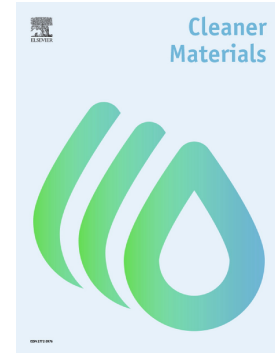


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Balanced Asphalt Binder Performance with Hazelnut Shell Biochar

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Abstract

Decarbonising road materials requires scalable strategies that enhance durability while valorising waste streams. This study investigates the effect of antioxidant biochar derived from European hazelnut shells (BH) on the operational and rheological performance of a PG 64–22 asphalt binder. BH was produced via slow pyrolysis at 300 °C and incorporated at contents of 2.5, 5.0 and 7.5 wt.% using two particle size fractions ($S < 20 \mu\text{m}$ and $L < 75 \mu\text{m}$). Owing to its carbon-rich composition and the presence of antioxidant compounds reported in previous studies, BH was evaluated as a sustainable modifier with the potential to improve the ageing resistance of asphalt binders. Commercial graphite was included as a control modifier for comparison. Operational properties were assessed through rotational viscosity and storage stability, while rheological performance was evaluated using the rutting parameter $G^*/\sin(\delta)$, Bending Beam Rheometer (BBR), Multiple Stress Creep and Recovery (MSCR), $|G^*|$ and δ master curves, rheological ageing index (RAI), and time sweep fatigue tests. BH increased rotational viscosity and mixing and compaction temperatures without compromising storage stability. At high temperatures, BH enhanced rutting resistance by increasing $G^*/\sin(\delta)$ and reducing the non-recoverable creep compliance (J_{nr}), particularly for the finer particle size fraction. At low temperatures, BH increased creep stiffness without significantly affecting the stress relaxation capacity compared with control binders. Master curve and RAI analysis indicated lower susceptibility to long-term ageing-induced stiffening at higher BH contents. BH also improved the fatigue response by increasing accumulated dissipated energy and tended to delay the 50 % reduction of the initial $|G^*|$ compared with the unmodified binder. Based on the combined rheological response, 5.0 wt.% BH with a particle size below $20 \mu\text{m}$ was selected as the final formulation for this type of modifier.

Keywords: Asphalt Binder; Hazelnut Shell Biochar; Particle Size; Biochar Content; Operational Properties; Rheological Properties

1. Introduction

Asphalt binder is a non-renewable resource widely used in pavement construction due to its outstanding adhesion, waterproofing, and mechanical resistance properties [1]. Improving its rheological behaviour and ageing resistance represents one of the main challenges in pavement engineering, as both factors directly affect the durability and maintenance costs of pavement structures [2]. Furthermore, periodic cycles of repair and reconstruction involve high energy and natural resource consumption, as well as increased greenhouse gas emissions, resulting in significant environmental impacts [3], [4]. In this context, various strategies have been developed to optimise the properties of asphalt binders, among which polymer modification and the incorporation of nanomaterials such as graphene and metal oxides stand out [5], [6], [7], [8]. However, global decarbonisation agendas and circular economy strategies have accelerated the development of sustainable asphalt modifiers. Biochar has consequently emerged as a compelling alternative, underpinned by its renewable nature, its potential to valorise agro-industrial residues, and its reduced environmental burden [9], [10], [11]. Biochar is a carbon-rich solid material obtained through the pyrolysis of residual biomass, including wood,

shells, straw, and manure, which would otherwise be incinerated or disposed of as waste [12], [13]. Its physicochemical properties, including porous structure, specific surface area, surface functional groups, and carbon content, depend on both the type of precursor biomass and the pyrolysis process conditions. Although its most widespread application is within the agricultural sector, where it is used to improve soil quality and nutrient retention [14], it has also been explored for water treatment, energy production and storage, environmental remediation, and catalyst support applications [15], [16], [17].

Within pavement engineering, a substantial number of research over the past decade has shown that biochar incorporation can significantly enhance the rheological performance of asphalt binders and improve their resistance to oxidative ageing [18]. In particular, biochar particle size and content have been identified as critical variables governing the effectiveness of the modification and its compatibility with the asphalt matrix. At high service temperatures, particles smaller than 75 μm have been shown to produce greater increases in the rutting parameter $G^*/\sin(\delta)$ and rotational viscosity compared with larger particles ($> 150 \mu\text{m}$) [19], [20]. Furthermore, the optimum particle size is often considered to be comparable to the thickness of the asphalt binder film coating the aggregate, with fractions below 20 μm recommended to minimise interference with binder–aggregate interactions [21].

Regarding biochar content, both the rutting parameter $G^*/\sin(\delta)$ and the complex modulus $|G^*|$ tend to increase as the modifier content increases [20], [22], [23]. For instance, wood-waste biochar incorporated at proportions of 2, 4, and 6 wt.% resulted in average increases in $G^*/\sin(\delta)$ of 4.55 %, 17.49 %, and 45.72 %, respectively, compared with the control binder [20]. Similarly, the incorporation of 15 wt.% switchgrass biochar produced increases of up to 35 % in $|G^*|$ [24]. In addition, biochar incorporation increases the softening point and reduces penetration, thereby enhancing resistance to permanent deformation, which is the principal distress mechanism at high temperatures [22], [24], [25]. At low temperatures, however, adverse effects have been reported, whereby biochar may increase creep stiffness and reduce the m -value in the BBR test, potentially compromising resistance to thermal cracking [21], [26]. Nevertheless, an appropriate selection of particle size and content (particles $< 75 \mu\text{m}$ and contents close to 5 %) has been shown to preserve binder flexibility and maintain balanced performance over a wide temperature range [20], [21], [26].

In this context, previous studies conducted by the authors [27], [28] investigated biochar derived from European hazelnut shells (BH, obtained from the species *Corylus avellana* L.) as an antioxidant biomodifier for asphalt binders, promoting the valorisation of an abundant agricultural residue [29]. Initially, the pyrolysis conditions that provided the most favourable physicochemical and antioxidant properties were identified, demonstrating that BH produced at 300 °C with a residence time of 1 h retained up to 40 % of the original antioxidant activity of the hazelnut shell. This antioxidant activity was confirmed through oxygen radical absorbance capacity (ORAC) measurements, while other antioxidant assays showed consistent trends. In addition, the total phenolic content, determined using the Folin–Ciocalteu method, revealed a high concentration of phenolic compounds, whereas Fourier-transform infrared spectroscopy (FTIR) confirmed the presence of oxygen-containing functional groups, including hydroxyl groups ($-\text{OH}$), in both the precursor biomass and the resulting BH [27]. Subsequently, high-performance liquid chromatography coupled with diode array detection and quadrupole time-of-flight mass spectrometry (HPLC–DAD Q–TOF) enabled the identification and quantification of individual phenolic compounds in BH, providing further evidence of its antioxidant potential [28]. The evaluation of BH-modified asphalt binders under unaged, short-term-aged, and long-term-aged conditions demonstrated reduced susceptibility to oxidative ageing compared with the base binder. In particular, the modified binders exhibited lower carbonyl, sulphoxide, and combined ageing indices, determined by FTIR, with the effect becoming more pronounced after long-term ageing and subsequently corroborated by X-ray photoelectron spectroscopy (XPS) [28]. Nevertheless, the influence of BH particle size and content on these properties has not yet been investigated. Accordingly, the present study evaluates the effect of these variables on the operational and rheological performance of BH-modified asphalt binders under unaged, short-term ageing and long-term ageing conditions. The findings contribute to a better understanding of the role of BH particle size and content in the response of asphalt binders and identify a formulation that provides balanced rheological performance. This study also contributes to the development of cleaner materials by exploring the valorisation of agro-industrial residues through their use as asphalt binder modifiers.

2. Materials and methods

2.1. Materials

A PG 64–22 asphalt binder, with a penetration of 51 dmm, a softening point of 50.7 °C and a rotational viscosity of 0.35 Pa·s at 135 °C, was used as both the control material and the base binder for modification. The biochar (BH) was produced through slow pyrolysis of European hazelnut shells at 300 °C, with a residence time of 1 h and a production yield of 56 wt.%. These conditions were established through a previous optimisation process [27] aimed at preserving the highest possible concentration of antioxidant compounds derived from the shell and ensuring adequate dispersion of BH within the asphalt matrix. Figure 1 schematically illustrates the BH production process and its incorporation into the asphalt binder. Based on the FTIR and X-ray photoelectron spectroscopy (XPS) analyses reported in the previous study [27], panel (d) presents the physicochemical interactions between the surface functional groups of BH and the asphalt binder.

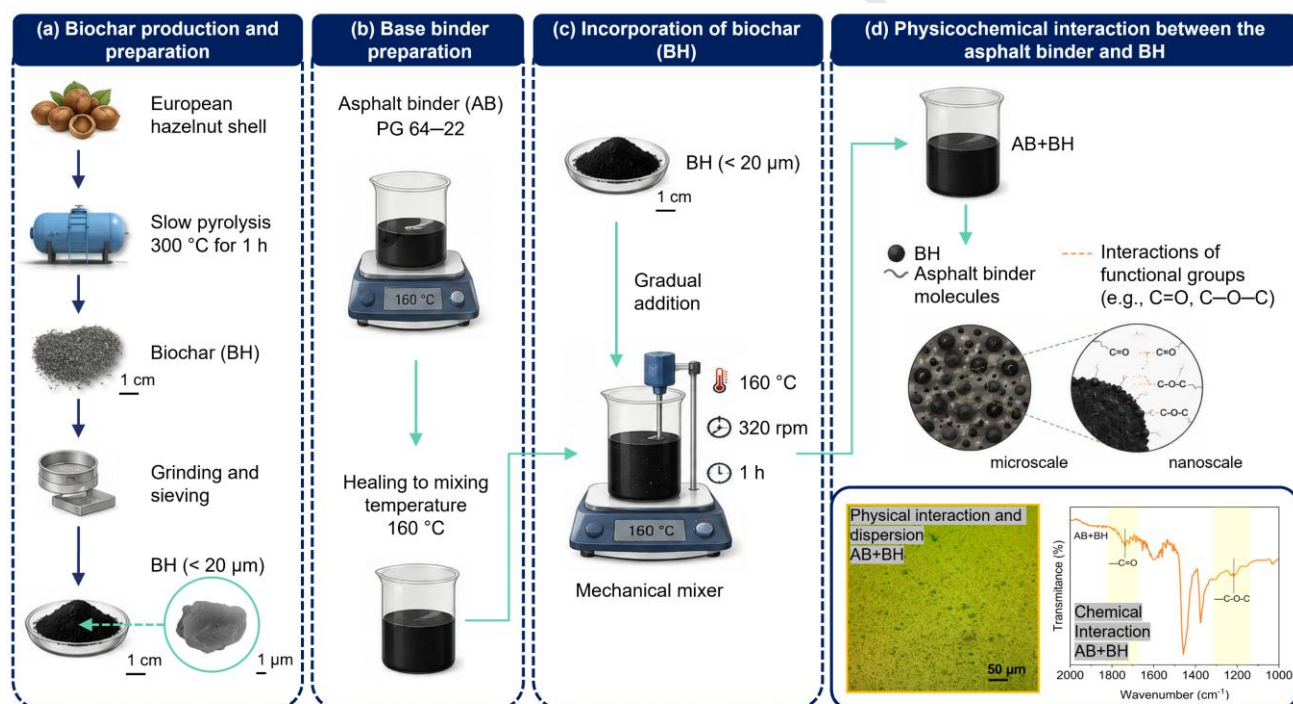


Figure 1. Schematic illustration of the BH production process, its incorporation into the asphalt binder, and the physicochemical interactions between BH and the asphalt binder, based on previous FTIR and XPS analyses [27].

This study evaluated two BH with different particle size distributions, obtained by dry sieving through 20 and 75 µm meshes and hereafter referred to as BH(S) and BH(L), respectively. These particle sizes were selected based on previous studies showing that BH powders prepared using these sieving conditions improved the physical and rheological properties of asphalt binders, outperforming particles larger than 150 µm [20], [21], [30], [31]. Laser diffraction analysis confirmed distinct particle size distributions for the two BH, with BH(L) also containing particles smaller than 20 µm (Figure 2). The morphology of both BH was characterised by scanning electron microscopy (SEM) (Figure 3), while their specific surface area (S_{BET}), pore volume (V_p) and average pore diameter (D_p) are summarised in Table 1. For comparison, a commercial graphite powder (CM) with a particle size below 20 µm was used as a reference modifier.

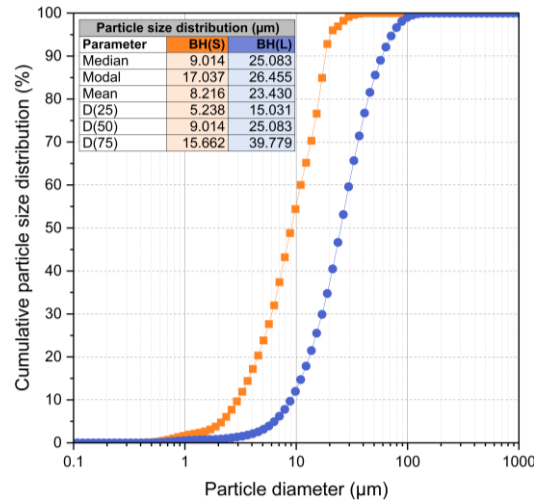


Figure 2. Cumulative particle size distributions of BH(S) and BH(L), determined by laser diffraction in dry dispersion mode, together with their corresponding particle size distribution parameters.

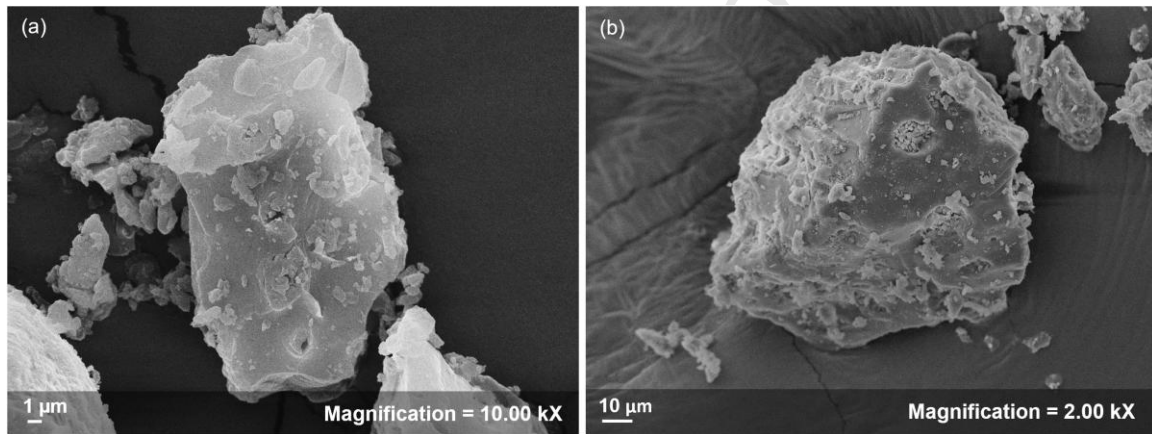


Figure 3. SEM micrographs of the two BH powders with different particle size distributions: (a) BH(S) and (b) BH(L).

Table 1. Specific surface area and pore characteristics of BH(S) and BH(L).

Physical properties	BH(S)	BH(L)
Specific surface area (S_{BET}) (m^2/g)	4.98*	8.66
Pore volume (V_p) (cm^3/g)	0.01*	0.02
Pore diameter (D_p) (nm)	3.86*	1.78

Note: Values marked with an asterisk * were obtained from [27].

2.2. Asphalt binder modification

The modification process was carried out following the procedure described by Martínez-Toledo et al. [27]. In that study, the homogeneous dispersion of BH particles within the asphalt matrix was verified by confocal laser scanning microscopy, and the same mixing protocol was adopted in the present work. Modification was performed using a mechanical mixer at 160 ± 5 °C for 30 min at 320 rpm, progressively incorporating the BH particles. Subsequently, the samples were conditioned in an oven at 120 °C for 6 h to promote binder adsorption into the modifier particles, in accordance with previous studies [25].

Table 2 presents the experimental matrix considered in this study. Samples AB and AB+0 correspond to the PG 64–22 asphalt binder. However, AB+0 was subjected to the modification process without the addition of modifiers. This procedure was adopted in order to isolate the oxidative effect of the modification process from the effect attributable to the modifiers themselves. Additionally, AB+CM corresponds to the asphalt binder

incorporating the commercial modifier. The BH-modified samples were labelled as follows: AB+BHXX(Y), where XX refers to the BH content expressed as a percentage of the asphalt binder mass (2.5, 5.0, or 7.5 wt.%), and Y refers to the particle size fraction: small (S: < 20 μm) or large (L: < 75 μm).

Table 2. Experimental matrix used for asphalt binder sample preparation.

Sample	Modifier type	Particle size (μm)	Modifier content (wt. %)
AB	—	—	—
AB+0	—	—	—
AB+CM	CM	< 20	5.0
AB+BH2.5(S)	BH	< 20	2.5
AB+BH5.0(S)	BH	< 20	5.0
AB+BH7.5(S)	BH	< 20	7.5
AB+BH2.5(L)	BH	< 75	2.5
AB+BH5.0(L)	BH	< 75	5.0
AB+BH7.5(L)	BH	< 75	7.5

The BH particle size and content were selected based on previous studies, as the adopted conditions have demonstrated balanced performance in terms of rutting resistance, fatigue, and cracking for biochars derived from different feedstocks [19], [24], [26], [31]. In particular, the available evidence suggests that low modification levels (≤ 2.5 wt.%) generally result in negligible changes in binder behaviour, whereas higher contents (> 7.5 wt.%), while enhancing rutting resistance, may induce excessive stiffening and increase susceptibility to low-temperature cracking. The CM content (5 wt.%) was established according to the manufacturer's recommendations and previous findings regarding its influence on the physical and rheological properties of asphalt binders [20], [32], [33], [34].

2.3. Experimental methodology

The experimental programme was structured in two stages. Firstly, the operational properties of all samples (Table 2) were evaluated to determine their suitability for industrial-scale application. Subsequently, the rheological properties of the asphalt binders were characterised at high and low service temperatures to compare the performance of the two BH with different particle size distributions (S: < 20 μm ; L: < 75 μm) and to select the particle size fraction for the subsequent stage of the study. Based on this sequential experimental procedure, additional viscoelastic parameters were then analysed to support the selection of the BH content (2.5, 5.0 or 7.5 wt.%) for detailed rheological evaluation. The methodological framework is presented in Figure 4.

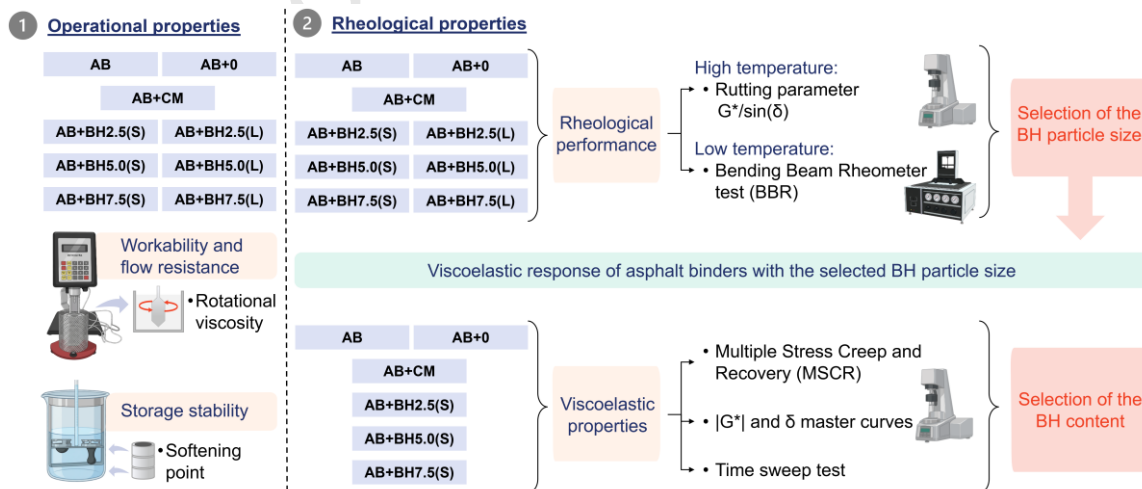


Figure 4. Methodological framework of the study.

2.3.1. Stage 1: Operational properties

The rotational viscosity of the asphalt binders was evaluated at 120, 135, 150 and 160 °C using a Brookfield viscometer according to AASHTO T316–19 [35]. Mixing and compaction temperatures were determined based on the equiviscous principle (ASTM D2493), corresponding to viscosities of 0.17 ± 0.02 Pa·s and 0.28 ± 0.03 Pa·s, respectively [36]. Storage stability was evaluated following ASTM D7173 [37]. The modified binders were poured into aluminium tubes and conditioned in a vertical position at 163 ± 5 °C for 48 h. The tubes were then cooled, sectioned into three equal parts, and the softening points of the upper and lower sections were determined according to ASTM D36 [38] using the ring-and-ball (R&B) method. Storage stability was assessed from the difference between the softening points of the upper and lower sections ($\Delta R\&B$). A binder was considered to exhibit adequate storage stability when $\Delta R\&B$ was lower than 2.5 °C [39], [40]. All measurements parameters were determined from three replicates.

2.3.2. Stage 2: Rheological properties

Short-term ageing was simulated using the Rolling Thin Film Oven (RTFO) test, in accordance with AASHTO T240 [41], whereas long-term ageing was simulated using the Pressure Ageing Vessel (PAV) was performed on samples previously aged using the RTFO procedure, following the procedure specified in AASHTO R28 [42].

Resistance to permanent deformation at high temperatures was evaluated on RTFO-aged binders using the rutting parameter $G^*/\sin(\delta)$, determined with a Dynamic Shear Rheometer (DSR) in accordance with AASHTO T315 [43]. All binders were tested at the high PG temperature of the control binder AB (64 °C) to enable a direct comparison of their rutting resistance under equivalent conditions.

Low-temperature performance was characterised using the BBR test, in accordance with AASHTO T313 [44], on PAV-aged binders. Creep stiffness and the relaxation parameter (m-value) were determined at the low PG temperature of the control binder AB (–12 °C), using the Superpave threshold values as evaluation criteria. All binders were evaluated at the same temperature to enable a direct comparison of their low-temperature response.

The MSCR test was conducted at 64 °C on RTFO-aged binders, in accordance with AASHTO T350 [45]. The non-recoverable creep compliance (J_{nr}), percentage recovery (R), and accumulated strain were determined at stress levels of 0.1 and 3.2 kPa, together with the $J_{nr,diff}$ parameter and the associated traffic classification.

Master curves of the complex modulus ($|G^*|$) and phase angle (δ) were obtained for unaged, RTFO-aged, and PAV-aged binders using frequency sweep tests (0.1–100 rad/s) performed in a DSR over temperature ranges of 5 to –25 °C (0.05 % strain, 4 mm plates) and 5 to 75 °C (0.5 % strain, 8 and 25 mm plates). The master curves were generated using the time–temperature superposition principle, with the horizontal shift factors (a_T) determined according to the Williams–Landel–Ferry (WLF) model [46]. The horizontal shift factors were determined by iterative least-squares optimisation to maximise the overlap between adjacent isothermal frequency sweep curves and subsequently fitted to the WLF equation. For all binders, the WLF constants were within the typical ranges reported for asphalt binders ($C_1 \approx 11 - 18$, $C_2 \approx 85 - 137$). The reduced angular frequency was calculated by multiplying the experimental angular frequency by the corresponding a_T . Reference temperatures of –10 °C and 25 °C were used for the 5 to –25 °C and 5 to 75 °C ranges, respectively. **Tables S.1** and **S.2** present the WLF fitting parameters and their corresponding coefficients of determination (R^2). To further evaluate the rheological changes associated with ageing, the rheological ageing index (RAI) was calculated as the ratio of $|G^*|$ after ageing to that of the corresponding unaged binder, as expressed in Equations (1) and (2):

$$RAI_{RTFO} = \frac{|G^*|_{RTFO}}{|G^*|_{unaged}} \quad (1)$$

$$RAI_{PAV} = \frac{|G^*|_{PAV}}{|G^*|_{unaged}} \quad (2)$$

The RAI was calculated at the same experimental temperature and angular frequency for each aged binder and its corresponding unaged condition, without applying time–temperature shifting. RAI values greater than unity indicate an increase in $|G^*|$ compared with the corresponding unaged condition.

Fatigue resistance was evaluated through a time sweep test under controlled-strain conditions using the DSR by determining the evolution of $|G^*|$ with the number of loading cycles [47], [48]. The tests were conducted on PAV-aged binders at the intermediate PG temperature of the control binder (25 °C), at a loading frequency of 10 Hz. The applied strain level was previously determined through an amplitude sweep test [48]. The accumulated dissipated energy was calculated by summing the energy dissipated in each cycle, according to Equations (3) and (4) [49].

$$\sum_{i=1}^n W_i \quad (3)$$

$$W_i = \pi G_i^* \varepsilon_i^2 \sin(\delta_i) \quad (4)$$

where n corresponds to the total number of loading cycles; W_i is the dissipated energy in cycle i (J/m³); G_i^* is the complex modulus in cycle i (Pa); ε_i is the applied strain in cycle i ; and δ_i is the phase angle in cycle i . Strain values were expressed in decimal form and phase angles in radians.

All measured parameters were determined from three replicate specimens obtained from the same prepared binder batch.

2.4. Statistical analysis

Analysis of variance (ANOVA) was performed on the operational and rheological properties of the asphalt binders at a significance level of 0.05 (p -value < 0.05). One-way ANOVA was used to compare the individual binder formulations, followed by Tukey's honestly significant difference (HSD) post hoc test when statistically significant differences were identified. For the BH-modified binders, two-way ANOVA was additionally performed to evaluate the effects of particle size, BH content, and their interaction on the rutting parameter $G^*/\sin(\delta)$, creep stiffness, and m -value. Each asphalt binder formulation was prepared as a single batch, from which the specimens required for each test were obtained. All tests were performed using three replicate specimens ($n = 3$). Thus, the reported variability represents the reproducibility of measurements obtained from the same batch. Results are presented as means $\pm$ standard deviations (SD).

3. Results and discussion

3.1. Effect of biochar addition on the operational properties of asphalt binders

Figure 5(a) shows that rotational viscosity increased as a result of both the modification process and the incorporation of CM and BH, in agreement with previous studies [20], [24], [28]. However, as the temperature increased, these differences decreased, likely because the higher molecular mobility of the binder at elevated temperatures reduced the influence of the particulate modifiers on flow resistance. At 120 °C, the highest increases were observed, with statistically significant differences identified among the samples (ANOVA, p -value < 0.001; **Table S.3**), as confirmed by Tukey's test (Figure 5(b)). In particular, AB+CM exhibited a viscosity approximately 17 % higher than AB and 9 % higher than AB+0. Nevertheless, its viscosity was slightly lower than those obtained for the BH-modified binders prepared using the same modifier dosage (5 wt.%). For the BH-modified binders, viscosity increases ranged from 12 to 27 % relative to AB and from 4 to 18 % relative

to AB+0, depending on BH particle size and content. The highest increase was observed for AB+BH7.5(S), with an increase of approximately 27 % relative to AB and 18 % relative to AB+0. These results indicate that, although the modification process itself induces an increase in flow resistance, the incorporation of BH produces an additional effect, which was more pronounced at smaller particle sizes and higher BH contents. The influence of particle size is consistent with that reported in other studies [19], [20], where smaller particles exhibited a greater effect on viscosity enhancement.

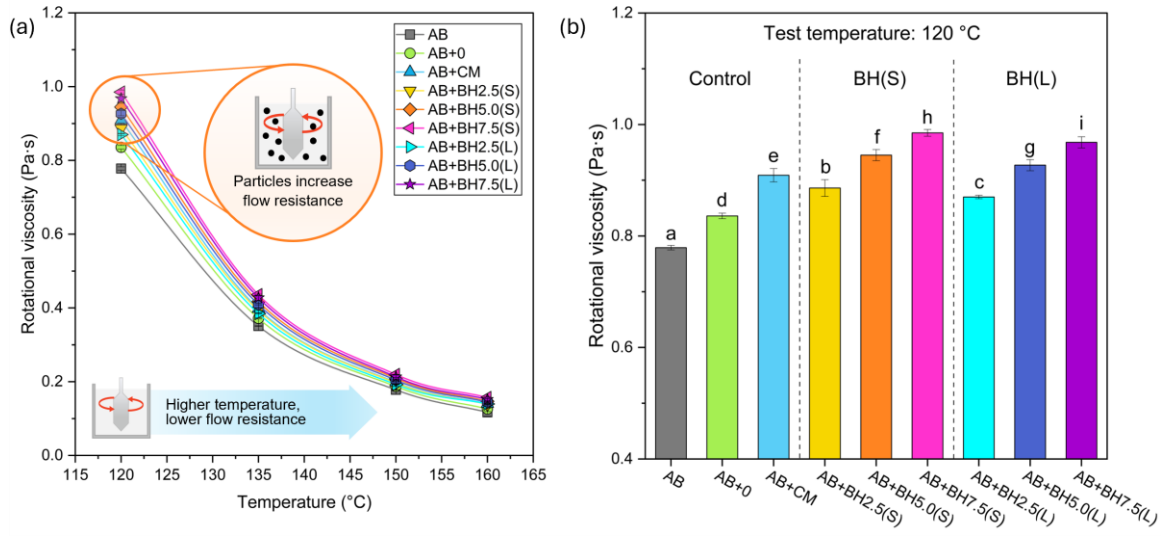


Figure 5. Results of flow resistance and workability: (a) rotational viscosity at 120, 135, 150, and 160 °C; and (b) rotational viscosity evaluated at 120 °C including the statistical classification through Tukey's post hoc test.

Table 3 presents the mixing and compaction temperatures of the binders, whose coefficients of determination ($R^2 > 0.99$) confirm an adequate fit of the linear model within the evaluated temperature range. Compared with AB, sample AB+0 exhibited a slight increase in both temperatures (+1 °C), suggesting a viscosity increase associated with the thermal ageing during the modification process [28]. Overall, the results indicate that both CM and BH increase the viscosity of the asphalt binder and, consequently, require higher mixing and compaction temperatures. The effect of BH depended on its content, with increases of up to 5 °C relative to AB observed for the highest BH content (7.5 wt.%). In contrast, particle size (S and L) did not show relevant differences to produce noticeable changes in mixing and compaction temperatures within the analysed range.

Table 3. Mixing and compaction temperature ranges determined using the equiviscous principle.

Sample	Mixing (°C)	Compaction (°C)	Equation	R^2
AB	149 – 154	139 – 143	$y = -0.0206x + 2.3461$	0.998
AB+0	150 – 155	140 – 144	$y = -0.0206x + 2.3711$	0.996
AB+CM	152 – 157	142 – 147	$y = -0.0204x + 2.3874$	0.993
AB+BH2.5(S)	152 – 157	141 – 146	$y = -0.0203x + 2.3595$	0.993
AB+BH5.0(S)	153 – 159	143 – 148	$y = -0.0203x + 2.3938$	0.994
AB+BH7.5(S)	154 – 159	144 – 148	$y = -0.0201x + 2.3801$	0.992
AB+BH2.5(L)	152 – 157	141 – 146	$y = -0.0200x + 2.3200$	0.991
AB+BH5.0(L)	153 – 158	142 – 147	$y = -0.0203x + 2.3808$	0.993
AB+BH7.5(L)	154 – 159	143 – 148	$y = -0.0203x + 2.3951$	0.993

Note: $y = \log_{10}(\text{rotational viscosity})$; $x = \text{temperature (°C)}$; $R^2 = \text{coefficient of determination}$.

Figure 6 presents the softening point (R&B) results measured in the upper and lower sections of the binders after the storage stability test, together with the difference between both sections ($\Delta R\&B$). One-way ANOVA of the individual $\Delta R\&B$ values indicated no statistically significant differences among the evaluated binders (p -

value = 0.663; **Table S.3**), suggesting comparable storage stability under the conditions evaluated. Moreover, all samples exhibited $\Delta R\&B$ values lower than 2.5 °C, a criterion commonly accepted as indicative of good storage stability [39], [40]. Accordingly, the storage stability test did not detect a significant difference in softening point ($\Delta R\&B$) between the upper and lower sections of the BH-modified binders, even at the highest BH content evaluated (7.5 wt.%).

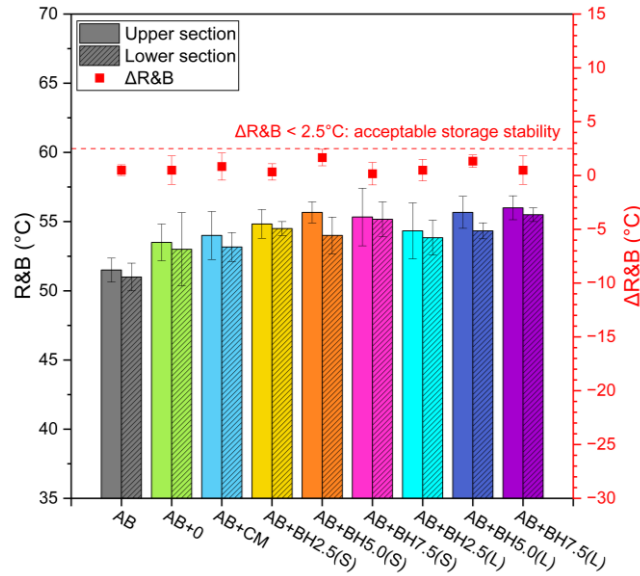


Figure 6. Softening point (R&B) of the upper and lower binder sections after the storage stability test and their difference ($\Delta R\&B$).

3.2. Effect of biochar addition on the rheological properties of asphalt binders

At high service temperatures, Figure 7(a) shows that the rutting parameter $G^*/\sin(\delta)$ increased as a consequence of both the modification process and the incorporation of modifiers, with statistically significant differences observed among the samples (one-way ANOVA, p -value < 0.001; **Table S.4**), with pairwise differences identified by Tukey's test (Figure 7(a)). In particular, AB+0 exhibited an increase of approximately 5 % relative to AB, suggesting that the modification process induces slight oxidative stiffening of the binder. The addition of BH produced a further increase in the parameter, reaching values approximately 10 – 11 % higher than AB+0 when the S particle size was used. For the BH-modified binders, two-way ANOVA (**Table S.5**) showed significant effects of particle size and BH content on $G^*/\sin(\delta)$ (p -value < 0.001), together with a significant particle size x content interaction (p -value < 0.001). The S particle size produced higher $G^*/\sin(\delta)$ values than the L particle size, particularly at lower BH contents, with the effects of particle size decreasing as BH content increased. The binders modified with BH(S) also exhibited $G^*/\sin(\delta)$ values comparable to those of AB+CM, particularly AB+BH5.0(S), with no statistically significant differences according to Tukey's test. Finally, all binders satisfied the Superpave specification for the rutting parameter ($G^*/\sin(\delta) \geq 2.2$ kPa at 64 °C after RTFO ageing), indicating that the incorporation of BH, especially at the S particle size, may improve rutting resistance while satisfying the specification requirements. This behaviour is consistent with that reported in other studies [19], [20], where smaller particle sizes promoted greater improvements in rheological performance.

At low service temperatures, Figure 7(b) presents the BBR test results, revealing statistically significant differences among the binders for both creep stiffness and m -value (one-way ANOVA, **Table S.4**), with p -values lower than 0.001 and 0.002, respectively. Creep stiffness increased with the incorporation of BH, particularly at 7.5 wt.%, with increases of approximately 15 % for AB+BH7.5(S) and 10 % for AB+BH7.5(L) relative to the control binders. In contrast, no significant differences in the m -value were observed between the BH-modified binders and the control binders according to Tukey's test (Figure 7(b)). Among the BH-modified binders, two-

way ANOVA (**Table S.5**) showed significant effects of both particle size and BH content on creep stiffness (p -value < 0.001), whereas their interaction was not significant (p -value = 0.803). Creep stiffness increased with BH content for both particle sizes, with higher values observed for the S particle size than for the L particle size. Similarly, both particle size (p -value = 0.013) and BH content (p -value = 0.005) significantly affected the m -value, while their interaction was not significant (p -value = 0.932). Nevertheless, the magnitude of these changes was small, with mean m -values ranging from 0.305 to 0.314 across the BH formulations. These results indicate that the influence of BH on low-temperature performance was mainly reflected in creep stiffness, while changes in the m -value remained small. Previous studies have reported increases in creep stiffness together with reductions in the m -value relative to the control binder following biochar incorporation [24], [26]. However, although creep stiffness increased in the present study, the m -value did not differ significantly between the BH-modified and control binders. Moreover, all binders satisfied the Superpave criteria (creep stiffness ≤ 300 MPa and m -value ≥ 0.300) at the evaluated temperature, indicating that the observed changes did not prevent compliance with the low-temperature specification limits.

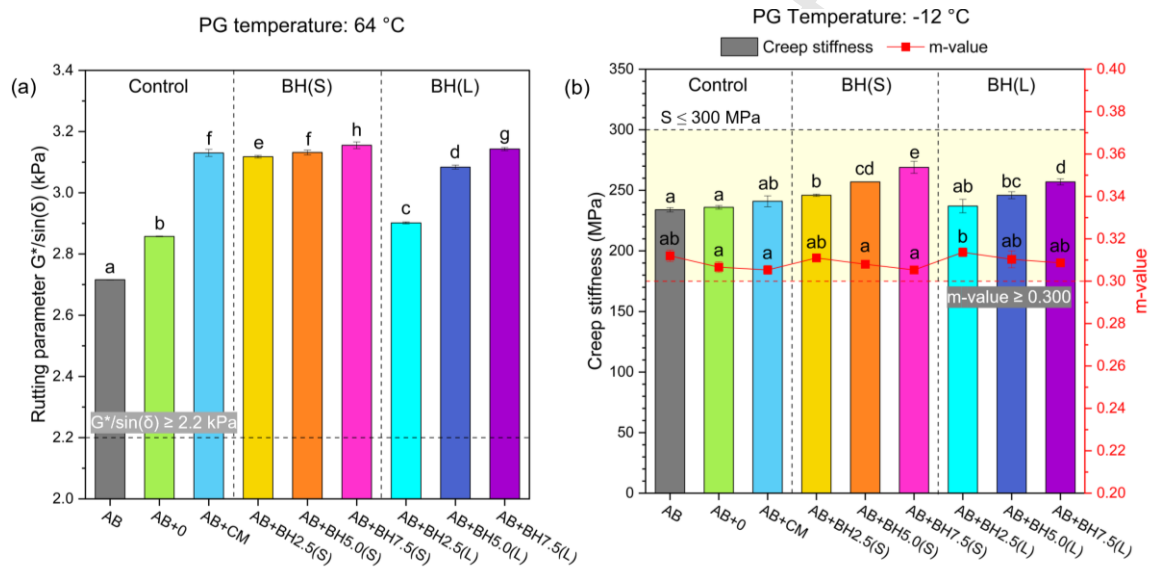


Figure 7. High- and low-temperature rheological performance: (a) rutting parameter $G^*/\sin(\delta)$ at 64 °C and (b) BBR creep stiffness and m -value at -12 °C for control and BH-modified binders (S and L). Statistical groupings from Tukey's post hoc test are included.

Overall, the results indicate that particle size and BH content both influenced binder performance, although their effects differ between high- and low-temperature responses. The S particle size provided the most favourable response at high temperature, producing higher $G^*/\sin(\delta)$ values, particularly at lower BH contents. At low temperature, the S particle size resulted mainly in a slight increase in creep stiffness, while changes in the m -value remained small and no significant differences were observed between the BH-modified and control binders. Moreover, all BH-modified binders remained within the Superpave BBR limits. Therefore, considering the rheological properties evaluated in this study, BH with the S particle size was selected because it provided the most favourable high-temperature response while maintaining adequate low-temperature performance.

3.3. Viscoelastic response of asphalt binders modified with the selected BH particle size

Using the selected BH particle size ($S < 20 \mu\text{m}$), the viscoelastic response of the binder was evaluated at different modifier contents (2.5, 5.0, and 7.5 wt.%). For this purpose, the MSCR test was performed at two stress levels (0.1 and 3.2 kPa), and the results are presented in Figure 8. Figure 8(a) shows that the incorporation of CM and BH significantly reduced $J_{nr0.1}$, as supported by the ANOVA results (p -value < 0.001 ; **Table S.6**). In this context, AB and AB+0 exhibited the highest values, whereas the modified binders showed reductions of approximately 22 % and were grouped within the same statistical subset according to Tukey's test. In contrast, $R_{0.1}$ did not

exhibit significant differences (ANOVA, p -value = 0.050; **Table S.6**), indicating that the observed improvement was mainly associated with a reduction in the viscous component rather than with an increase in the elastic character of the binder. Figure 8(b) shows that the $Jnr_{3.2}$ parameter followed the same trend, with reductions of approximately 19 % in the binders modified with CM and BH relative to AB and AB+0. ANOVA revealed statistically significant differences among the samples (p -value < 0.001; **Table S.6**), although no significant differences were identified among the different BH-modified binders, suggesting a saturation effect in rheological improvement under the evaluated conditions. Although these results confirm the effectiveness of the modifiers in enhancing rutting resistance under high stress conditions [21], [30], [50], their traffic classification according to AASHTO M332 remained within the standard category (S: $2 < Jnr_{3.2} \leq 4.5$, $Jnr,diff < 75$ %; Table 4) [51]. Regarding the $R_{3.2}$ parameter, all evaluated binders exhibited predominantly viscous behaviour under high stress levels, indicating a low elastic recovery capacity.

Table 4. Non-recoverable creep compliance difference ($Jnr,diff$) of the evaluated asphalt binders.

Sample	$Jnr,diff$ (%)
AB	26.73 ± 0.79
AB+0	24.40 ± 0.80
AB+CM	22.90 ± 0.81
AB+BH2.5(S)	26.65 ± 0.79
AB+BH5.0(S)	29.09 ± 0.77
AB+BH7.5(S)	27.83 ± 0.78

Note: Values are presented as mean $\pm$ standard deviation (SD).

Figure 8(c) presents the evolution of accumulated strain for the evaluated binders at 0.1 and 3.2 kPa during the MSCR test, showing the characteristic stepwise behaviour associated with loading and recovery cycles. At both stress levels, AB and AB+0 exhibited the highest strain values, whereas the binders containing modifiers showed reduced strain accumulation, consistent with their lower Jnr values (Figure 8(a) and 8(b)). At 3.2 kPa, the binders modified with BH exhibited reductions in accumulated strain ranging from 18 to 25 % relative to AB, depending on BH content, reflecting a greater separation between the curves and a more pronounced non-linear response under increased loading. This behaviour is consistent with the increase in binder stiffness associated with the presence of solid particles that restrict deformation under load [28]. Although differences among the BH-modified binders were apparent in the accumulated strain curves, the statistical analysis of $Jnr_{0.1}$ and $Jnr_{3.2}$ did not identify significant differences among the evaluated BH contents. AB+CM exhibited accumulated strain comparable to that of AB+BH7.5(S), with reductions in accumulated strain close to 25 %. These results indicate that, under the evaluated conditions, a higher BH content was required to achieve a mechanical response comparable to that of the CM-modified binder.

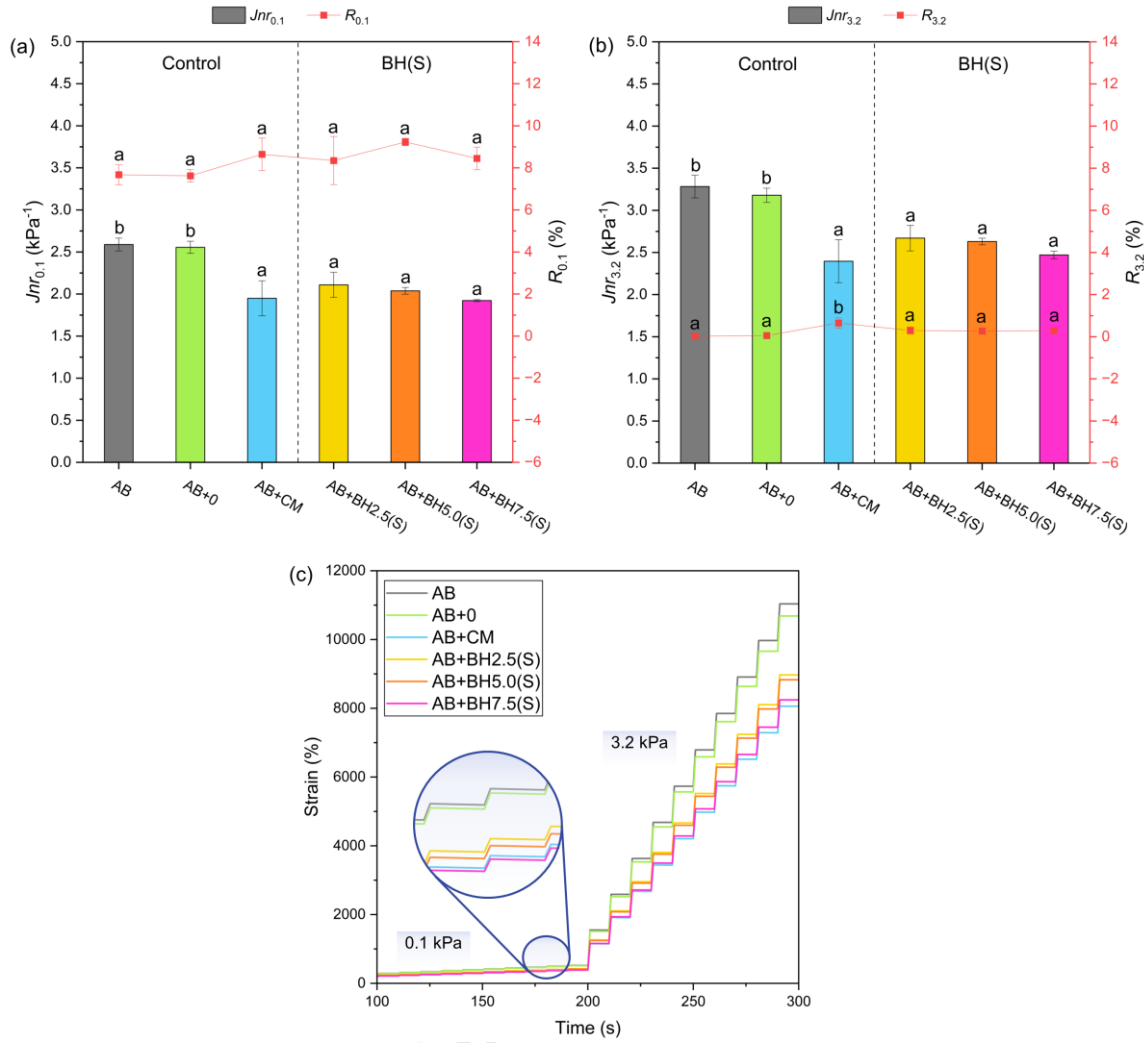


Figure 8. MSCR test results at 64 °C: (a) Jnr and (b) R values at 0.1 and 3.2 kPa, and (c) accumulated strain evolution under both stress levels. Statistical groupings from Tukey's post hoc test are included in (a) and (b).

The master curves presented in Figures 9 and 10 provide a detailed analysis of the rheological evolution of the binders under different ageing conditions (unaged, RTFO and PAV). The results are evaluated within the linear viscoelastic regime as a function of reduced angular frequency at the corresponding reference temperatures.

The master curves of the complex modulus $|G^*|$ and phase angle δ obtained for the 5 to -25 °C temperature range, shown in Figure 9, reveal the typical viscoelastic behaviour of the binders, characterised by an increase in stiffness and a decrease in phase angle with increasing reduced angular frequency. This reflects the transition from a viscoelastic regime with viscous predominance at low frequencies ($< 10^{-2}$ rad/s) towards a predominantly elastic response at high frequencies ($> 10^2$ rad/s). In the unaged condition (Figure 9(a) and 9(b)), $|G^*|$ values ranged from 10^6 to 10^9 Pa, whilst δ decreased from approximately 50 ° to approximately 5 °. The $|G^*|$ curves exhibited similar shapes among the binders, with slight separation in the intermediate-to-high frequency range, where AB+BH7.5(S) showed slightly higher stiffness, consistent with the greater BH content and the resulting contribution of the particulate phase [28]. The δ values were comparable among all samples, except for AB+0, indicating that the BH-modified binders exhibited a viscoelastic response similar to that of AB.

Following RTFO ageing (Figure 9(c) and 9(d)), a general increase in $|G^*|$ was observed, particularly at low frequencies, consistent with oxidation and the loss of volatile fractions associated with short-term ageing [52]. In this case, the physical reinforcing effect attributed to the BH particles remained evident in AB+BH7.5(S), which maintained the highest stiffness levels under this ageing condition. Simultaneously, the reduction in δ

values indicated an increase in elastic character, particularly for AB+CM at low frequencies. This behaviour may be attributed to the high specific surface area of CM [21], which could promote binder–filler physical interactions and contribute to the reinforcement of the elastic component of the binder.

PAV ageing produced the most evident rheological differences within the 5 to –25 °C range (Figure 9(e) and 9(f)), with higher $|G^*|$ values and greater separation among the binder curves than under the unaged and RTFO-aged conditions. After PAV ageing, AB exhibited the lowest $|G^*|$ values, followed by AB+0, whereas the incorporation of CM and BH resulted in higher stiffness. For the BH-modified binders, $|G^*|$ generally increased with modifier content, indicating that the reinforcing contribution of the particles remained evident after long-term ageing. Nevertheless, the δ curves showed similar trends among the binders across the frequency spectrum, indicating that the differences were more pronounced in $|G^*|$ than in the balance between the viscous and elastic components of the material. This observation is consistent with the BBR results (Figure 7(b)), where BH increased creep stiffness without significantly affecting the m -value compared with the control binders. However, the higher $|G^*|$ values observed after ageing may reflect both the reinforcing effect of the modifiers and the changes induced by ageing. Therefore, the rheological ageing index (RAI), shown in Figure 11, was used to assess the change in $|G^*|$ with respect to the corresponding unaged binder.

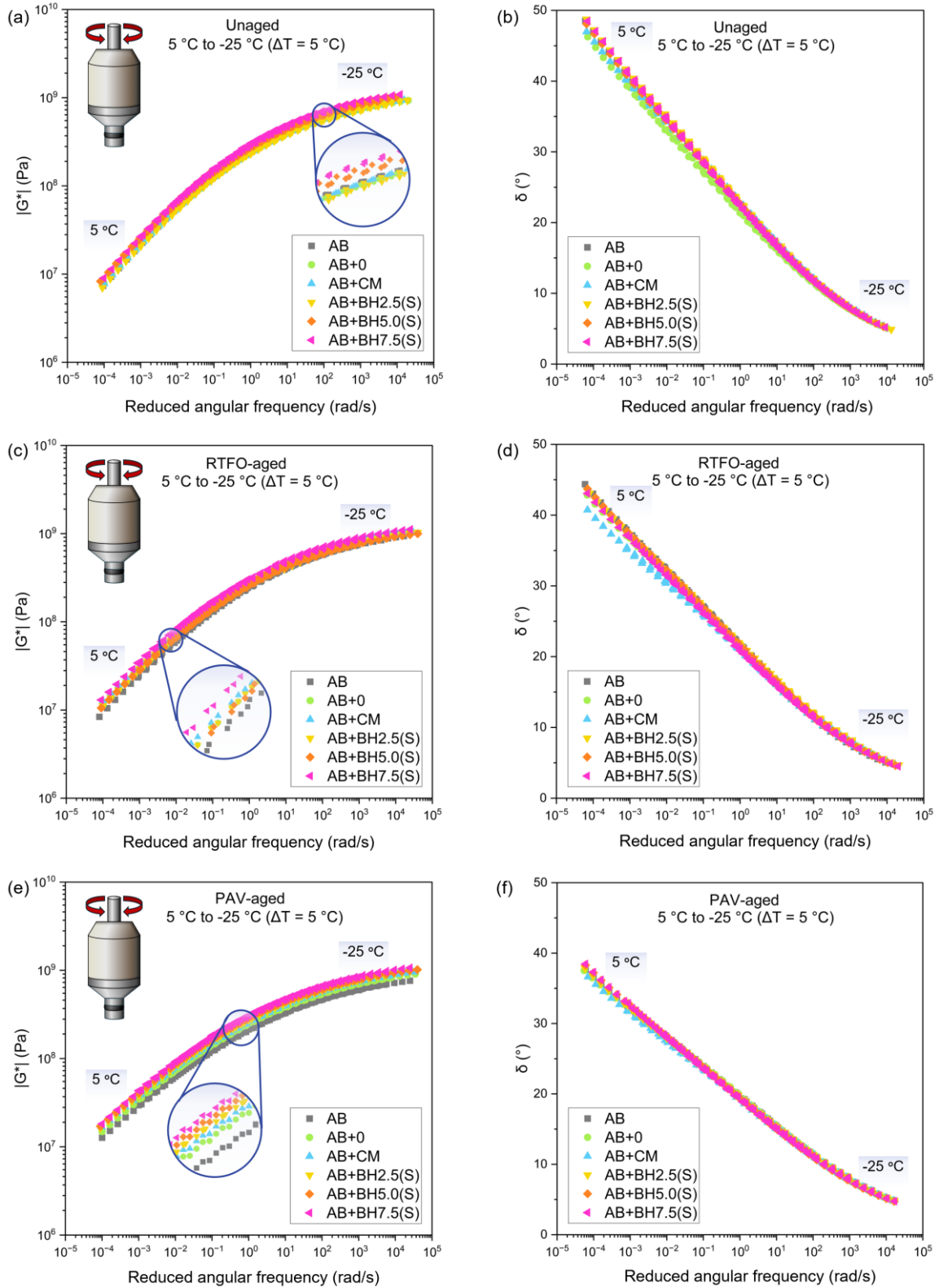


Figure 9. Master curves of $|G^*|$ and δ versus reduced angular frequency for binders tested from 5 to -25 °C under the following conditions: (a) and (b) unaged, (c) and (d) RTFO-aged, and (e) and (f) PAV-aged.

The master curves obtained for the 5 to 75 °C temperature range, shown in Figure 10, present the binder response under intermediate- and high-temperature service conditions, where viscous behaviour predominates at low reduced angular frequencies ($< 10^{-2}$ rad/s). In the unaged condition (Figure 10(a) and 10(b)), both $|G^*|$ and δ exhibited similar values for the BH-modified binders and the control binder (AB), indicating that the

incorporation of BH did not produce substantial variations in stiffness or viscoelastic balance within this thermal range.

Following RTFO ageing (Figure 10(c) and 10(d)), the BH-modified binders exhibited $|G^*|$ values similar to those of AB, whilst AB+CM showed higher stiffness in the low-frequency region. This behaviour may be associated with the physical reinforcing effect of the CM particles, which can restrict the mobility of the surrounding binder molecules and increase resistance to deformation under loading. Regarding the δ values, all binders converged towards similar values at high frequencies. In contrast, at intermediate frequencies (approximately between 10^{-5} and 10^1 rad/s), AB+CM exhibited δ values comparable to those of AB+0, whereas the BH-modified binders showed an increase in δ with increasing modifier content, reaching values close to those of AB at 5 and 7.5 wt.% BH. This indicates that the binders containing higher BH contents retained a greater viscous contribution than AB+0 and AB+CM after RTFO ageing. This behaviour was further evaluated using the experimental frequency sweep data obtained directly from the DSR at 55 °C. The δ values at an angular frequency of 6.28 rad/s (1 Hz), corresponding to the frequency range highlighted in the master curve, were obtained by interpolation between the two adjacent experimental frequencies. ANOVA revealed statistically significant differences among the binders (p -value < 0.001; **Table S.6**), while Tukey's test identified the differences shown in Figure 12(a). Finally, at low frequencies, the differences among the binders tended to diminish due to the proximity to the viscous limit ($\delta \approx 90^\circ$).

Following PAV ageing (Figure 10(e) and 10(f)), the BH-modified binders showed lower $|G^*|$ values than AB+0 and AB+CM in the intermediate- and low-frequency ranges, approaching those of AB. Differences were also observed in the δ curves at intermediate frequencies (approximately between 10^{-5} and 10^1 rad/s), where δ tended to increase with BH content. In particular, the binders containing 5 and 7.5 wt.% BH showed higher δ values than AB+0 and AB+CM, indicating a greater viscous contribution within this frequency range. These differences were further evaluated using the experimental frequency sweep data obtained directly from the DSR at 55 °C. The δ values at an angular frequency of 6.28 rad/s (1 Hz), corresponding to the frequency range highlighted in the master curve, were obtained by interpolation between the two adjacent experimental frequencies. ANOVA revealed statistically significant differences among the binders (p -value < 0.001; **Table S.6**). Tukey's test further showed that the binders containing 5 and 7.5 wt.% BH exhibited significantly higher δ values than the control binders (Figure 12(b)), indicating a greater viscous contribution after PAV ageing. In contrast, AB+CM exhibited the lowest δ values, indicating a more elastic response. Finally, at high frequencies, all samples converged towards similar δ values, whereas at low frequencies the differences tended to diminish as δ approached the viscous limit ($\delta \approx 90^\circ$).

Figure 11 presents the RAI as a function of experimental angular frequency for the binders after RTFO and PAV ageing at -25, 5 and 55 °C. At -25 °C (Figure 11(a) and 11(b)), RAI values remained close to unity for all binders under both ageing conditions, with only minor variations over the angular frequency range evaluated. This behaviour indicates that $|G^*|$ was less sensitive to ageing-induced changes at -25 °C, where the high stiffness and predominantly elastic response of the binders may attenuate the differences between the unaged and aged conditions. At 5 °C (Figure 11(c) and 11(d)), the RAI exhibited a stronger dependence on angular frequency, with higher values at low frequencies that progressively decreased as frequency increased. This behaviour was more pronounced after PAV ageing, indicating greater changes in $|G^*|$ relative to the unaged condition, particularly at low frequencies. Differences among the binders were also more evident in this region. Following RTFO ageing, AB+CM and AB+BH2.5(S) exhibited the highest RAI values, whereas increasing the BH content to 5 and 7.5 wt.% was associated with lower index values. The most pronounced changes were observed at 55 °C (Figure 11(e) and 11(f)), where the RAI increased considerably towards lower frequencies, particularly after PAV ageing. At this temperature, greater molecular mobility, particularly at low frequencies, may favour a more pronounced manifestation of ageing-induced changes in binder stiffness, thereby making differences among the samples more evident. AB+CM and AB+BH2.5(S) exhibited the highest RAI values, whereas AB+BH5.0(S) and AB+BH7.5(S) showed lower values comparable to those of AB, despite their higher particulate content and the associated contribution to binder reinforcement. These results are consistent with

previous studies [27], [28], which reported lower oxidation indices and a lower abundance of oxygen-containing functional groups in BH-modified binders, suggesting that the previously reported antioxidant activity of BH may contribute to limiting ageing-related hardening even as the particle content in the binder increases.

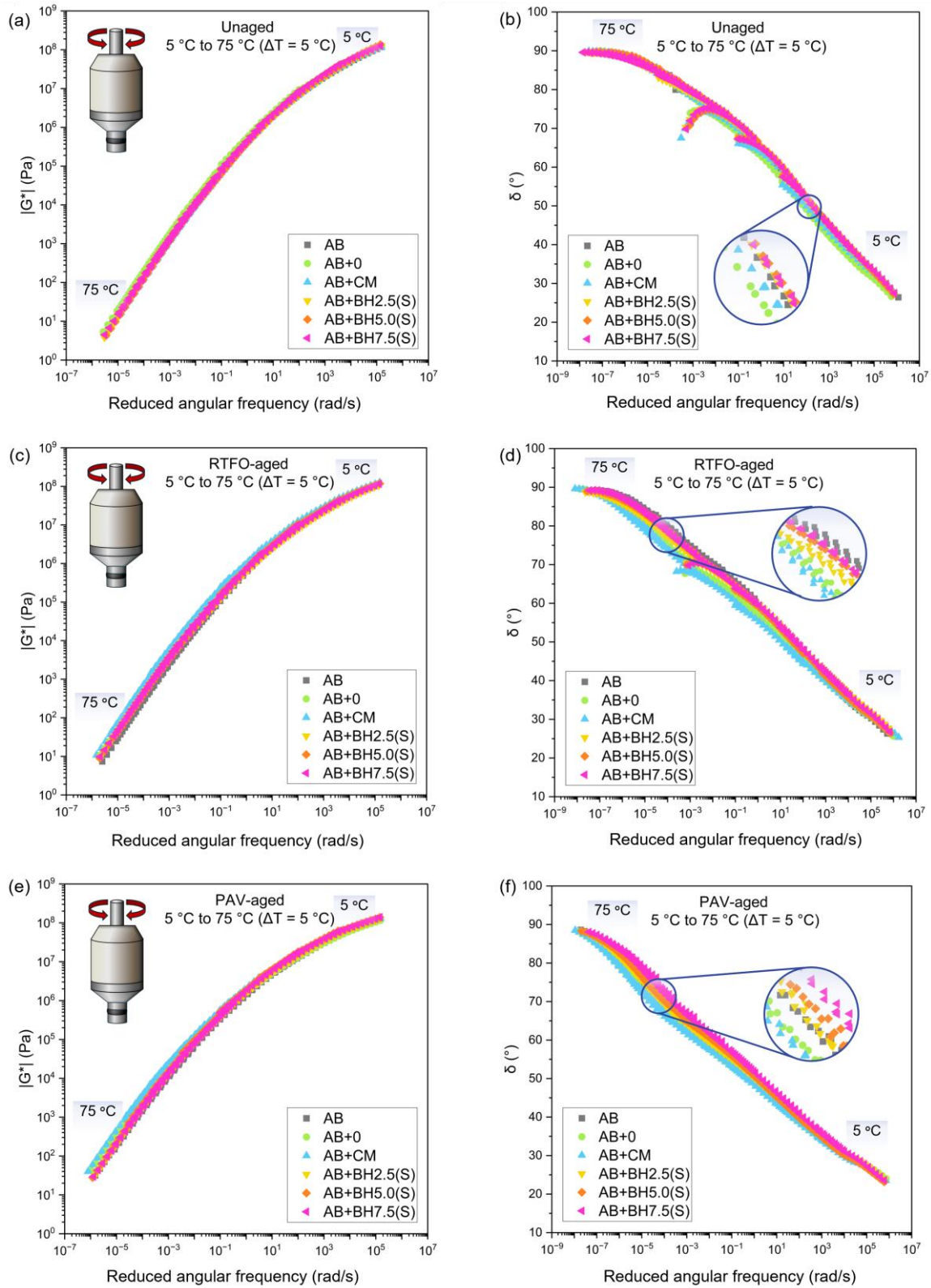


Figure 10. Master curves of $|G^*|$ and δ versus reduced angular frequency for binders tested from 5 to 75 °C under the following conditions: (a) and (b) unaged, (c) and (d) RTFO-aged, and (e) and (f) PAV-aged.

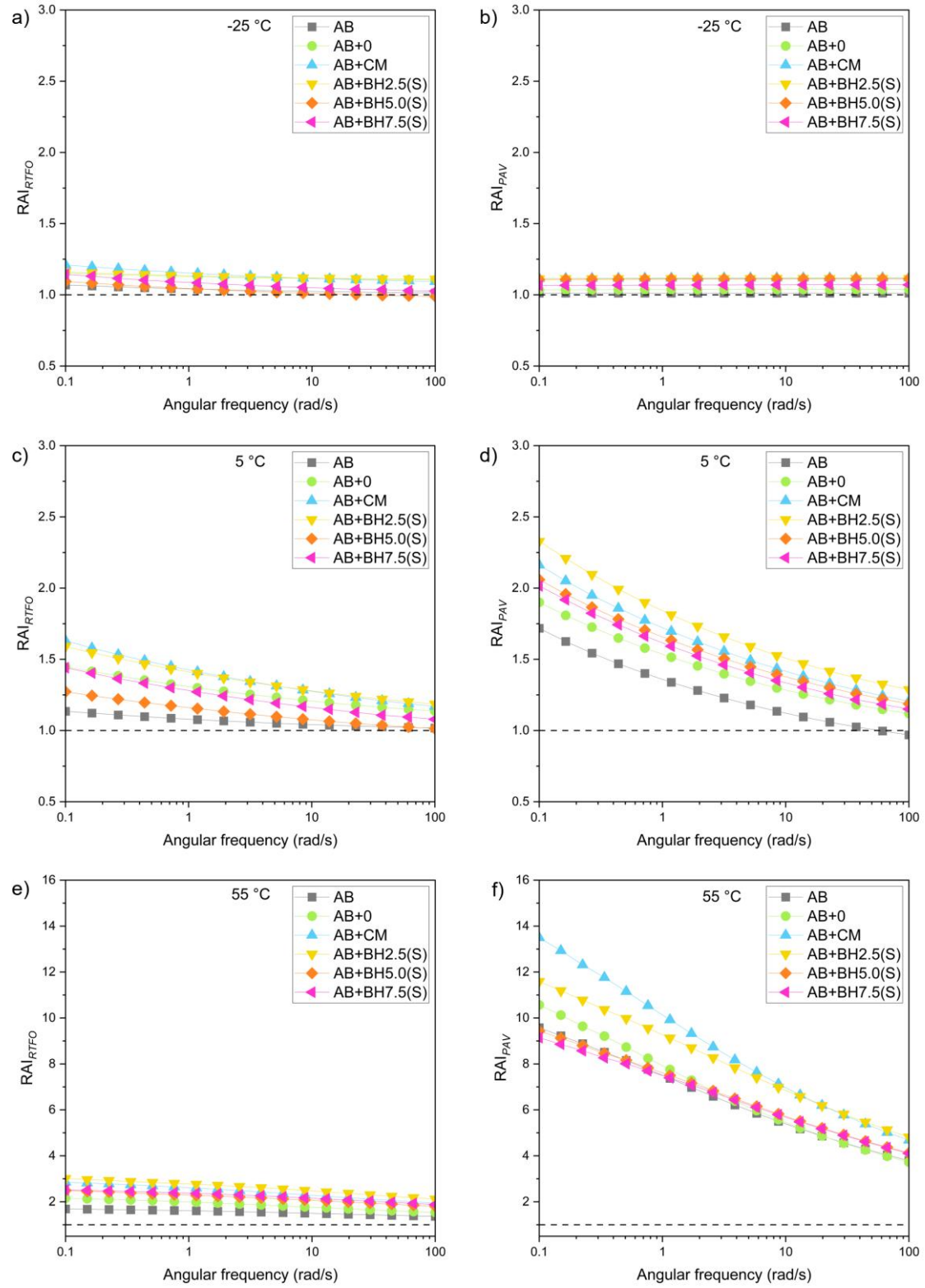


Figure 11. Rheological ageing index (RAI) of binders after RTFO and PAV ageing at: (a), (b) -25 °C, (c), (d) 5 °C and (e), (f) 55 °C.

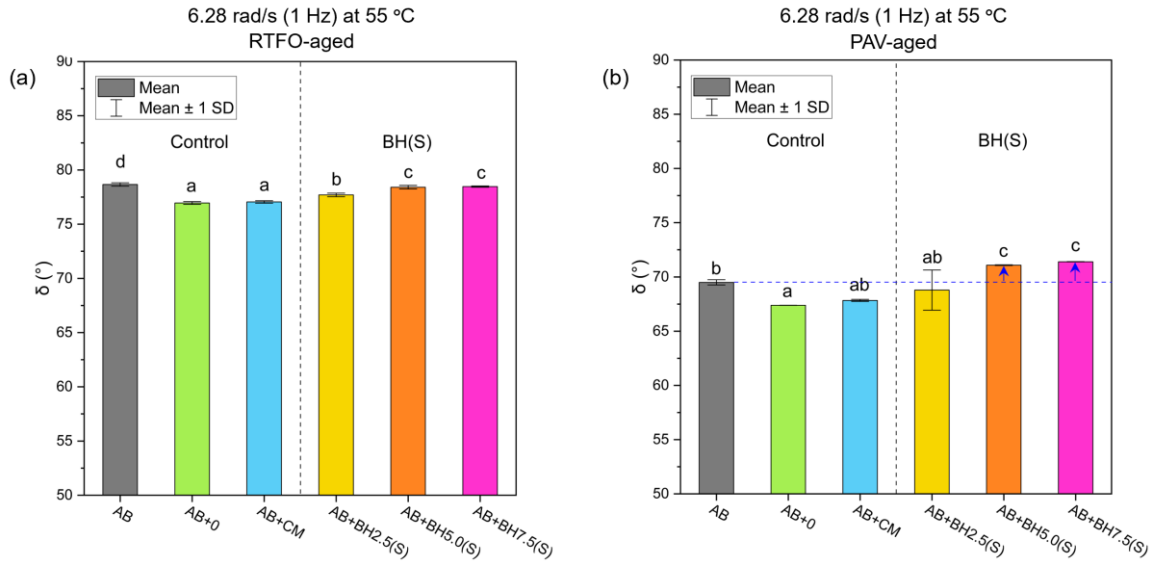


Figure 12. δ values at an angular frequency of 6.28 rad/s (1 Hz) and 55 °C for (a) RTFO-aged and (b) PAV-aged binders. Statistical groupings from Tukey's post hoc test are included. In panel (b), the blue dashed line represents the mean δ value of AB, while the blue arrows highlight the higher δ values of AB+BH5.0(S) and AB+BH7.5(S) relative to AB.

To evaluate fatigue resistance, a strain sweep test was conducted to delimit the linear viscoelastic (LVE) and non-linear viscoelastic (NLVE) regimes of the evaluated binders. The criterion adopted to identify the LVE limit corresponded to the strain associated with a 10 % reduction to the initial complex modulus ($|G^*|_0$), following the criterion established by the Strategic Highway Research Program (SHRP) [48], [53]. As shown in Figure 13(a), the modified binders exhibited lower LVE limits than AB, with values between 2 and 3 % strain. Based on these results, a strain amplitude of 8 % was selected for the fatigue tests to ensure NLVE conditions for all binders under the same loading condition. The comparative fatigue performance should therefore be interpreted within the context of the selected strain amplitude.

Fatigue damage evolution was evaluated through a time sweep test under controlled-strain conditions (8 %), monitoring the evolution of $|G^*|$ as a function of the number of loading cycles. The initial complex modulus ($|G^*|_0$) was defined as the value recorded at cycle 100, following a criterion adopted to exclude the effects of thixotropy and transient non-linearity associated with the initial loading cycles [54]. The time sweep results (Figure 13(b)) show that AB and AB+0 exhibited lower $|G^*|$ values than those obtained for the binders modified with CM and BH throughout the entire test. This indicates that the reinforcing effect associated with the modifier particles is also manifested under cyclic loading conditions at 25 °C, where the presence of the particulate phase contributes to increasing binder stiffness. Analysis of damage evolution showed that the binders containing BH tended to reach a 50 % reduction in $|G^*|_0$ after a greater number of cycles than the control binders. Specifically, AB+BH2.5(S), AB+BH5.0(S) and AB+BH7.5(S) reached this threshold at cycles 933, 1019, and 999, respectively, under the selected testing conditions. This trend is consistent with the rheological response observed after PAV ageing, particularly the higher δ values at intermediate frequencies reported in the master curve analysis (Figure 10(f)). These findings are also consistent with previous studies by the authors, in which lower oxidation indices and reduced ageing susceptibility were reported for BH-modified binders [27], [28].

In terms of accumulated dissipated energy (Figure 13(c)), the binders modified with BH exhibited higher average values than the control binders AB, AB+0 and AB+CM. In particular, AB+BH5.0(S) recorded the highest accumulated dissipated energy, with statistically significant differences confirmed by ANOVA (p -value < 0.001 ; **Table S.6**) and Tukey's test. These results indicate that, under the selected testing conditions, AB+BH5.0(S) exhibited the highest accumulated dissipated energy while also reaching the greatest number of loading cycles before a 50 % reduction in the initial complex modulus ($|G^*|_0$).

The combined analysis of the damage trajectories within the $|G^*|$ - δ space (Figure 13(d)) supports this interpretation, showing that the BH-modified binders evolved towards states with higher δ values while maintaining higher $|G^*|$ values than the control binders. This behaviour suggests that BH increased binder stiffness whilst maintaining a greater viscous contribution during fatigue damage evolution under the selected testing conditions.

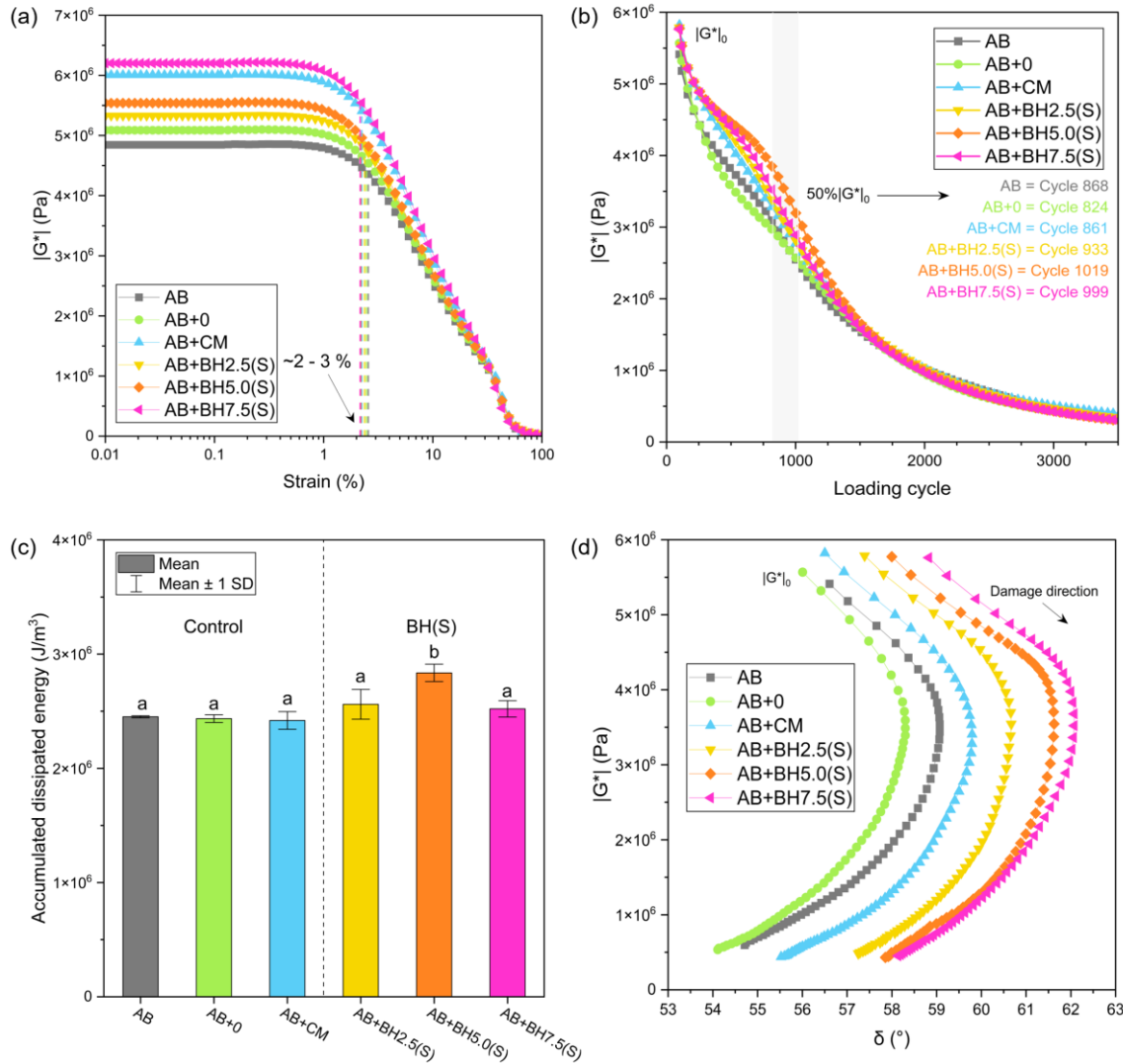


Figure 13. Fatigue resistance evaluation: (a) amplitude sweep test and LVE–NLVE limits, (b) $|G^*|$ evolution with loading cycles, (c) accumulated dissipated energy with Tukey statistical grouping, and (d) damage trajectory in the $|G^*|$ – δ space.

Overall, the results of the viscoelastic evaluation of the binders prepared with the selected BH particle size ($S < 20 \mu\text{m}$) suggest that the incorporation of 5.0 wt.% BH can improve several rheological properties of asphalt binders under the evaluated conditions. In the MSCR test, all BH contents resulted in significantly lower J_{nr} values than the control binders, with no significant differences among the BH-modified formulations. Furthermore, the master curve and RAI analyses showed lower susceptibility to ageing-induced stiffening in the binders containing 5.0 and 7.5 wt.% BH than in that containing 2.5 wt.%. Finally, during the fatigue test, AB+BH5.0(S) tended to reach a 50 % reduction in $|G^*|$ after a greater number of loading cycles and achieved the highest accumulated dissipated energy, with statistically significant differences for the latter parameter. Therefore, considering the response to permanent deformation, ageing, and fatigue together, 5.0 wt.% BH(S) was selected as the final formulation for this type of modifier.

4. Conclusions

In this study, the influence of the particle size and content of biochar derived from European hazelnut shell (BH), previously characterised by its antioxidant activity and phenolic composition, on the operational and rheological properties of a PG 64–22 asphalt binder was evaluated. Overall, smaller particle sizes and higher BH contents promoted greater improvements in the rheological performance of the modified binders, particularly at high service temperatures. Based on the experimental results obtained, the following conclusions can be drawn:

- The incorporation of BH increased the rotational viscosity of the binder and moderately raised the mixing and compaction temperatures, with this effect being more pronounced at higher modifier contents and smaller particle sizes ($< 20 \mu\text{m}$). Furthermore, all modified binders maintained good storage stability.
- At high temperatures, the incorporation of BH increased the $G^*/\sin(\delta)$ parameter and reduced the J_{nr} parameter, indicating improved resistance to permanent deformation, particularly for particle sizes smaller than $20 \mu\text{m}$. At the evaluated low temperature, BH increased creep stiffness without significantly affecting the stress relaxation capability compared with the control binders, and all binders satisfied the Superpave BBR criteria.
- Master curve analysis showed that BH modification influenced the viscoelastic response of the binder depending on the ageing condition. After PAV ageing, binders containing 5 and 7.5 wt.% BH retained a greater viscous contribution and exhibited ageing-induced changes in $|G^*|$ comparable to those of AB, despite the reinforcing contribution of the BH particles. This behaviour is consistent with the reduced oxidation and ageing susceptibility previously reported for BH-modified binders.
- Under cyclic fatigue loading conditions, the binders modified with BH tended to reach a 50 % reduction in the initial $|G^*|$ after a greater number of loading cycles than the control binders. In addition, the BH-modified binders exhibited higher accumulated dissipated energy and maintained higher $|G^*|$ values throughout the time sweep test under the selected testing conditions.
- Based on the combined response to permanent deformation, ageing, and fatigue, AB+BH5.0(S) was selected as the final formulation for this type of modifier, corresponding to 5.0 wt.% BH with a particle size below $20 \mu\text{m}$.

Overall, the results demonstrate the potential of BH as a sustainable modifier for the evaluated PG 64–22 asphalt binder, capable of improving its rheological performance whilst promoting the valorisation of agro-industrial waste within a circular economy framework. Furthermore, the performance achieved by BH, particularly at smaller particle sizes, was comparable to and, in some cases, better than that obtained with commercial graphite powder (CM), especially under long-term ageing conditions. Nevertheless, further studies are required to validate its performance at asphalt mixture scale, assess its long-term durability under field conditions, and investigate its applicability to asphalt binders from different crude oil sources and performance grades.

5. Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work, the authors used ChatGPT in order to enhance the readability and grammar of the text. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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Highlights

1. Biochar from hazelnut shells (BH) was used as a sustainable asphalt modifier.
2. BH delayed long-term ageing-induced stiffening, possibly by its antioxidant properties.
3. BH particle size and content influenced binder performance improvements.
4. BH enhanced rutting and fatigue resistance, preserving low-temperature performance.
5. BH enabled agro-industrial waste valorisation within a circular economy.

Declaration of interests

☐ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☒ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Gonzalo Valdes Vidal reports financial support was provided by National Agency for Research and Development (ANID). If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.