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Viscoelastic Properties, Rutting Resistance, and Fatigue Resistance of Waste Wood-Based Biochar-Modified Asphalt

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Abstract: The purpose of this study is to explore the viscoelastic properties, rutting resistance, and fatigue resistance of waste wood-based biochar-modified asphalt. The biochar with 2%, 4%, and 8% mixing amounts and two kinds of particle size, 75–150 µm and <75 µm, were used as modifiers of petroleum asphalt. Meanwhile, in the control group, a graphite modifier with a particle size of 0–75 µm and mixing amount of 4% was used for comparison. Aged asphalts were obtained in the laboratory by the Rolling Thin Film Oven (RTFO) test and the Pressure Aging Vessel (PAV) test. The viscoelastic properties, rutting resistance, and fatigue resistance of biochar-modified asphalt were evaluated by phase angle, critical high temperature, and fatigue cracking index by the Dynamic Shear Rheometer (DSR) test. In addition, the micromorphology of biochar and graphite was compared and observed by using the scanning electron microscope (SEM). The results show that increasing the mixing amount of biochar gave a higher elastic property and significantly better rutting resistance of the modified asphalt at high temperature. Compared with graphite, the biochar has a rougher surface and more pores, which provides its higher specific surface area. Therefore, it is easier to bond with asphalt to form a skeleton network structure, then forming a more stable biochar–asphalt base structure. In this way, compared to graphite-modified asphalt, biochar-modified asphalt showed better resistance to rutting at high temperature, especially for the asphalt modified with biochar of small particle size. The critical high temperature $T^{(G^*/\sin\delta)}$ of 4% Gd, 4% WD, and 4% Wd was 0.31 °C, 1.57 °C, and 2.92 °C higher than that of petroleum bitumen. In addition, the biochar asphalt modified with biochar of small particle size had significantly better fatigue cracking resistance than the asphalt modified with biochar of large particle size. The fatigue cracking indexes for 2% Wd, 4% Wd, and 8% Wd were 29.20%, 7.21%, and 37.19% lower by average than those for 2% WD, 4% WD, and 8% WD at 13–37 °C. Therefore, the waste wood biochar could be used as the modifier for petroleum asphalt. After the overall consideration, the biochar-modified asphalt with 2%–4% mixing amount and particle size less than 75 µm was recommended.

Keywords: asphalt pavement; biochar; biochar modified asphalt; viscoelastic properties; rutting resistance; fatigue resistance; performance evaluation

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1. Introduction

Biochar is a kind of organic compound product rich in carbon. It has many varieties, which is mainly produced by pyrolysis technology [1,2]. Most of the existing research on biochar has worked on the environmental science and soil science fields. The biochar plays a great role in agricultural soil improvement, environmental pollution remediation, soil carbon pool enhancement, and greenhouse gas emission reduction [3,4]. The biochar, acting as a soil modifier, can promote nutrient cycling to improve soil productivity and plant growth [5–8]. However, according to recent studies, in many cases, crop productivity cannot be improved or even decreased due to pesticides and water adsorption [9]. The reason is that biochar used in the soil can store toxic substances by absorbing or other physicochemical reactions, thus bringing a negative effect on the environment [10]. Furthermore, inactivated biochar is hard to isolate from soil [9,11]. In light of this, if it is piled without control, the biochar in air will serve as a threat to our human body [12].

At present, bio-based materials and carbon-based materials, such as carbon black, carbon fiber, and graphite, have been used for asphalt modification. Among them, carbon fiber modifier was proven to increase the rutting resistance of asphalt exposed to high temperature and alleviate its oxidation process [13,14], while it contributed to the increase in asphalt fatigue life [15] as well as improved the flexural strength and self-healing ability of asphalt concrete [16]. Adding lignin fibers to bitumen increased the rutting resistance of asphalt binder and the adhesion between asphalt binder and aggregates [17]. Rapeseed and oleic imidazolines could improve the low-temperature crack resistance and permanent deformations resistance of polymer-modified asphalt [18]. The asphalt concrete modified with graphite improved the thermal conductivity, rheological properties, and high-temperature properties of asphalt concrete [19]. The combination of graphite and carbon fiber to modify the asphalt concrete increased its electrical conductivity and mechanical properties [20]. In addition, carbon black modifiers can be applied to improve the properties of asphalt at high temperature, the anti-aging performance, and the thermal conductivity [21]. In consideration of these good performances from the carbon-based material for pavement, the biochar may be used in road engineering. However, little studies were made to work out the application of biochar as the modifier for road materials. It was found that 1 wt% biochar from food waste could improve the impermeability of mortar [22]; paper mill sludge and paper pulp-based biochar improved the mechanical strength of concrete [23]; and pig manure-based biochar reduced the temperature sensitivity and shear sensitivity of asphalt [24]. In addition, according to studies, 10 wt% of mixing amount of switchgrass-based biochar significantly reduced the temperature sensitivity of asphalt and improved the moisture susceptibility, crack resistance, and rutting resistance of asphalt [25,26]. Furthermore, biochars derived from straw, rice husk, palm fruit ash, and soybean powder decreased the temperature sensitivity of asphalt while improving the high-temperature stability reflected by dynamic shear modulus [27,28]. In addition, biochar-modified asphalt pavement was proven to remove 97.2% of nitrate and 56% of chromium from rainwater, thus improving the groundwater quality [29].

In summary, it is possible to use the biochar in the asphalt. However, in light of its varieties, biochars may have performance differences due to different material sources. Few studies on the evaluation of the properties of waste wood-based biochar-modified asphalt exist currently. In addition, the application of biochar to pavement materials can both reduce the environmental pollution caused by biochar and reuse waste biomass resources. Therefore, this study worked on the waste wood-based biochar as a modifier for petroleum asphalt to evaluate the viscoelastic properties, rutting resistance, and fatigue resistance of biochar-modified asphalt with different biochar mixing amounts and particle sizes. In parallel, the waste wood-based biochar-modified asphalt was compared with petroleum asphalt and graphite-modified asphalt to probe the possibility of using biochar as an asphalt modifier, and to recommend biochar-modified asphalt with better performance.

2. Test Material

2.1. Matrix Asphalt

Petroleum asphalt with performance grade PG 58-28 was used as the base asphalt, and its performance meets the requirements of specification [30]. The detailed technical indexes are shown in Table 1.

Table 1. Basic properties of petroleum asphalt PG 58-28.

Evaluation Index	Result	Specification Requirements	Specification
Specific gravity	1.03	-	-
Rotational viscosity (135 °C, Pa·s)	0.350	<3.0	AASHTO T316-13
Rutting resistance factor of the original asphalt $G^*/\sin\delta$ (58 °C) (kPa)	1.995	>1.0	AASHTO T315-12
Rutting resistance factor of aging asphalt after RTFOT $G^*/\sin\delta$ (58 °C) (kPa)	5.018	>2.2	AASHTO T315-12
m-value (−18 °C)	0.31	>3.0	AASHTO T313-12

2.2. Graphite and Graphite-Modified Asphalt

For comparison, the graphite modifier was used, with the particle size less than 75 μm, density 2.25 g/cm^3, and minimum particle thickness of 0.11 mm [19]. The graphite-modified asphalt was prepared by adding 4% mixing amount of graphite into a base binder of PG 58-28 and stirring them at 120 °C for about one hour by a mixer with a mixing speed of 3000–5000 revolutions per minute.

2.3. Biochar and Biochar-Modified Asphalt

Biochar with a density of 0.4 g/cm^3 was prepared from waste wood by a special pyrolysis process [31]. To study the effect of biochar with different mixing amounts and particle sizes, three mixing amounts were used: 2%, 4%, and 8%, as well as particle sizes of 75–150 μm and <75 μm. The biochar-modified asphalt was prepared by adding biochar with different particle sizes and mixing amounts into the base binder of PG 58-28 and stirring them at 120 °C for about one hour by a mixer with a mixing speed of 3000–5000 revolutions per minute.

This study worked on 1 graphite-modified asphalt and 1 petroleum asphalt as the control, and 6 biochar-modified asphalt, altogether 8 kinds of asphalt, as shown in Table 2. Here, "W" represents waste wood-based biochar, "G" represents graphite, "D" represents larger particle sizes of 75–150 μm, and "d" represents smaller particle sizes of <75 μm.

Table 2. Test samples.

Asphalt Type	Instruction
PG 58-28	Petroleum asphalt
2% WD	Biochar-modified asphalt, with 2% of biochar mixing amount, in 75–150 μm particle size
4% WD	Biochar-modified asphalt, with 4% of biochar mixing amount, in 75–150 μm particle size
8% WD	Biochar-modified asphalt, with 8% of biochar mixing amount, in 75–150 μm particle size
2% Wd	Biochar-modified asphalt, with 2% of biochar mixing amount, in 0–75 μm particle size
4% Wd	Biochar-modified asphalt, with 4% of biochar mixing amount, in 0–75 μm particle size
8% Wd	Biochar-modified asphalt, with 8% of biochar mixing amount, in 0–75 μm particle size
4% Gd	Graphite-modified asphalt, with 4% of biochar mixing amount, in 0–75 μm particle size

3. Test Method

3.1. Rolling Thin Film Oven (RTFO) Test

The RTFO test was used to simulate the asphalt aging during storage, transportation, mixing, and paving. A short-time aging test was conducted on the asphalt according to the specification AASHTO T240 [32], where the test temperature was 163 ± 0.5 °C, the aging time was controlled for 85 min, the weight of the asphalt sample in the sample bottle was 35 g ± 0.5 g, and the flow rate of air injection in the oven was 4000 L/min ± 200 mL/min.

3.2. Pressure Aging Vessel (PAV) Test

The PAV test was designed to accelerate the asphalt aging with high temperature and pressed air, to simulate the oxidative aging of asphalt during 5–10 years of road use. The samples used were residues from the RTFO test of asphalt. The PAV test was performed based on the specification AASHTO R28 [33], where the air pressure was 2.1 ± 0.1 Mpa, the aging time was controlled for 20 h, the mass of the asphalt sample in the aging plate was 50 g ± 5 g, and the asphalt film was about 3.2 mm thick.

3.3. Dynamic Shear Rheometer (DSR) Test

The DSR test was performed on the asphalt according to the specification AASHTO R28 [34]. The phase angle (δ) and rutting index (G*/sinδ) of the original and RTFO-aged sample were measured, and the critical high temperature of asphalt was calculated based on specification AASHTO M32-10 [30]. These were used to characterize the viscoelastic properties and rutting resistance of asphalt exposed to high temperatures of 52–76 °C with a temperature interval of 6 °C. The fatigue cracking index (G*sinδ) of the PAV aged sample was measured to evaluate the fatigue resistance of asphalt exposed to medium temperatures of 13–37 °C with a temperature interval of 6 °C. Three parallel samples were tested each time.

The critical high temperature refers to the minimum temperature of $T_o^{(G^*/\sin\delta)}$ and $T_R^{(G^*/\sin\delta)}$. $T_o^{(G^*/\sin\delta)}$ is the critical high temperature of original asphalt, the rutting index at that temperature of original asphalt is 1.0 kPa at 1.59 Hz (10 rad/s); $T_R^{(G^*/\sin\delta)}$ is the critical high temperature of RTFO aged asphalt, the rutting index at that temperature of RTFO aged asphalt is 2.2 kPa at 1.59 Hz (10 rad/s), as shown in Formula (1).

$$T^{(G^*/\sin\delta)} = \min.\ (T_o^{(G^*/\sin\delta)}, T_R^{(G^*/\sin\delta)}) \tag{1}$$

where $T^{(G^*/\sin\delta)}$ = Critical high temperature of asphalt; $T_o^{(G^*/\sin\delta)}$ = Critical high temperature of original asphalt; $T_R^{(G^*/\sin\delta)}$ = Critical high temperature of RTFO-aged asphalt.

3.4. Electron Microscope (SEM) Test

The microtopography of biochar and graphite was observed by using Hitachi (Schaumburg, IL, USA) S-4700 and JEOL (Peabody, KS, USA)-6400, where Hitachi S-4700 had an acceleration voltage of 1–5 kV, an emission current of 9 A, and the working distance of 2.8–8 mm; JEOL-6400 had an acceleration voltage of 20 kV and the working distance of 39 mm. The prepared biochar or graphite particles were moved to a double-sided adhesive carbon tape, which was then coated on an aluminum sample holder.

4. Results and Analyses

4.1. Analysis of Viscoelastic Properties

The viscoelastic properties of asphalt can be evaluated by its phase angle. The larger phase angle presents a higher viscous component of asphalt. The phase angle of the original asphalt and RTFO aged asphalt is shown in Figure 1.

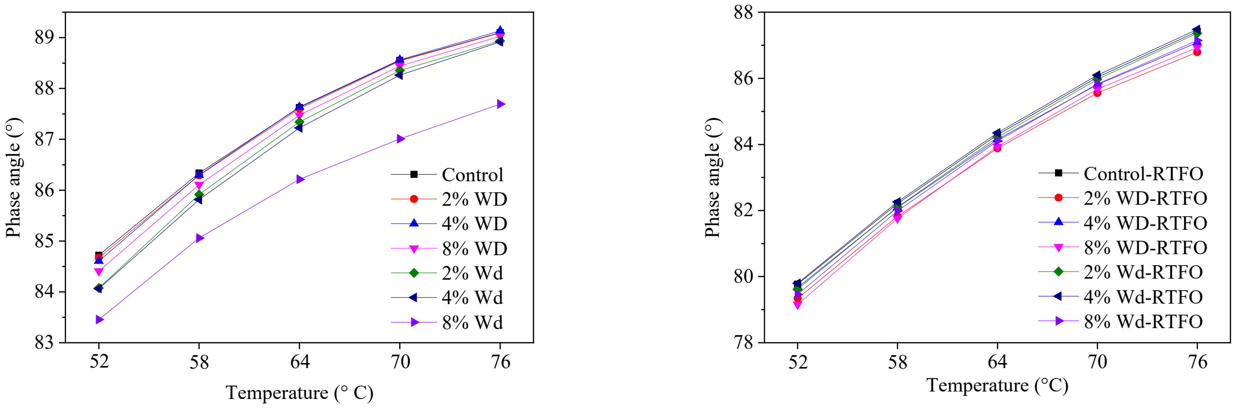

Figure 1. Phase angle (δ) of biochar-modified asphalt. (**a**) Phase angle (δ) of original sample; (**b**) Phase angle (δ) of RTFO aged sample.

From Figure 1a, the phase angle of asphalt increases when the temperature rises, indicating that the asphalt is more likely to exhibit viscous properties at higher temperature. However, the adding of biochar lowered the phase angle of the asphalt. It can be found that the phase angle of biochar-modified asphalt decreased with the addition of biochar. Compared with asphalt, biochar exhibits harder and more rigid characteristics. When harder biochar was added into the asphalt, the asphalt volume was filled, thus improving the elastic composition. The particle size of biochar had a significant effect on the phase angle of the asphalts. As shown in Figure 1a, at 52–76 °C, the phase angles of 2% Wd, 4% Wd, and 8% Wd are 0.31°, 0.39°, and 1.21° smaller than those of 2% WD, 4% WD, and 8% WD, respectively. This demonstrated that the biochar-modified asphalt with a small particle size had the smaller phase angle than that of biochar-modified asphalt with a large particle size. The reason could be that the biochar of small particle size has a high surface area accompanied by a porous structure, resulting in stronger adhesion between them and the matrix asphalt. In this way, a strong structure is formed [31].

According to Figure 1b, the phase angles of all asphalt are relatively similar, which means that compared with petroleum asphalt and biochar-modified asphalt of large particle size, the phase angle of biochar-modified asphalt of small particle size had the least change after the RTFO aging. Therefore, the viscoelastic properties of asphalt modified by biochar of small particles were less affected by high temperature, which had the best aging resistance.

To compare the viscoelastic properties of asphalt affected by different modifiers, the phase angles of PG 58-28, 4% Gd, 4% WD, and 4% Wd before and after RTFO aging test are shown in Figure 2. Compared with 4% WD, 4% Gd had a smaller phase angle and more elastic components, which may be caused by the large volume filling of small hard particles. After short-term aging, the phase angle change of 4% Wd was slightly smaller than that of 4% Gd by 0.072 °C on average. This may be caused by the surface difference that graphite has a relatively smooth surface, while biochar has a complex multi-hole structure. Adding the latter may enhance the formation of the biochar–matrix asphalt structure. Therefore, the elastic components of asphalt modified with biochar of small particle size were higher, and the viscoelastic properties were least affected by high temperature.

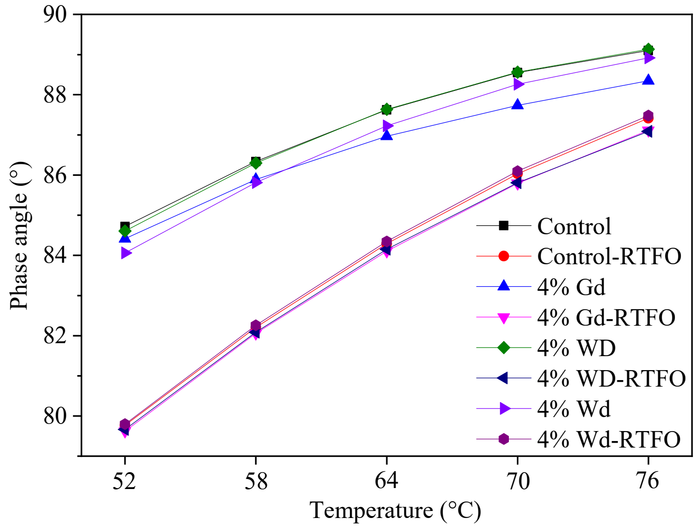

Figure 2. Phase angle of different types of asphalt (δ).

4.2. Analysis of Rutting Resistance

After the DSR test, the temperature ($T_o^{(G^*/\sin\delta)}$) at which the rutting index ($G^*/\sin\delta$) of original asphalt was 1.0 kPa at 1.59 Hz (10 rad/s) was obtained as well as the temperature ($T_R^{(G^*/\sin\delta)}$) at which the rutting index ($G^*/\sin\delta$) of RTFO aged asphalt was 2.2 kPa at 1.59 Hz (10 rad/s). The critical high temperature of petroleum asphalt, biochar-modified asphalt, and graphite modified asphalt is shown in Figures 3 and 4.

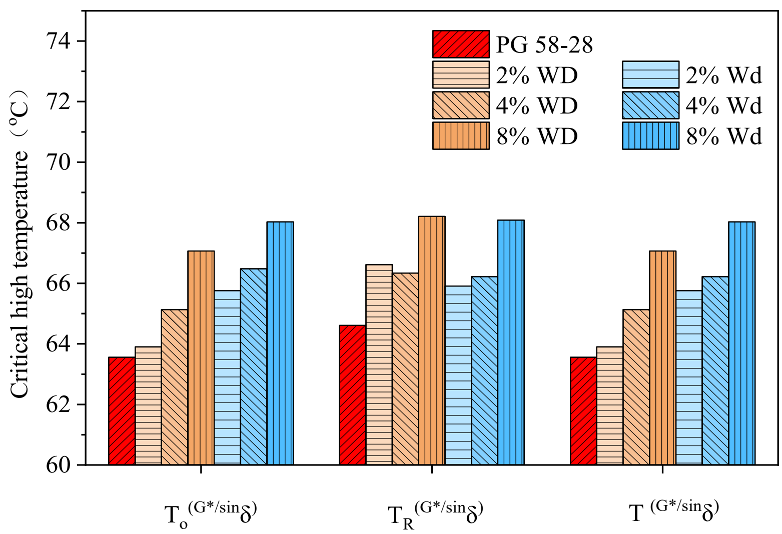

Figure 3. Critical high temperature of biochar-modified asphalt.

The smaller one of $T_o^{(G^*/\sin\delta)}$ and $T_R^{(G^*/\sin\delta)}$ was considered as the critical high temperature of the asphalt sample, which can more intuitively characterize its rutting resistance under high temperature. As shown in Figure 3, the greater the amount of biochar, the higher the critical high temperature of asphalt, indicating that biochar-modified asphalt had better rutting resistance at high temperature. In this regard, the volume filling of harder biochar particles is one of the main reasons, which increases the elastic components of asphalt to a certain extent. This is in line with the above analysis of viscoelastic properties. For another possible reason, the porous structure of biochar led to its higher surface area, thus causing higher adhesion between biochar and petroleum asphalt [31]. In addition, the resistance to rutting of biochar-modified asphalt exposed to high temperature increased with the increase in biochar mixing amount. This is because increasing the mixing amount of biochar makes it more easily for hard porous particles to come into contact with each other. In this way, a skeleton frame is formed in asphalt to enhance its rutting resistance. After analyzing the influences of biochar of different particle sizes on the critical high temperature of modified asphalt, it was seen that the asphalt modified with biochar of small particle size had better rutting resistance under high-temperature conditions than the asphalt modified with biochar of large particle size. This can be explained by the large volume filling of harder biochar particles and the higher specific surface area. With the same mass, biochar with a smaller particle size has a larger volume and higher specific surface area than biochar with a larger particle size.

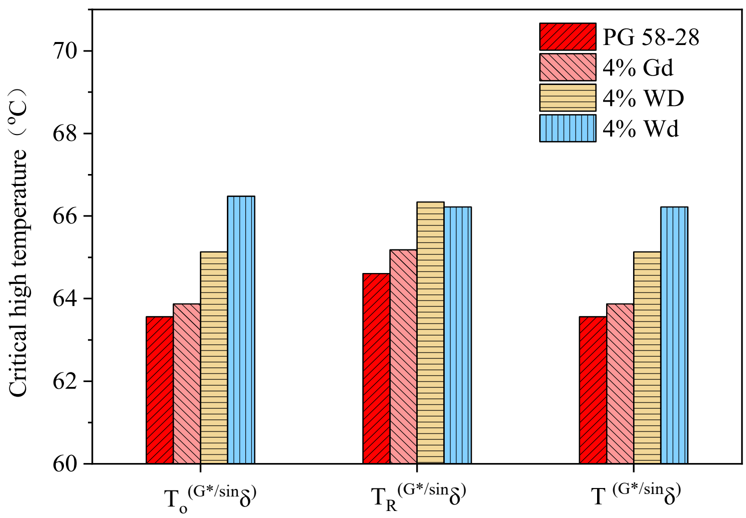

Figure 4. Critical high temperature of different types of asphalt.

After comparing the critical high temperature of different types of asphalt, it was found that biochar-modified asphalt had better rutting resistance than petroleum asphalt and graphite-modified asphalt. For example, the critical high temperature $T^{(G^*/\sin\delta)}$ of 4% Gd, 4% WD, and 4% Wd was 0.31 °C, 1.57 °C, and 2.92 °C higher than that of petroleum bitumen. The addition of carbon-based particles increased the elastic components of asphalt and enhanced the properties at high temperature of asphalt. Furthermore, the asphalt modified with biochar of small particle size had the best high-temperature performance. As the density of biochar is lower than that of graphite, the particle volume of biochar is higher than that of graphite at the same mixing amount. In addition, under the same mass, smaller-sized biochar particles have a higher surface area and porous structure, which promotes the asphalt to generate a greater adhesion to biochar and to form a biochar–asphalt base structure, thus reducing the influence of high temperature on asphalt. These results are consistent with those of the elastic components test.

4.3. Analysis of Fatigue Resistance

In this study, the fatigue cracking index (G*sinδ) was utilized to characterize the fatigue resistance of biochar-modified asphalt, as shown in Figure 5.

The higher fatigue cracking index (G*sinδ) represents the worse anti-fatigue cracking ability of asphalt. According to Figure 5, as the temperature rises, the asphalt fatigue cracking index (G*sinδ) decreased, indicating that the fatigue resistance of asphalt was better at higher temperature. The added biochar reduced the fatigue resistance of asphalt. The G*sinδ of biochar-modified asphalt was basically higher compared to petroleum asphalt PG 58-28, and it increased with the higher mixing amount of biochar. Moreover, the size of biochar particles had a significant influence on the fatigue resistance of biochar-modified asphalt. Compared with the asphalt modified with biochar of large particle size, the asphalt modified with biochar of small particle size had a lower fatigue cracking index. For example, the fatigue cracking indexes for 2% Wd, 4% Wd, and 8% Wd were 29.20%, 7.21%, and 37.19% lower by average than those for 2% WD, 4% WD, and 8% WD at 13–37 °C. It demonstrated that the fatigue cracking resistance of asphalt modified with biochar of small particle size was better than that of asphalt modified with biochar of large particle size. Furthermore, little difference exists between the fatigue resistance of 2% Wd and that of petroleum asphalt, especially when the temperature was 19–37 °C; the fatigue resistance of 2% Wd was similar to or even better in comparison with asphalt PG 58-28.

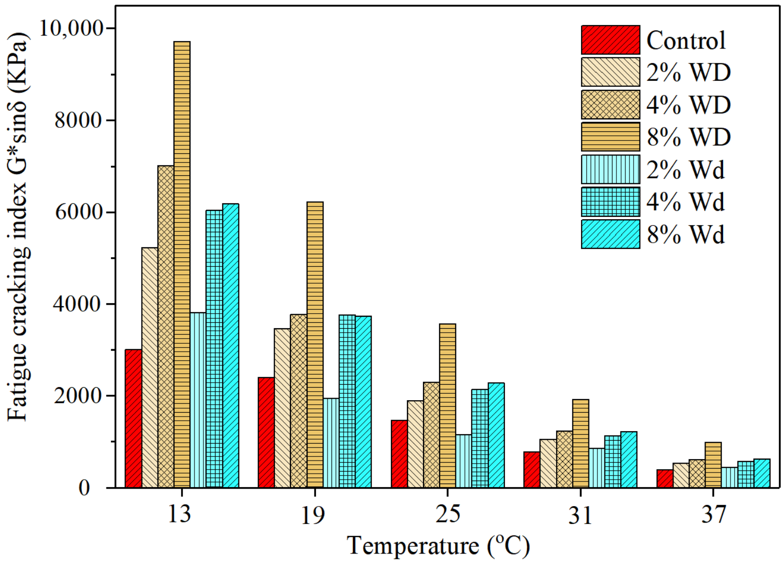

Figure 5. Fatigue cracking index of biochar-modified asphalt (G*sinδ).

To compare the effects of different modifiers on the fatigue resistance of asphalt, the fatigue cracking index of PG 58-28, 4% Gd, 4% WD, and 4% Wd is shown in Figure 6. The G*sinδ of 4% Gd was higher than that of petroleum asphalt, but lower than 4% WD and 4% Wd. It indicated that graphite-modified asphalt owned better fatigue cracking resistance compared to biochar-modified asphalt. However, there was no significant difference in the fatigue cracking index (G*sinδ) of 4% Gd and 4% Wd; the G*sinδ of 4% Wd was only 408.00 kPa higher than that of 4% Gd. This demonstrated that smaller particle-sized biochar-modified asphalt had a similar fatigue cracking resistance to graphite-modified asphalt.

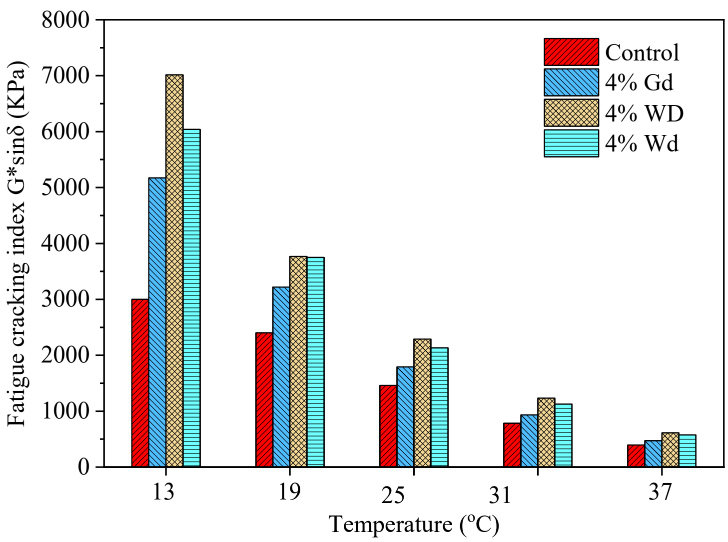

Figure 6. Fatigue cracking index of different types of asphalt (G*sinδ).

4.4. Analysis of Microstructure

The SEM images for biochar and graphite are shown in Figure 7.

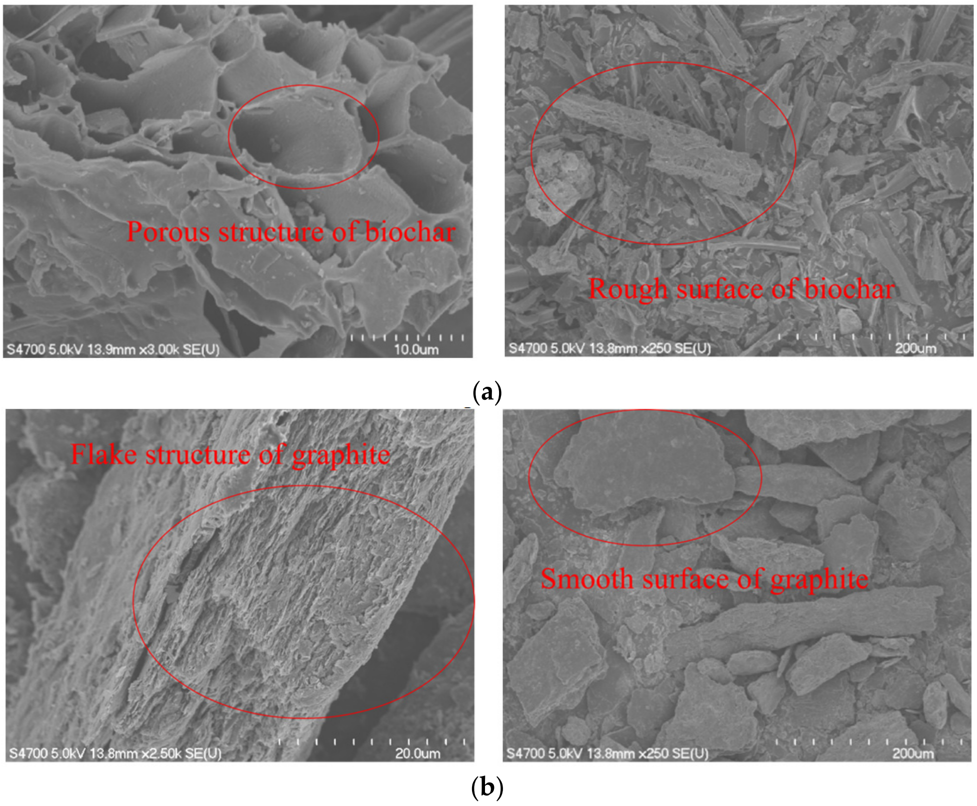

Figure 7. SEM images of biochar and graphite. (**a**) Images of biohar; (**b**) Images of graphite.

According to Figure 7, the biochar particles are mostly tubular or flake, with a significant porous structure and rough surface, while the flake graphite surface is relatively smooth. On one hand, this porous, fibrous, and hollow structure on the surface of biochar particles enables it to have a larger specific surface area, which makes it fully bond with asphalt. On the other hand, the irregular shape of biochar particles makes it easier to lap with asphalt to form a skeleton network structure, forming a more stable biochar–asphalt base structure. Therefore, it promotes the modified asphalt to have better road performance. To some extent, it can explain the reason why biochar-modified asphalt has better high-temperature performance than graphite-modified asphalt. As the biochar utilized for this study was obtained from waste wood by the thermal pyrolysis process, its microstructure still retains part of the structural characteristics of biomass raw materials. The microstructures of biochar obtained from different kinds of biomass raw materials may be different, including the macroscopic performance. Therefore, before the application of biochar, biomass raw materials should be selected according to the application requirements so as to achieve the best application effect.

4.5. Statistical Analysis

In order to study whether the change in properties of asphalt after the use of biochar was significant, a statistical analysis was performed based on SPSS software. The tests of the between-subjects effects on phase angle, critical temperature, and fatigue cracking index of modified asphalts are shown in Tables 3–5, respectively. A p-value of less than 0.05 is considered to indicate that the factor has a significant influence on the property of asphalt.

As can be seen from Table 3, all p-values were higher than 0.05. This indicated that the influence of type, mixing amount, and particle size of biochar on the phase angle of asphalt was not significant. However, for the influence factor of mixing amount of biochar, the p-value of 0.116 is the smallest. The mixing amount of biochar had a significantly higher influence on the phase angle of asphalt than the type and particle size of biochar. From Table 4, it can be found that the type, mixing amount, and particle size of biochar all had a significant influence on high-temperature performance. This is consistent with the above results of rutting resistance. According to Table 5, the type of modifier, with a p-value of 0.860, has no significant influence on the fatigue resistance of asphalt. However, both the mixing amount and particle size of biochar exhibited a significant influence on the fatigue resistance of asphalt. Overall, the resistance to rutting and fatigue were significantly affected by the mixing amount and particle size of biochar.

Table 3. Tests of between-subjects effects on phase angle.

Source	df	Mean Square	Statistic Value (F)	Significance Probability (p)
Corrected model	10	57.482	129.509	0.000
Intercept	1	247,350.122	557,285.216	0.000
Type of modifier	1	0.172	0.388	0.535
Mixing amount of biochar	2	0.986	2.222	0.116
Particle size of biochar	1	0.491	1.106	0.297

Table 4. Tests of between-subjects effects on critical high temperature.

Source	df	Mean Square	Statistic Value (F)	Significance Probability (p)
Corrected model	7	8.709	43.976	0.000
Intercept	1	52,912.657	267,169.545	0.000
Type of modifiers	1	8.789	44.379	0.000
Mixing amount of biochar	2	10.056	50.775	0.000
Particle size of biochar	1	3.721	18.788	0.000

Table 5. Tests of between-subjects effects on fatigue cracking index.

Source	df	Mean Square	Statistic Value (F)	Significance Probability (p)
Corrected model	9	18,420,737.266	28.445	0.000
Intercept	1	100,839,554.363	155.714	0.000
Type of modifiers	1	20,424.173	0.032	0.860
Mixing amount of biochar	2	6,459,913.709	9.975	0.000
Particle size of biochar	1	6,210,405.133	9.590	0.004

5. Conclusions

In this study, biochar was utilized as a modifier of petroleum asphalt. Three mixing amounts were used: 2%, 4%, and 8%, as well as two particle sizes of 75–150 μm and <75 μm. In addition, flake graphite with a particle size <75 μm and mixing amount of 4% was used as the control modifier. The viscoelastic properties, rutting resistance, and fatigue resistance of biochar-modified asphalt were evaluated by using RTFO, PAV, and DSR tests, and the microstructures of biochar and graphite were observed by using SEM. Statistical analysis was performed. The following conclusions were obtained:

1. Asphalt showed more obvious viscous characteristics with increasing temperature. The addition of biochar particles promoted the improvement of the elastic components of asphalt, and it increased with the increasing amount of biochar. Compared to asphalt PG 58-28, graphite-modified asphalt, and biochar-modified asphalt of large particle size, the elastic components of biochar-modified asphalt of small particle size was higher, whose viscoelastic properties were least affected by high temperature.

2. The addition of biochar increased the critical high temperature of asphalt, and the rutting resistance of biochar-modified asphalt significantly increased with the increase in biochar mixing amount. The biochar-modified asphalt had better rutting resistance at high temperature than petroleum asphalt and graphite-modified asphalt, especially for the asphalt modified with biochar of small particle size.
3. The fatigue cracking resistance of asphalt reduced with the addition of biochar. However, the fatigue resistance of 2% Wd was similar to or even better than that of petroleum asphalt, and 4% Wd had a similar fatigue cracking resistance to 4% Gd. The particle size of biochar had a significant influence on the fatigue resistance of biochar-modified asphalt. The binder modified with biochar of small particle size had better fatigue cracking resistance than the asphalt modified with biochar of large particle size.
4. The biochar had a rougher surface and more pores than graphite, which provided its larger specific surface area. This made it easier to bond with asphalt to form a skeleton network structure, thus forming a more stable biochar–asphalt base structure. Therefore, biochar-modified asphalt showed better rutting resistance at high temperature.

Overall, the addition of a biochar modifier can increase the elastic property and rutting resistance at high temperature while maintaining good resistance to fatigue. Therefore, it may be widely applied in the future. After full consideration, the biochar-modified asphalt with 2%–4% mixing amount and particle size less than 75 μm was recommended.

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References

1. Jaiswal, A.K.; Elad, Y.; Graber, E.R.; Frenkel, O. Rhizoctonia solani suppression and plant growth promotion in cucumber as affected by biochar pyrolysis temperature, feedstock and concentration. *Soil Biol. Biochem.* **2014**, *69*, 110–118. [CrossRef]
2. Muñoz, E.; Curaqueo, G.; Cea, M.; Vera, L.; Navia, R. Environmental hotspots in the life cycle of a biochar-soil system. *J. Clean. Prod.* **2017**, *158*, 1–7. [CrossRef]
3. Qi, L.; Pokharel, P.; Chang, S.X.; Zhou, P.; Niu, H.; He, X.; Wang, Z.; Gao, M. Biochar application increased methane emission, soil carbon storage and net ecosystem carbon budget in a 2-year vegetable–rice rotation. *Agric. Ecosyst. Environ.* **2020**, *292*, 106831. [CrossRef]
4. Xu, L.; Fang, H.; Deng, X.; Ying, J.; Lv, W.; Shi, Y.; Zhou, G.; Zhou, Y. Biochar application increased ecosystem carbon sequestration capacity in a Moso bamboo forest. *For. Ecol. Manag.* **2020**, *475*, 118447. [CrossRef]
5. Mukherjee, A.; Zimmerman, A.R. Organic carbon and nutrient release from a range of laboratory-produced biochars and biochar–soil mixtures. *Geoderma* **2013**, *193–194*, 122–130. [CrossRef]
6. Masiello, C.; Dugan, B.; Brewer, C.; Spokas, K.; Novak, J.; Liu, Z.; Sorrenti, G. *Biochar Effects on Soil Hydrology, Biochar for Environmental Management*, 2nd ed.; Routledge: London, UK, 2015.
7. Suliman, W.; Harsh, J.B.; Abu-Lail, N.I.; Fortuna, A.-M.; Dallmeyer, I.; Garcia-Pérez, M. The role of biochar porosity and surface functionality in augmenting hydrologic properties of a sandy soil. *Sci. Total Environ.* **2017**, *574*, 139–147. [CrossRef]

8. Ali, S.; Rizwan, M.; Qayyum, M.F.; Ok, Y.S.; Ibrahim, M.; Riaz, M.; Arif, M.S.; Hafeez, F.; Al-Wabel, M.I.; Shahzad, A.N. Biochar soil amendment on alleviation of drought and salt stress in plants: A critical review. *Environ. Sci. Pollut. Res.* **2017**, *24*, 12700–12712. [CrossRef]
9. Six, J. Biochar: Is There a Dark Side? 2014. Available online: https://ethz.ch/en/news-and-events/eth-news/news/2014/04/biochar-is-there-a-dark-side.html (accessed on 1 April 2014).
10. Downie, A. Biochar Production and Use: Environmental Risks and Rewards. Ph.D. Thesis, University of South Wales, Newport, UK, 2011.
11. An, C.; Huang, G. Environmental concern on biochar: Capture, then what? *Environ. Earth Sci.* **2015**, *74*, 7861–7863. [CrossRef]
12. Brick, S.; Lyutse, S. *Biochar: Assessing the Promise and Risks to Guide US Policy*; Natural Resources Defense Council: NRDC Issue Paper; NRDC: New York, NY, USA, 2010.
13. Cleven, M.A. Investigation of the Properties of Carbon Fiber Modified Asphalt Mixtures. Master's Thesis, Michigan Technological University, Houghton, MI, USA, 2000.
14. Yao, H.; You, Z.; Li, L.; Goh, S.W.; Lee, C.H.; Yap, Y.K.; Shi, X. Rheological properties and chemical analysis of nanoclay and carbon microfiber modified asphalt with Fourier transform infrared spectroscopy. *Constr. Build. Mater.* **2013**, *38*, 327–337. [CrossRef]
15. Khattak, M.J.; Khattab, A.; Rizvi, H.R.; Zhang, P. The impact of carbon nano-fiber modification on asphalt binder rheology. *Constr. Build. Mater.* **2012**, *30*, 257–264. [CrossRef]
16. Yoo, D.-Y.; Kim, S.; Kim, M.-J.; Kim, D.; Shin, H.-O. Self-healing capability of asphalt concrete with carbon-based materials. *J. Mater. Res. Technol.* **2018**, *8*, 827–839. [CrossRef]
17. Norgbey, E.; Huang, J.; Hirsch, V.; Liu, W.J.; Wang, M.; Ripke, O.; Li, Y.; Annan, G.E.; Ewusi-Mensah, D.; Wang, X.; et al. Unravelling the efficient use of waste lignin as a bitumen modifier for sustainable roads. *Constr. Build. Mater.* **2020**, *230*, 116957. [CrossRef]
18. Wróbel, M.; Woszuk, A.; Ratajczak, M.; Franus, W. Properties of reclaimed asphalt pavement mixture with organic rejuvenator. *Constr. Build. Mater.* **2021**, *271*, 121514. [CrossRef]
19. Wang, Z.; Dai, Q.; Guo, S. Laboratory performance evaluation of both flake graphite and exfoliated graphite nanoplatelet modified asphalt composites. *Constr. Build. Mater.* **2017**, *149*, 515–524. [CrossRef]
20. Liu, X.; Wu, S. Study on the graphite and carbon fiber modified asphalt concrete. *Constr. Build. Mater.* **2011**, *25*, 1807–1811. [CrossRef]
21. Cong, P.; Xu, P.; Chen, S. Effects of carbon black on the anti aging, rheological and conductive properties of SWd/asphalt/carbon black composites. *Constr. Build. Mater.* **2014**, *52*, 306–313. [CrossRef]
22. Gupta, S.; Kua, H.W.; Koh, H.J. Application of biochar from food and wood waste as green admixture for cement mortar. *Sci. Total. Environ.* **2018**, *619–620*, 419–435. [CrossRef]
23. Akhtar, A.; Sarmah, A.K. Novel biochar-concrete composites: Manufacturing, characterization and evaluation of the mechanical properties. *Sci. Total Environ.* **2018**, *616*, 408–416. [CrossRef]
24. Walters, R.C.; Fini, E.H.; Abu-Lebdeh, T. Enhancing Asphalt Rheological Behavior and Aging Susceptibility Using Bio-Char and Nano-Clay. *Am. J. Eng. Appl. Sci.* **2014**, *7*, 66–76. [CrossRef]
25. Zhao, S.; Huang, B.; Ye, X.P.; Shu, X.; Jia, X. Utilizing bio-char as a bio-modifier for asphalt cement: A sustainable application of bio-fuel by-product. *Fuel* **2014**, *133*, 52–62. [CrossRef]
26. Zhao, S.; Huang, B.; Shu, X.; Ye, P. Laboratory investigation of biochar-modified asphalt mixture. *Transp. Res. Rec. J. Transp. Res. Board* **2014**, *2445*, 56–63. [CrossRef]
27. Dong, W.; Ma, F.; Li, C.; Fu, Z.; Huang, Y.; Liu, J. Evaluation of Anti-Aging Performance of Biochar Modified Asphalt Binder. *Coatings* **2020**, *10*, 1037. [CrossRef]
28. Tarar, M.A.; Khan, A.H.; ur Rehman, Z.; Qamar, S.; Akhtar, M.N. Performance characteristics of asphalt binders modified with sunflower flour: A sustainable application of renewable resource derived material. *Constr. Build. Mater.* **2020**, *242*, 118157. [CrossRef]
29. Thomas, K.M.; Mathew, N.V.; Rajalekshmi, P.R.; Kumar, R.S.; Koshy, R.Z. Water Quality and Performance Assessment of Porous Asphalt Mix Modified Using Charcoal Powder. *J. Sustain. Eng. Proc. Ser.* **2019**, *1*, 123–136. [CrossRef]
30. AASHTO. *Standard Specification for Performance-Graded Asphalt Binder (M320-10)*; American Association of State Highway and Transportation Officials: Washington, DC, USA, 2013.
31. Zhang, R.; Dai, Q.; You, Z.; Wang, H.; Peng, C. Rheological Performance of Bio-Char Modified Asphalt with Different Particle Sizes. *Appl. Sci.* **2018**, *8*, 1665. [CrossRef]
32. AASHTO. *Effect of Heat and Air on a Moving Film of Asphalt Binder (Rolling Thin-Film Oven Test)*; American Association of State Highway and Transportation Officials: Washington, DC, USA, 2013.
33. AASHTO. *Standard Practice for Accelerated Aging of Asphalt Binder Using a Pressurized Aging Vessel (PAV)*; American Association of State Highway and Transportation Officials: Washington, DC, USA, 2013.
34. AASHTO. *Standard Method of Test for Determining the Rheological Properties of Asphalt Binder Using a Dynamic Shear Rheometer (DSR) (T 315-12)*; American Association of State Highway and Transportation Officials: Washington, DC, USA, 2013.