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Physical Properties and Rheological Characteristics of Cigarette Butt-Modified Asphalt Binders

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Abstract: Cigarette butt (CB) waste is abundant and difficult to biodegrade, which is dangerous for both the environment and human health. The key reason CBs are littered is that people do not know much about the harm CBs pose to the environment. Recycling CBs in infrastructure construction can help raise people's awareness. To promote the recycling of CB waste, this paper aimed to determine the feasibility of using CBs as a modifier for asphalt binders. In this research, CBs were preprocessed and mixed with virgin asphalt binder as a fiber modifier. Comprehensive laboratory investigations, including a softening point test, viscosity test, storage stability test, and temperature sweep test, were performed, along with a frequency sweep test, to evaluate the performance of the modified samples. During this investigation, samples were prepared with 1%, 2%, 3%, and 4% CBs. The results of the CB-modified samples were compared with the sample consisting of fresh bitumen (0% fiber). The results show that the physical and rheological properties of bitumen with incorporated CBs improved significantly, and CBs could be used instead of virgin cellulose fiber as a fiber modifier. However, CB-modified asphalt reduced the storage stability and low-temperature performance of the samples. Further research should focus on improving the storage stability and low-temperature performance of CB-modified asphalt binders to facilitate their application in asphalt pavements.

Keywords: road engineering; cigarette butts; modified asphalt; physical properties; rheological characteristics

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1. Introduction

Although smoking is harmful to health, a great number of cigarettes are produced and consumed worldwide. According to statistics, China produced about 2.4 trillion cigarettes in 2022. The World Health Organization reported that almost 340 to 680 million kilograms of cigarette butts (CBs) are discarded annually worldwide [1,2]. In China, CBs are commonly thrown away, incinerated, or buried along with other garbage. Since some hazardous substances are contained in CBs, the improper disposal of CBs can pose a severe environmental threat. Therefore, it is highly important to recycle and utilize CB waste.

In general, CBs consist of unburned tobacco, filters, ash, and paper wrap [3]. Among them, the filters, which are composed of plasticized cellulose acetate and polypropylene fiber bundles, are notoriously hard to decompose [3–5]. Poisonous substances are usually kept in the filters [4–8]. In recent decades, research has been conducted on recycling CBs in various fields, such as the paper industry [9], activated carbon preparation [10], sound absorption agent preparation [11], clay brick preparation [12], and more. However, the total amount of CBs recycled is still limited. Therefore, it is necessary to expand the utilization strategies of CB waste, which could bring considerable benefits in terms of both environmental and economic aspects [13–17].

In addition, asphalt binders are a typical viscoelastoplastic material. Hence, asphalt pavement is prone to permanent deformation at high temperatures and cracks at low temperatures. High-performance or modified asphalt binders are required to ensure the durability of asphalt pavement. Furthermore, as the economy and society develop, there is typically an increase in the volume and weight of traffic. Therefore, various modifiers or additives have been used to improve pavement's performance and the durability of asphalt binders and pavement. The cellulose acetate in CBs is a thermosetting material [18]. It is worth trying to modify asphalt with recycled CBs.

In previous studies, CBs were encapsulated in asphalt or wax and mixed with asphalt mixtures. The test results indicated that bitumen-encapsulated CB-modified asphalt is resistant to permanent deformation at high temperatures and cracking at low temperatures [19–21]. Furthermore, limited research has been conducted to explore using recycled CBs as an additive in asphalt mixtures. Liu, H. (2021) derived recycled cellulose diacetate (rCDA) from CBs. rCDA has been used as a fiber stabilizer of stone mastic asphalt (SMA) mixtures [3]. Similarly, Piergiorgio Tataranni (2021) used shredded CBs as stabilizing fibers for stone mastic asphalts [22]. The optimal CB content was only discovered and recommended by observing rutting and cracking in rheological property tests [23]. It has been proven that bitumen can seal off harmful substances to some extent. Recycled cigarette filter waste can be used as an eco-friendly alternative to common modifiers typically used in asphalt mixtures. We know that previous research on CBs in asphalt mixtures mainly involved encapsulating the butts with paraffin or asphalt. The encapsulated butts were then used to replace a portion of the aggregate in the mixture. Experiments have shown that asphalt-encapsulated CBs used in asphalt pavement exhibit good resistance to high-temperature permanent deformation and low-temperature cracking. Encapsulation also helps control the leaching of toxic substances from the CBs. However, the preparation process for encapsulated butts is relatively complex and may pose difficulties in practical engineering applications. Subsequent studies mainly focused on extracting cellulose acetate fibers from CBs and using them as fiber stabilizers in SMA mixtures. Studies have also directly shredded CBs and used them as fiber stabilizers. Although CBs have been proven to be an environmentally friendly asphalt mixture modifier, these studies primarily concentrated on the performance testing of asphalt mixtures. Few studies have focused on how CBs affect the physical and rheological properties of asphalt binders.

This paper aimed to promote the application of recycled CBs in road engineering through more comprehensive tests of CB-modified asphalt, such as physical and rheological tests. Firstly, the CBs were collected and disinfected, and then, the modified asphalt binders with different contents of CBs were prepared via high-speed shearing and stirring equipment. Secondly, the samples required for the test were prepared separately. Then, the penetration test, softening point temperature test, ductility test, viscosity test, storage stability test, temperature sweep test, and frequency sweep test were carried out to investigate the effect of CBs on the properties of asphalt binder, providing a reference for the application of CBs in road construction.

2. Materials and Methods

2.1. Materials

2.1.1. Asphalt

In this study, the virgin asphalt binder was provided by Star-Energy (China) Co., Ltd., Ningbo, China. It has a penetration of 84.7 (0.1mm), a softening point temperature of 45.8 °C, and a ductility (25 °C) exceeding 100 cm.

2.1.2. Preparation of CB Modifier

Discarded CBs were collected and provided by a community in Chongqing, China. Figure 1 presents the pretreatment procedure for the CB waste in the laboratory. Firstly, the gathered CBs were treated with ultraviolet-light disinfection. Then, the CBs were put in a high-speed mixer with a shearing rate of 500 rpm for 1 h. Finally, the CB additive was obtained to prepare the modified asphalt binder.

(**a**) Recycled CBs (**b**) CBs in shearing (**c**) CB additive

Figure 1. Laboratory process of preparing CBs.

2.1.3. Preparation of CB-Modified Asphalt Binder

The preparation of the CB-modified asphalt binder is shown in Figure 2**,** which can be divided into three steps.

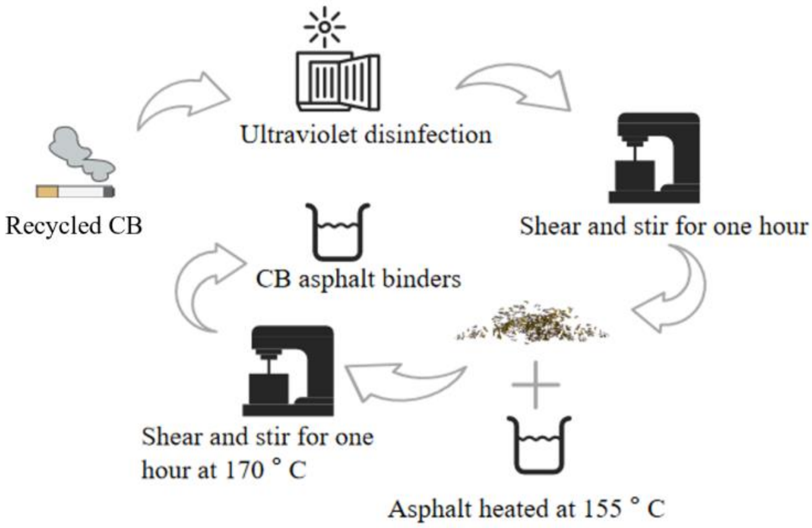

Figure 2. Laboratory process of preparing CB-modified asphalt binders.

Step 1: The virgin asphalt binder was heated to 155 °C to obtain a fluid state.

Step 2: The CB modifier was added to the asphalt binder. The CB contents were 1%, 2%, 3%, and 4% by the weight of the asphalt binder. In subsequent research, three samples were provided for each percentage of CB-modified asphalt.

Step 3: The CB-modified asphalt binders were heated to 170 °C and mixed using high-speed shearing equipment. The shearing rate was 4500 r/min, and the shearing time was 60 min.

Finally, the CB-modified asphalt binder was obtained and used for the physical and rheological property tests.

2.2. Experimental Methods

2.2.1. Physical Property Tests

Firstly, the penetration, softening point temperature, and ductility of the CB-modified asphalt binders were tested according to the Chinese specifications JTG E20-2011 T0604, T0606, and T0605 [24], respectively. The penetration test was performed at three temperature levels (5, 15, and 25 °C) to investigate the temperature susceptibility of the CB-modified asphalt binders. The ductility test was conducted at a temperature of 15 °C and a loading rate of 50mm/min.

2.2.2. Viscosity Test

The viscosity of the CB-modified asphalt binders was tested at three temperature levels (115, 135, and 175 °C) using the Brookfield viscometer method according to the Chinese specification JTG E20-2011 T0625 [24].

2.2.3. Storage Stability Test

The storage stability of the CB-modified asphalt binders was studied according to the Chinese specification JTG E20-2011 T0661 [24]. Firstly, the modified asphalt binders were heated and put into aluminum tubes with a diameter of 25 mm and a height of 150 mm. Secondly, the tubes were placed in an oven of 163 °C for 48 h and then in a refrigerator at −18 °C for 4h. Finally, each tube was cut into three equal pieces. The bottom and top samples were adopted to conduct the softening point temperature test, and their difference values were used to evaluate the storage stability of the asphalt binders.

2.2.4. Temperature Sweep Test

In this experiment, both high-temperature and low-temperature sweep tests were employed to assess the rheological properties of the CB-modified asphalt binders. The complex modulus (G*) and phase angle (δ) of the modified asphalt binders were obtained using a dynamic shear rheometer (DSR). The test temperatures ranged from −10 to 80 °C within an increment of 1 °C, and the test frequency was 10 rad/s. In addition, the rutting and cracking factors were generated to evaluate the high- and low-temperature performances, respectively.

2.2.5. Frequency Sweep Test

The primary curves of the complex modulus for the CB-modified asphalt binders were generated using the frequency sweep test. The test temperatures were −10, 5, 20, 35, and 50 °C, and the test frequencies ranged from 0.1Hz to 10Hz. Then, the Williams–Landel–Ferry equation was used to establish the primary curves of the complex modulus at a reference temperature of 20 °C.

3. Results and Discussion

3.1. Physical Properties

Figure 3 shows the penetration of the CB-modified asphalt binders at different temperatures (5, 15, and 25 °C). Penetration can represent the ability of asphalt to resist the shear-loading effect. The asphalt binder with lower penetration showed better stability under loading. As seen in Figure 3, the penetration values of the asphalt binders decreased with the CB modifier content at the same temperature. When the content of the CB additive increased from 0% to 4%, the penetration value of the asphalt binder decreased from 84.7 to 76.0 dmm at 25 °C. The penetration of the asphalt binders decreased by 7.6%, 8.5%, 9.1%, and 10.3% when the CB contents were 1%, 2%, 3%, and 4%, respectively. This is because the acetate fibers in the CBs had certain adsorption properties [25,26], which adsorbed the light components in the asphalt, thereby increasing the viscosity and hardness of the asphalt. It can be concluded that the CB modifier improved the asphalt binder’s ability to resist the shearing loading effect.

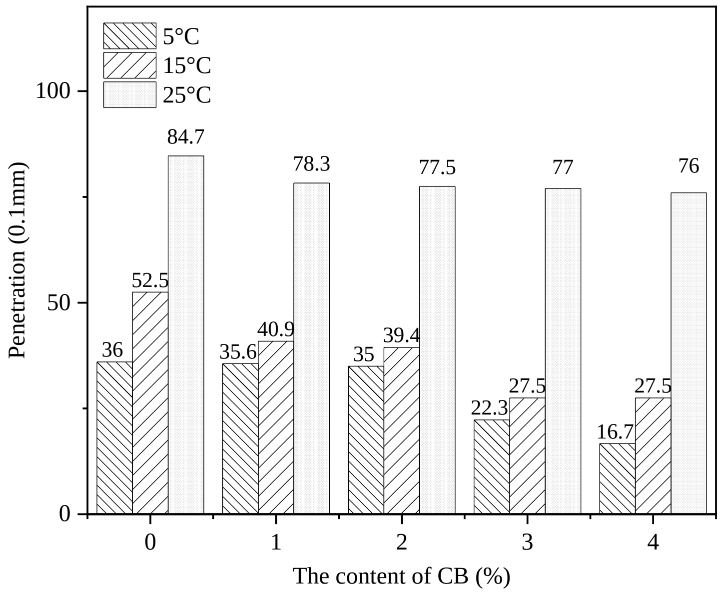

Figure 3. Penetration test results of CB-modified asphalt.

Figure 4 shows the softening point temperature and ductility test results of the CB-modified asphalt binders. The softening point temperature can represent the high-temperature stability of the asphalt binder. As seen in Figure 4, the softening point temperature of the CB-modified asphalt increased with the CB content. The softening point temperatures of the CB-modified asphalt binders were 45.8, 47., 49.7, 52.2, and 53.4 °C when the CB contents were 0%, 1%, 2%, 3%, and 4% CB, respectively. This implies that the CB modifier improved the high-temperature stability of the asphalt binders.

In addition, the ductility of the asphalt binders at 15 °C decreased with the CB content. The ductility of the asphalt binders showed a sudden drop when the CBs were added. This implies that the CB modifier degraded the flexibility of the asphalt binder, which might have increased the risk of temperature-related cracking for asphalt pavement. Asphalt binders containing a CB additive are similar to asphalt mortar, which consists of asphalt binders and mineral powder. A ductility test might not be suitable to evaluate the low-temperature property of CB-modified asphalt binders. Therefore, further studies should be conducted to better understand the low-temperature performance of CB-modified asphalt binders and mixtures.

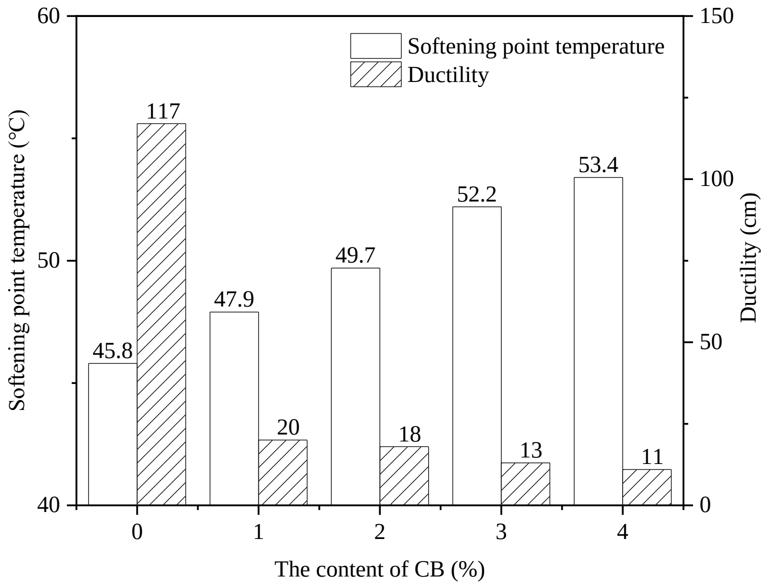

Figure 4. Softening point temperature and ductility test results of CB-modified asphalt.

CBs are mostly composed of acetate fibers with a relatively strong adsorption capacity. The powder of CBs has a certain adsorption capacity for asphalt, reducing the light fractions in the asphalt and thereby increasing the viscosity and hardness of the asphalt, decreasing its temperature sensitivity [25,26]. As a result, the softening point of asphalt shows a significant upward trend. This indicates that CBs enhance the high-temperature deformation resistance of asphalt. With the increase in hardness of the asphalt modified by CBs, its brittleness also increases, leading to a reduction in ductility. This suggests that CBs impair the low-temperature performance of asphalt.

3.2. Brinell Viscosity

The effect of the CB modifier on the viscosity of the asphalt binders was investigated, and the test results are shown in Figure 5. Greater viscosity means the asphalt binder is less prone to shearing deformation under the loading effect. As seen in Figure 5, the viscosity of the asphalt binders increased with the CB content at the same temperature. When the temperature was 135 °C, the viscosity values of the asphalt binders were 460.3 mPa·s, 535.7 mPa·s, 580.4 mPa·s, 728.2 mPa·s, and 771.8 mPa·s when the CB contents were 0%, 1%, 2%, 3%, and 4% CB, respectively. When the content of CBs was more than 3%, the viscosity of the CB asphalt increased significantly at 135 °C, exceeding 58%. It can be concluded that the CB modifier improved the resistance of the asphalt binders to deformation.

It is recognized that the construction temperature of the asphalt mixture strongly depends on the viscosity of the asphalt binder. The greater the viscosity, the higher the mixing temperature and compaction temperatures. Because the CB modifier increased the viscosity of the asphalt binder, it should be noted that the construction temperatures of the CB-modified asphalt mixtures were relatively higher than that of the virgin asphalt binder. According to the viscosity–temperature curves, the mixing temperatures of the mixtures were 168–173 °C for the matrix asphalt and 173–176 °C for the 4% CB asphalt, whose temperature was increased by about 5 °C. The compaction temperatures of the mixtures were as follows: that of the matrix asphalt was 154–161 °C, while that of the 4% CB asphalt was 165–169 °C, increasing by about 10 °C. Therefore, the CB content should be controlled at a certain amount in the admixture to reduce the energy consumed during construction mixing.

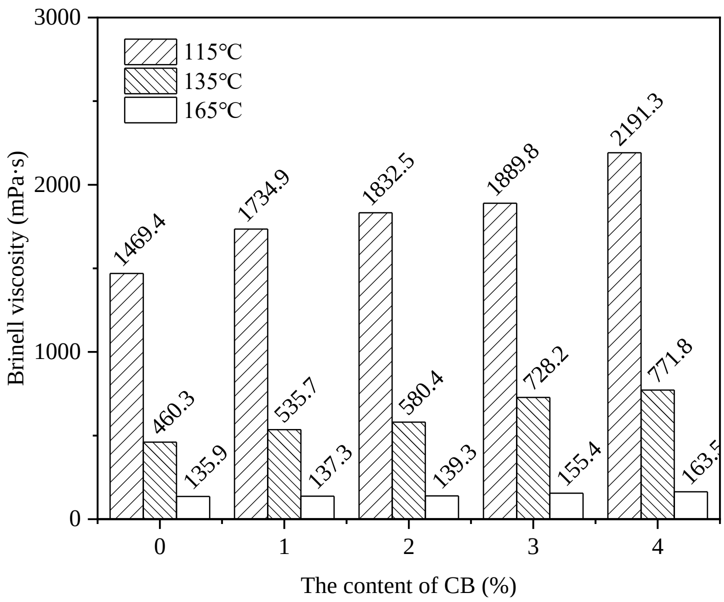

Figure 5. Brinell viscosity test results of CB-modified asphalt.

3.3. Storage Stability

Figure 6 illustrates the storage stability test results of the CB-modified asphalt binders. The greater the difference value of the softening point temperature between the upper and lower tubes, the worse the storage stability of the modified asphalt binder. As shown in Figure 6, for the virgin asphalt binder, the difference value of the softening point temperature was only 0.2 °C, signifying the best storage stability. However, the difference values of the softening point temperature for the modified asphalt binders increased with the CB content. When the CB content was 4%, the difference values of the softening point temperature increased to 6.6 °C. Since the density of the CB modifier was lower than that of the asphalt binder, the CB modifier was expected to float upward during the storage process at a relatively high temperature. Consequently, the softening point temperature of the upper section was greater than that of the bottom section. The difference value of softening point temperature became more significant with the CB content, implying that CBs would degrade the storage stability of asphalt binder.

The main reason is that asphalt materials can be divided into asphaltene, resin, saturate, and aromatic components. There are strong interactions between asphaltene and resin components and between resin and light components. Asphaltenes adsorb resins to form colloidal nuclei and adsorb light components, dispersing in the dispersion system composed of light components [27]. After CBs are compatible with asphalt, they show stronger interactions with light components, and these interactions increase with the addition of CBs. However, compared with heavy components, such as resins, CBs have weaker adsorption of light components, which means they can only adsorb a small number of light components and hardly form strong cross-linked structures. Therefore, the asphalt adhesives modified by CBs showed poor early storage stability and segregation.

According to the Chinese specification JTG F40-2004 [28], the difference value of the softening point temperature should be less than 2.5 °C. When the CB content was 2%, the difference value of the softening point temperature increased to 3.6 °C. Currently, the storage stability of asphalt binders cannot meet the Chinese specification requirements. Therefore, further study is needed to focus on improving the storage stability of CB-modified asphalt binder, which will promote its engineering application.

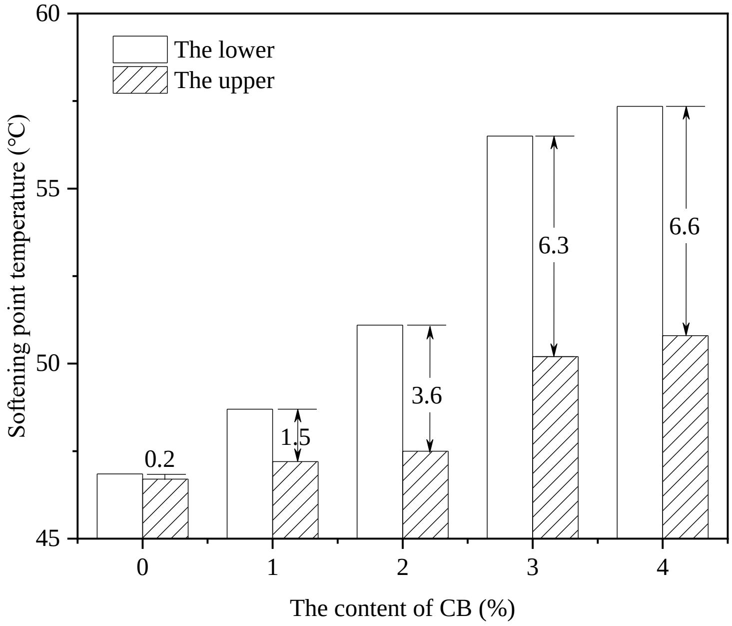

Figure 6. Storage stability performances of different CB-modified asphalt samples.

3.4. Temperature Sweep Tests

Figures 7 and 8 show the complex modulus (G*) and phase angle (δ) values for the CB-modified asphalt binders when the temperature regions were 30–80 °C and −10–30 °C, respectively. The complex modulus represents the stiffness of asphalt binder. The greater the complex modulus, the less prone the asphalt binder is to deformation. The tangent value of the phase angle is the proportion of the viscous part of the asphalt binder to the elastic. The greater the phase angle, the more susceptible the asphalt binder is to permanent deformation. As seen in Figures 7 and 8, the complex moduli of all the CB-modified asphalt binders showed a decreasing trend within the whole temperature region, while the phase angles tended to increase. At the same temperature, the complex modulus increased with the CB content while the phase angle decreased. It can be concluded that the CB modifier had a strengthening effect on the asphalt binder and decreased the risk of permanent deformation.

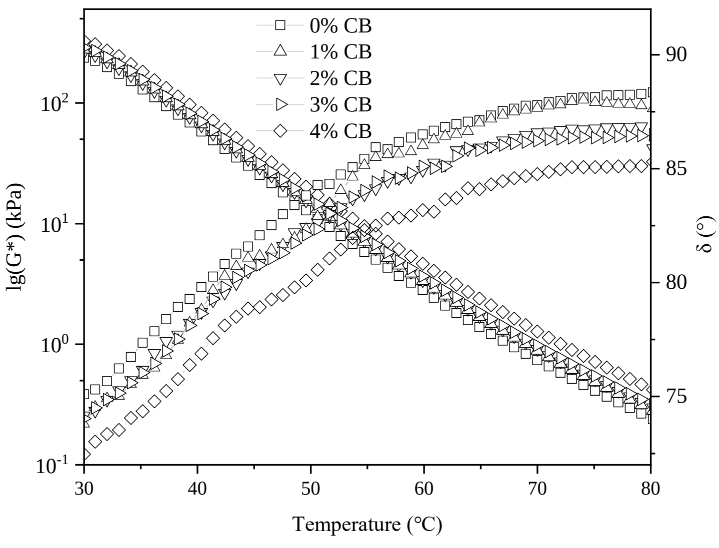

Figure 7. Relationship of complex modulus (G*) and phase angle (δ) values of CB-modified asphalt samples in the temperature region of 30 to 80 °C.

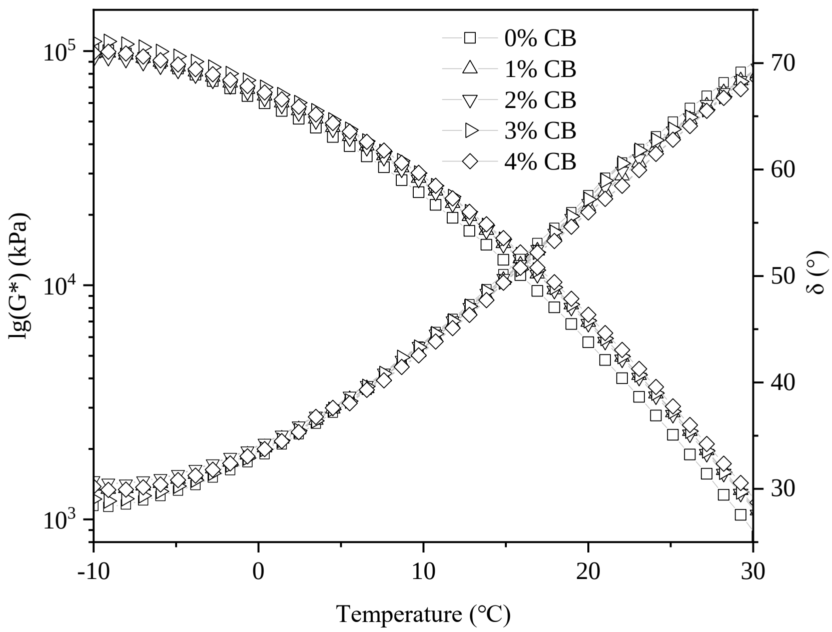

Figure 8. Relationship of complex modulus (G*) and phase angle (δ) values of CB-modified asphalt samples in the temperature region of−10–30 °C.

The rutting factor (G*/sinδ) was used to evaluate the effect of the CB modifier on the high-temperature performance of the asphalt binders, and the results are shown in Figure 9. The smaller the rutting factor, the greater the risk of permanent deformation. As seen in Figure 9, the rutting factor showed a decreasing trend across the whole temperature region (30–80 °C). This implies that the asphalt binder was more prone to permanent deformation at high temperatures. At the same temperature, the rutting factor increased with the CB modifier content, indicating that the CB modifier improved the high-temperature stability of the asphalt binder.

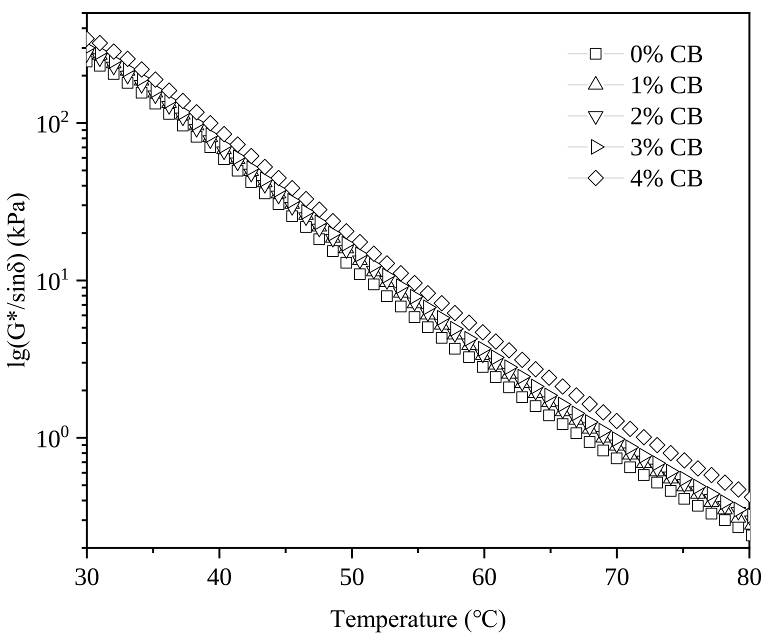

Figure 9. Rutting factor (G*/sinδ) values of CB-modified asphalt samples.

In addition, the cracking factor (G*cos2δ/sinδ) was adopted to assess the resistance of the asphalt binders to non-loading cracks at low temperatures, and the results are shown in Figure 10. The smaller the cracking factor, the better the resistance to cracking. The cracking factor tended to decrease across the whole temperature region (−10–30 °C), implying that the asphalt binder was less susceptible to cracking when the temperature increased. This is due to asphalt being a temperature-sensitive viscoelastic material. As the temperature increases, the asphaltenes in asphalt form a network structure at high temperatures, enhancing the overall stability of the asphalt and making it less prone to deformation. At the same time, the high temperature causes the asphalt to gradually soften and increase in fluidity, making it less likely to crack. This is reflected in Figure 10, where the higher the temperature, the lower the cracking factor, and the less likely the asphalt mixture was to crack. At the same temperature, as the content of the CB modifier increased, the cracking factor increased, and the asphalt mixture became more prone to cracking. Therefore, it can be inferred that the CB modifier made the asphalt binder more susceptible to cracking. Because asphalt mixtures are more likely to crack at low temperatures compared with high-temperature environments, further attention must be paid to improving the low-temperature performance of CB-modified asphalt binders.

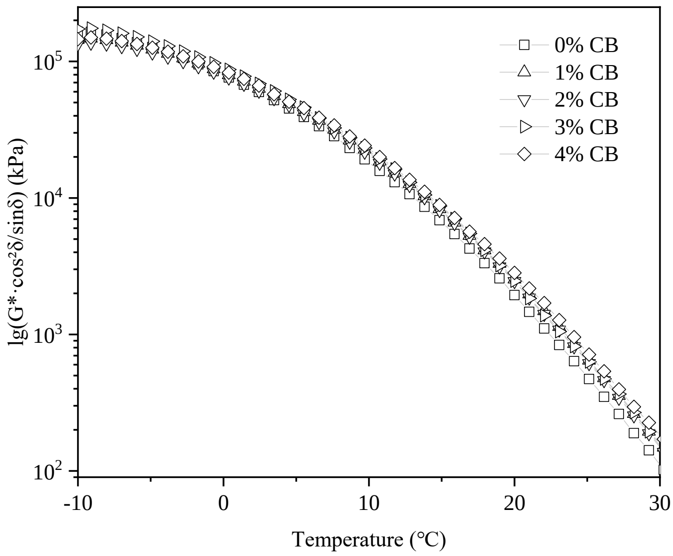

Figure 10. Cracking parameter ($G^*\cos^2\delta/\sin\delta$) values of CB-modified asphalt samples.

3.5. Frequency Sweep Test

According to the Williams–Landel–Ferry equation, the primary curves of the complex modulus (G*) for all the asphalt binders were developed, and the results are shown in Figure 11. The reference temperature was 20 °C. As seen in Figure 11, the relatively low-frequency region represents the high temperature, and the relatively high-frequency region represents the low temperature.

Across the whole frequency region, the complex modulus increased with the loading frequency. This implies that the complex modulus increased while the temperature decreased, which is in accordance with the results shown in Figures 7 and 8. At the same frequency, the complex modulus increased with the CB modifier content. In general, the low-frequency region represents the high temperature, and the high-frequency region represents the low temperature. The effect of the CB modifier on the complex modulus of the asphalt binder in the high-temperature region was more significant than that at a low temperature. When the frequency was 10^4 Hz (low-temperature region), the complex modulus of the asphalt binder with 4% CB modifier was 1.16 times that of the virgin asphalt binder. When the frequency was 10^{-4} Hz (high-temperature region), the corresponding results increased to 2.03 times.

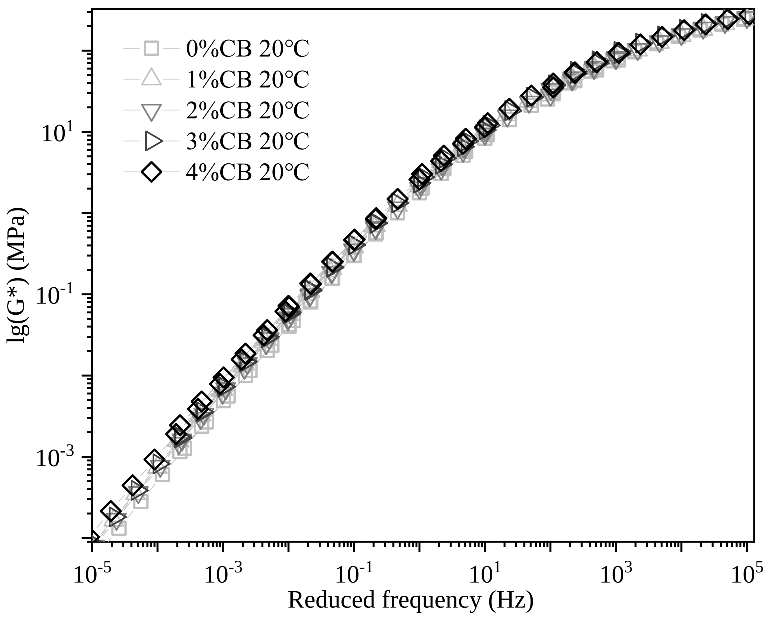

Figure 11. Complex moduli of different asphalt samples.

4. Conclusions

This study investigated the effect of CBs on the physical properties, storage stability, and rheological properties of asphalt binders. The main findings can be concluded as follows:

(1) The cellulose acetate fibers in CBs can add consistency and hardness to asphalt. With the increase in the CB content in the asphalt mixtures, the penetration value of the CB-modified asphalt adhesive decreased, the softening point increased, the ductility dropped significantly, and the viscosity increased, leading to poorer storage stability. CBs enhanced the shear resistance and high-temperature stability of the asphalt binder but reduced its low-temperature crack resistance and storage stability.

(2) CBs can enhance the high-temperature stability of asphalt binders but reduce their low-temperature stability. At the same temperature, as the CB content increased, the G* of the modified asphalt binder gradually increased, indicating a higher proportion of elastic components in the modified asphalt and an improved resistance to deformation. The G* of the modified asphalt also showed an upward trend with the increasing loading frequency, suggesting that the asphalt's resistance to deformation improved with the increase in the loading frequency.

(3) This study provides a reference for applying CB-modified asphalt. Recycling CBs contributes to environmental protection and promotes recycling efforts. However, when the content of CBs reaches 2%, the storage stability of the modified asphalt fails to meet the requirements of the Chinese standard JTG F40-2004 for SBS-modified asphalt. Further research is needed to improve the storage stability and low-temperature stability of CB-modified asphalt mixtures.

In this study, multiple tests were conducted on the penetration, ductility, and softening point of the modified asphalt. However, the data variability was particularly high. Therefore, we selected several sets of better data as representatives for analysis. Temperature scanning, frequency scanning, and rheological property tests were conducted with fewer replicates. In future research, we will further supplement and improve this part of the content.

Author Contributions: X.H.: data curation, validation, and writing—original draft. X.C. and J.Y.: conceptualization and writing—review and editing. J.Y. and L.Z.: project administration and funding acquisition. G.C.: data curation and investigation. Y.Y.: investigation and methodology. All authors have read and agreed to the published version of the manuscript.

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Institutional Review Board Statement: This study did not require ethical approval.

Informed Consent Statement: This study did not involve humans.

Data Availability Statement: The original contributions presented in this study are included in this article; further inquiries can be directed to the corresponding author.

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Conflicts of Interest: Xianglong Chen was employed by the China Harbour Engineering Company Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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