the upper PG from 58 to 76°C and confirming enhanced high-temperature performance. MSCR results further revealed higher %R and lower  $J_{nr}$  for CB-CR-modified conglomerates, with CR primarily contributing to elastic recovery and CB enhancing stiffness.
- **Storage stability:** All CB-CR-modified asphalt binders showed SP differences below 4°C between top and bottom sections of the aluminum tube, except VG10-CB25:CR75 and VG10-CB0:CR100, which exceeded the limit. The poor homogeneity was attributed to excessive CR content, compromising storage stability and practical applicability.
- **Compatibility rankings:** The compatibility ranking, based on six parameters, identified VG10-CB50:CR50 as the optimum blend, delivering superior high-temperature performance and elastic recovery without sacrificing workability or storage stability.
- **Recommendations:** This preliminary investigation provided valuable insights about the feasibility of incorporating CB and CR into the asphalt binder through the wet process, leading to the production of superior performing CB-CR-modified asphalt binders. However, further research is recommended to assess the low-temperature performance of CB-CR modified binders and rheological behavior at sub-zero temperatures as well as evaluate the compatibility of the CB-CR-modified binder with various asphalt mixture gradations. Overall, the study is expected to contribute to the development of sustainable, high-performance infrastructure solutions by addressing environmental challenges associated with CB and CR.

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