

Rheological Properties of Bitumen Modified with Crumb Rubber and Devulcanized Rubber [†]

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Abstract

The modification of bitumen with recycled rubber materials has gained significant attention due to its potential to enhance pavement performance and support sustainable waste management. In this study, the rheological properties of bitumen modified with crumb rubber (CR) and devulcanized crumb rubber (DCR) were investigated. Rubber modifiers were added at concentrations of 5–25% by weight, and the rheological behavior was evaluated using a Dynamic Shear Rheometer at 1.59 Hz over a temperature range of 46–96 °C. Key parameters, including storage modulus (G'), loss modulus (G''), and complex viscosity (η^*), were analyzed. The results indicate that increasing rubber content significantly enhances stiffness and viscosity, improving resistance to deformation at elevated temperatures. Moreover, DCR-modified binders exhibit higher rheological performance compared to CR systems, indicating better compatibility with the bitumen matrix. Overall, devulcanized rubber demonstrates superior efficiency as a modifier and shows strong potential for improving the durability and high-temperature performance of asphalt binders.

Keywords: bitumen binder; rheological properties; storage modulus; loss modulus; complex viscosity

1. Introduction

Bitumen is widely used as a binder in asphalt pavements due to its adhesive properties and ability to provide flexibility and durability under traffic loading. However, conventional bitumen exhibits significant temperature susceptibility, which leads to rutting at high temperatures and thermal cracking at low temperatures. Therefore, improving the rheological performance of bitumen has become an important objective in pavement engineering research [1–3].

One of the most effective methods for enhancing the performance of bituminous binders is modification with polymeric or rubber-based additives. In recent decades, crumb rubber (CR) derived from waste tires has gained significant attention as a modifier for asphalt binders. The use of crumb rubber not only improves the mechanical and rheological properties of asphalt binders but also contributes to sustainable waste management by recycling end-of-life tires [4–6]. Previous studies have shown that the incorporation of



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crumb rubber can significantly increase the elasticity, stiffness, and resistance of asphalt binders to permanent deformation and fatigue damage [7,8].

The interaction between crumb rubber particles and bitumen mainly occurs through swelling and absorption of light aromatic fractions from the binder. This swelling process results in an increase in viscosity and the formation of a rubber-bitumen network structure that improves the viscoelastic properties of the binder [9,10]. However, conventional crumb rubber modification also has some limitations, including high processing temperatures, poor compatibility with bitumen, and storage stability problems caused by phase separation between rubber particles and the asphalt matrix [11].

To overcome these limitations, devulcanized crumb rubber (DCR) has recently been proposed as an alternative modifier for asphalt binders. Devulcanization is a process that partially breaks the sulfur cross-links in vulcanized rubber, producing a more reactive material with improved molecular mobility and compatibility with bitumen [12,13]. As a result, devulcanized rubber particles can disperse more uniformly in the bitumen matrix and interact more effectively with its components.

Several studies have reported that the use of devulcanized rubber can significantly improve the rheological performance and storage stability of modified asphalt binders compared to conventional crumb rubber modification [14–16]. In particular, devulcanized rubber has been shown to enhance the elasticity of the binder, increase complex viscosity, and improve resistance to permanent deformation at high service temperatures [17].

The rheological behavior of modified bitumen is commonly evaluated using a Dynamic Shear Rheometer (DSR), which measures key viscoelastic parameters such as storage modulus, loss modulus, and complex viscosity. These parameters provide valuable insight into the elastic and viscous components of binder behavior and their response to temperature and loading conditions [18–20].

Therefore, the objective of this study is to investigate the rheological properties of bitumen modified with crumb rubber (CR) and devulcanized crumb rubber (DCR) and to evaluate the influence of rubber content and temperature on the viscoelastic characteristics of the modified binders.

2. Materials and Methods

2.1. Materials

Base paving-grade bitumen was used as the binder material in this study. The modification of the binder was carried out using two types of rubber modifiers: crumb rubber (CR) and devulcanized crumb rubber (DCR) material supplied by “Shinline” JSC, Almaty, Kazakhstan. Crumb rubber was obtained from recycled waste tires through mechanical grinding processes, while devulcanized rubber was produced by partially breaking the sulfur cross-links in vulcanized rubber to improve compatibility with the bitumen matrix [21].

Rubber modifiers were incorporated into the base bitumen at concentrations of 5%, 10%, 15%, 20%, and 25% by weight of the binder. The modified binders were prepared using a high-temperature mixing process to ensure uniform dispersion of rubber particles in the bitumen [21].

The purpose of using both crumb rubber and devulcanized rubber was to compare the rheological behavior of conventional rubber-modified bitumen and binders modified with devulcanized rubber, which is expected to exhibit improved interaction with the bitumen matrix.

2.2. Preparation of Rubber-Modified Bitumen

The rubber-modified binders were prepared by mixing the base bitumen with the rubber modifier at elevated temperature. The base bitumen was first heated to the required

mixing temperature and then the rubber particles were gradually added to the binder under continuous stirring [21].

The mixing process was continued until a homogeneous mixture was obtained. Different rubber concentrations were used to investigate the influence of modifier content on the rheological properties of the binder.

2.3. Rheological Testing

The rheological properties of the modified binders were evaluated using a Dynamic Shear Rheometer (DSR). The tests were performed at a constant oscillation frequency of 1.59 Hz, which corresponds to typical traffic loading conditions.

Rheological properties were determined using an Anthon Paar SmartPave 102 Dynamic Shear Rheometer (DSR) (Anton Paar GmbH, Graz, Austria), with a 25-plate configuration at 10 rad/s oscillation and 12% shear strain. The test procedure, according to EN 14770 [22], involves determining the viscoelastic properties of bitumen (including complex shear modulus G^* and phase shift angle δ) at different temperatures, which are gradually increased.

Three main rheological parameters were measured:

- Storage modulus (G')—characterizing the elastic component of binder behavior;
- Loss modulus (G'')—describing the viscous component of deformation;
- Complex viscosity (η^*)—indicating the overall resistance of the binder to deformation.

The tests were conducted at six different temperatures: 46 °C, 56 °C, 66 °C, 76 °C, 86 °C, and 96 °C.

These temperatures were selected to represent the typical temperature range experienced by asphalt pavements during service.

3. Results and Discussion

3.1. Storage Modulus Analysis

The storage modulus (G') describes the elastic component of the binder response and reflects the ability of the material to recover after deformation. The temperature dependence of the storage modulus for CR- and DCR-modified binders is presented in Figure 1.

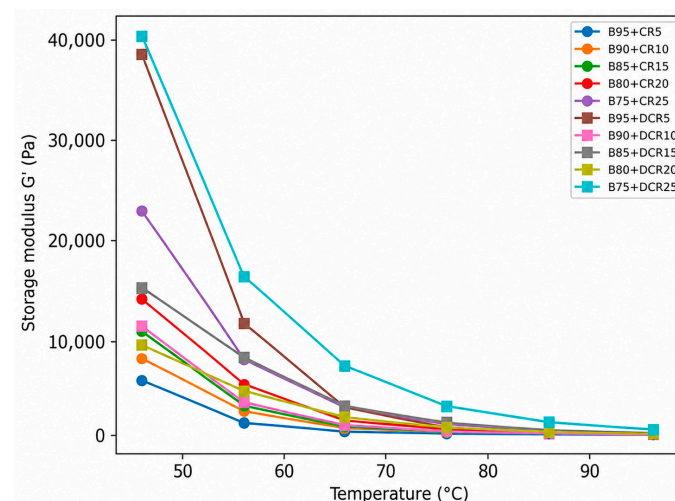


Figure 1. Temperature dependence of storage modulus G' .

The results show a strong decrease in the storage modulus with increasing temperature. For example, the binder containing 5% crumb rubber exhibits a storage modulus of approximately 5610 Pa at 46 °C, which decreases to about 5.9 Pa at 96 °C. This dramatic reduction demonstrates the temperature sensitivity of bituminous materials.

However, increasing rubber content significantly enhances the storage modulus values. At 25% crumb rubber content, the storage modulus reaches 23,100 Pa at 46 °C, which is more than four times higher than the value for the binder containing 5% rubber. This increase indicates that rubber particles contribute to the formation of a stronger elastic network within the binder.

Devulcanized rubber modified binders show even higher storage modulus values in several cases. For example, the binder containing 25% DCR exhibits a modulus of 41,400 Pa at 46 °C, indicating significantly enhanced stiffness and elastic response.

These findings are consistent with previous studies, where an increase in storage modulus with rubber content was attributed to the swelling of rubber particles and the formation of a reinforcing elastic network within the bitumen matrix [5,7]. In addition, devulcanized rubber has been reported to provide improved dispersion and compatibility with bitumen, resulting in higher stiffness and enhanced viscoelastic response compared to conventional crumb rubber systems [15].

These results demonstrate that rubber modification improves the elastic component of the binder behavior and increases resistance to deformation under loading.

3.2. Loss Modulus Analysis

The loss modulus (G'') represents the viscous component of the binder response and reflects the ability of the material to dissipate energy during deformation. The variation in the loss modulus with temperature is shown in Figure 2.

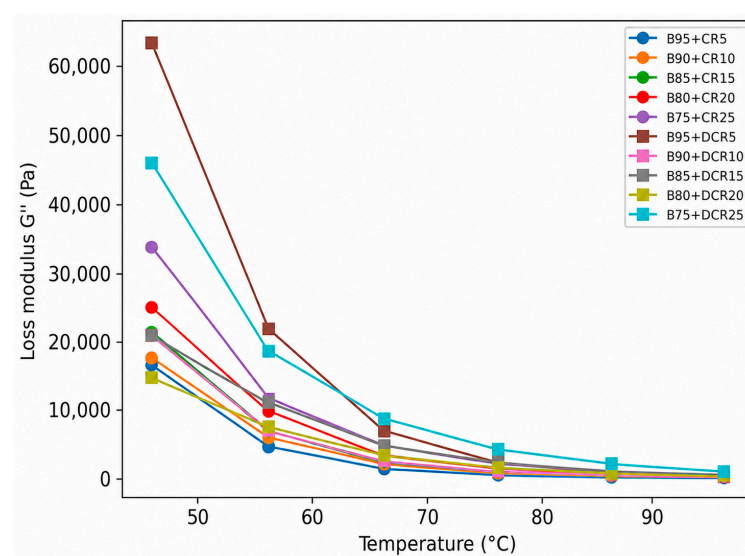


Figure 2. Temperature dependence of loss modulus G'' .

Similar to the storage modulus, the loss modulus decreases with increasing temperature for all binder compositions. This behavior is related to the softening of the binder structure as temperature increases.

For instance, the binder with 5% crumb rubber exhibits a loss modulus of approximately 16,700 Pa at 46 °C, which decreases to about 96 Pa at 96 °C.

Increasing rubber content leads to higher loss modulus values. The binder with 25% crumb rubber shows a loss modulus of about 34,000 Pa at 46 °C, indicating significantly improved viscous resistance.

In the case of devulcanized rubber modification, even higher values are observed. The binder with 25% DCR reaches a loss modulus of approximately 46,400 Pa at 46 °C, which demonstrates the enhanced energy dissipation capacity of DCR-modified binders.

These results are consistent with previous studies, where the increase in loss modulus with rubber content was attributed to enhanced viscous response caused by the swelling and partial dissolution of rubber particles in the bitumen matrix [5,10]. Furthermore, devulcanized rubber has been reported to improve interfacial interaction and dispersion within the binder, leading to higher energy dissipation capacity compared to conventional crumb rubber systems [15].

3.3. Complex Viscosity Analysis

Complex viscosity (η^*) is an important rheological parameter that characterizes the resistance of the binder to flow under oscillatory loading. The temperature dependence of complex viscosity is presented in Figure 3.

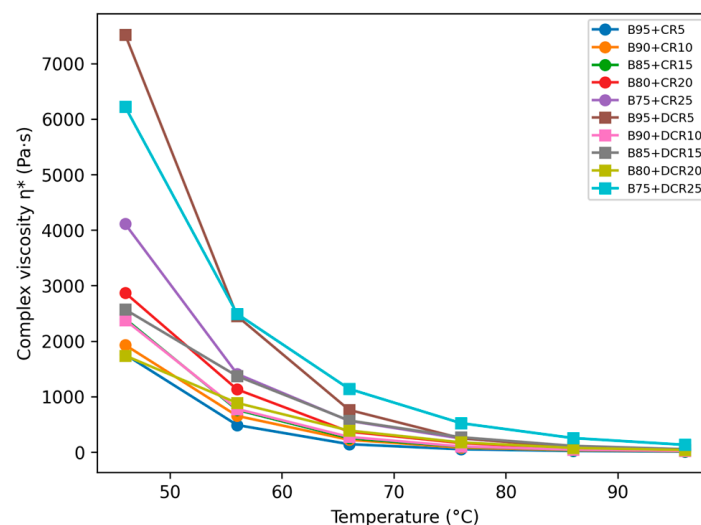


Figure 3. Temperature dependence of complex viscosity η^* .

As expected, the complex viscosity decreases with increasing temperature for all binder compositions. This decrease reflects the reduction in binder stiffness and increased molecular mobility at higher temperatures.

For example, the binder containing 5% crumb rubber shows a viscosity of approximately 1760 Pa·s at 46 °C, which decreases to about 9.7 Pa·s at 96 °C.

Increasing rubber concentration significantly increases viscosity values. The binder with 25% crumb rubber exhibits a viscosity of about 4110 Pa·s at 46 °C, indicating improved resistance to flow and deformation.

In the case of devulcanized rubber modification, even higher viscosity values are observed. The binder containing 25% DCR reaches approximately 6220 Pa·s at 46 °C, demonstrating stronger structural reinforcement of the binder.

These findings are consistent with previous studies, where the increase in complex viscosity with rubber content is associated with the swelling of rubber particles and the formation of a more structured binder network that restricts flow behavior [5,7]. Moreover, devulcanized rubber has been reported to enhance compatibility with bitumen and promote a more homogeneous microstructure, resulting in higher viscosity values compared to conventional crumb rubber systems [16].

3.4. Influence of Rubber Content

The influence of rubber concentration on the rheological properties of the binder is shown in Figures 4 and 5.

The results clearly indicate that increasing rubber content leads to a significant increase in both storage modulus and viscosity. This trend suggests that rubber particles contribute to the formation of a reinforcing network structure within the bitumen matrix.

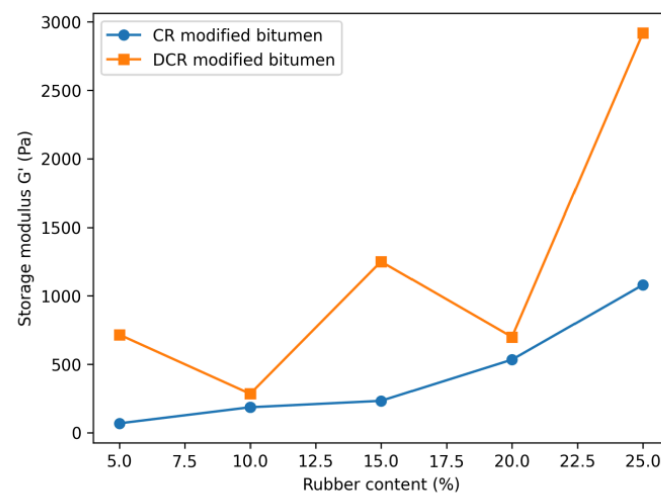


Figure 4. Effect of rubber content on storage modulus at 76 °C.

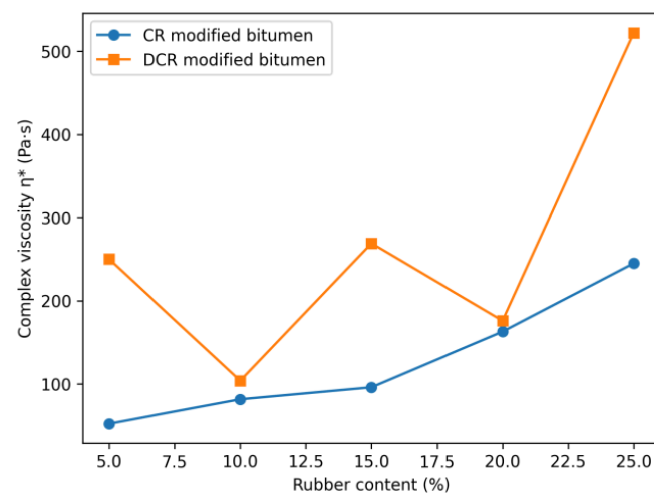


Figure 5. Effect of rubber content on complex viscosity at 76 °C.

The most pronounced increase in rheological properties is observed at rubber contents above 20%, indicating that higher rubber concentrations significantly enhance binder stiffness and deformation resistance, indicating improved resistance to rutting and permanent deformation.

These results are consistent with previous studies, where the increase in rheological properties at higher rubber contents is attributed to the formation of a continuous rubber-bitumen network structure, which enhances stiffness and deformation resistance [4,5]. In particular, it has been reported that rubber contents above 20% lead to a transition from dispersed particles to an interacting network, significantly improving high-temperature performance and rutting resistance [4,7].

3.5. Comparison of CR- and DCR-Modified Binders

A direct comparison between crumb rubber- and devulcanized rubber-modified binders is presented in Figure 6.

The results show that devulcanized rubber modification generally provides higher rheological parameters compared to conventional crumb rubber modification. This behav-

ior can be explained by improved compatibility between devulcanized rubber particles and the bitumen matrix.

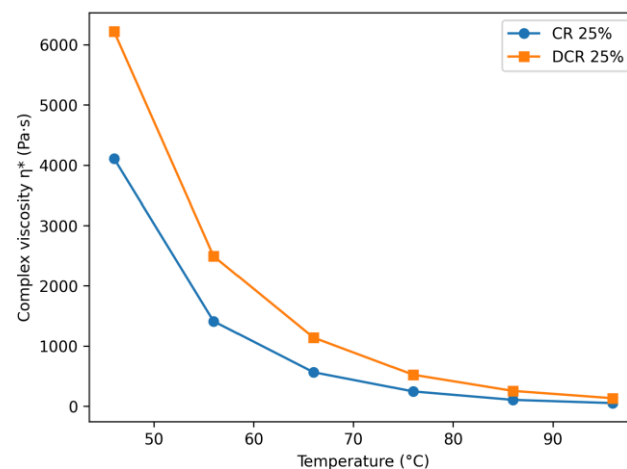


Figure 6. Comparison of complex viscosity of CR- and DCR-modified bitumen (25%).

The devulcanization process partially breaks sulfur cross-links in rubber molecules, allowing better dispersion of rubber particles in the binder. As a result, DCR-modified binders exhibit improved structural integrity and enhanced viscoelastic properties.

These findings are in good agreement with previous studies, which report that devulcanized rubber exhibits superior compatibility with bitumen due to the partial breakdown of sulfur cross-links and increased molecular mobility [12,13]. This structural modification facilitates better dispersion of rubber particles within the bitumen matrix and enhances interfacial interactions, resulting in improved viscoelastic performance [15]. In contrast, conventional crumb rubber retains a highly cross-linked structure, which limits its interaction with the binder and may lead to phase separation and reduced efficiency of modification [4]. Furthermore, several studies have shown that DCR-modified binders exhibit higher complex viscosity and storage modulus values, particularly at high temperatures, indicating improved resistance to rutting and permanent deformation [15,16]. The enhanced rheological behavior observed in DCR systems can therefore be attributed to the formation of a more homogeneous and interconnected microstructure compared to CR-modified binders. Overall, the results confirm that devulcanized rubber provides a more effective modification mechanism, leading to improved structural integrity and long-term performance of asphalt binders.

4. Conclusions

This study investigated the rheological behavior of bitumen modified with crumb rubber and devulcanized crumb rubber using a Dynamic Shear Rheometer.

The main conclusions are as follows:

1. The storage modulus, loss modulus, and complex viscosity of modified binders decrease with increasing temperature due to the thermally sensitive nature of bitumen.
2. Increasing rubber content significantly improves the rheological properties of the binder.
3. The storage modulus increases from 5610 Pa to 23,100 Pa when crumb rubber content increases from 5% to 25% at 46 °C.
4. Devulcanized rubber-modified binders exhibit higher stiffness, with storage modulus reaching 41,400 Pa at 46 °C for 25% DCR.
5. The complex viscosity increases from 1760 Pa·s to 4110 Pa·s when rubber content increases from 5% to 25%.

6. Devulcanized rubber modification provides superior rheological performance compared with conventional crumb rubber modification due to improved compatibility with the bitumen matrix.

Overall, the results demonstrate that rubber modification significantly enhances the high-temperature performance of bitumen, and devulcanized rubber represents a promising modifier for producing high-performance asphalt binders.

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