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Research Article

Effects of Waste Cooking Oil on the Antiageing Ability of Bitumen

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Ageing is considered one of the significant issues faced by bituminous mixtures. The short-term ageing phase involves a significant rise in the bitumen's viscosity, which may lead to early raveling and cracking. Antiageing additives are prescribed to reduce the effects of short-term ageing. However, most antiageing additives have detrimental health effects and also affect water quality. In recent times, waste cooking oil (WCO) has gained attention as a potential antiageing additive considering its peptizing ability. In this study, the antiageing ability of WCO is investigated considering the rheological and chemical parameters of the bitumen (binder). The rheological test included oscillation, frequency sweep, and multiple stress creep recovery (MSCR). The chemical test included the extraction of asphaltene from the bitumen. PG 64-10 and VG30 binders were short-term aged and modified with 3.0 and 5.0% WCO, respectively. The master curves indicated the presence of optimum WCO content for the binders, where the WCO short-term aged modified binders overlapped with that of unaged binders. The Burger model fitted for the creep phase of the bitumen indicated a significant increase in the viscous strain when the WCO addition exceeded the optimum value. The ageing indices based on rheological and chemical parameters depicted an excellent correlation. The optimum values of WCO based on rheological, chemical, and ageing indices were found to be in tandem. Overall, WCO has the potential to function as an antiageing additive, and the optimum value should be identified meticulously, as adding beyond the optimum may lead to permanent deformation.

1. Introduction

Bitumen is a commonly used binding material in road pavements worldwide [1]. It binds with the mineral aggregates to form the bituminous mixtures, which provides a water-proof coat for the lower layers. During the mixing, construction, and operation of the bituminous pavement, its physical properties change significantly throughout its life. These changes primarily occur due to environmental conditions like temperature, rainfall, UV radiation, etc. This is defined as the ageing of the bituminous pavement [2]. Ageing causes changes in the physical, rheological, and chemical properties of bitumen due to variations in its chemical composition during the construction process and throughout its service life [3, 4]. Figure 1 shows the typical variation in the viscosity of the bitumen in the mixture. It can be seen that the rate of change of viscosity during the short-term ageing is significantly higher compared to long-term ageing as a thin film of bitumen is exposed to high temperature for at least 2–4 hours [5].

The drastic change in the viscosity due to short-term ageing results in distresses. The common distresses include thermal cracking, raveling, moisture damage, and early fatigue cracking in bituminous pavement [6–8].

As ageing is an inherent characteristic of bitumen, owing to the presence of volatile components, several additives have been proposed as antiageing additives, which can reduce the extent of ageing [9]. As short-term ageing mainly involves the loss of volatile components, it is reversible, unlike oxidative ageing, which is irreversible. Some of the most commonly investigated antiageing additives include dilauryl thiodipropionate, furfural [10], imidazoline [11], hydrated lime [12], and diatomite [13]. These additives result in an antiageing effect by either physically and/or chemically altering the bitumen. In recent times, oil-based antiageing additives have gained attention, and one of them is waste cooking oil (WCO) [14].

Past studies have found that 1.0 to 5.0% of WCO is needed to restore the rheological and physical properties of

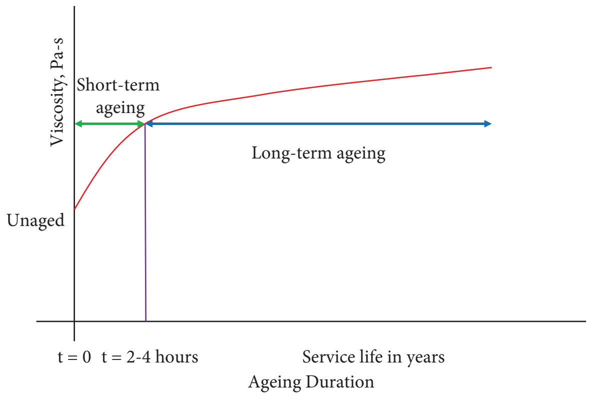


Figure 1: Typical variation in viscosity of bitumen in a bituminous mixture [5].

the aged binder as compared to the unaged binder [15–19]. Cao et al. [20] showed that WCO could be used effectively as a rejuvenator for utilizing high reclaimed asphalt pavement (RAP) content. Also, the study showed that the linear viscoelastic range of aged bitumen could be increased using WCO. It was found that rejuvenated bitumen with 5% WCO had the same fatigue life as the virgin binder. As the amount of WCO increased, the fatigue life of rejuvenated bitumen increased exponentially. The same results were observed in other studies [19, 21, 22]. These studies show that at a particular dose of WCO, the rheological and physical properties of bitumen can be revived in the unaged binder. Lyu et al. [23] concluded that WCO with crumb rubber could be utilized to enhance the ageing resistance of rubberized bitumen. Azahar et al. [24] observed that the ageing rate of bitumen might be reduced by adding WCO in the bitumen. However, the optimum amount of WCO was not determined in this study. Zhang et al. [25] have utilized bio-bitumen to improve the high-temperature performance and antiageing properties of bio-bitumen. The bio-bitumen was produced using bio-oil with up to 30% content in the synthesis process. The study observed that bitumen binder with bio-oil may considerably improve the high temperature and antiageing performance of bitumen using distilled water. Zeng et al. [26] conducted a study on the bio-bitumen produced using castor oil. The findings showed that bio-bitumen had adequate ageing resistance. Gökalp and Uz [14] have utilized WCO as an antiageing agent; the study concluded that almost all of the negative effects taking place during short-term ageing could be fixed by adding six percent WCO to the binder. However, the negative effect of long-term ageing can be avoided by adding seven percent WCO to the binder. Yan et al. [19] concluded in their study that bitumen with 8% WCO showed the same rheological and physical properties after long-term ageing as compared to the unaged binder.

Although many studies have been conducted on using WCO in binder as a rejuvenator, there are very few studies investigating the potential of WCO as an antiageing agent in bitumen. Also, minimal research has been found investigating the ageing indices (AI) in terms of chemical and rheological properties with WCO. Higher maltenes of WCO increases the maltenes proportion in modified binder, resulting in lower asphaltene-to-maltene ratio after short-term ageing as compared to unmodified binder. Therefore, the WCO can be utilized as an antiageing agent, which was investigated in this study contributing to the state-of-the-art.

The main objective of the study is to investigate the antiageing potential of WCO in bitumen against the short-term ageing. In this direction, the scope of the study included the following:

(i) Selection of different binders
(ii) Ageing of modified binder with waste cooking oil (WCO)
(iii) Physical tests include penetration and softening points
(iv) Rheological investigations using a dynamic shear rheometer (DSR)
(v) Determination of asphaltene content
(vi) Assessing the effect of WCO on bitumen stripping
(vii) Formulation of ageing indices

The research outline of the study is shown in Figure 2.

2. Research Significance

Ageing is considered one of the inherent causes of distresses in the bitumen. From the stage of mixture production up until the end of the design life, the thin coating of the bitumen is subjected to conditions that increase its stiffness. The increased stiffness of the bitumen will increase its tendency to undergo crack distress, decreasing the serviceable life of the bituminous pavements. With the present research works focusing on perpetual/long-lasting bituminous pavements, ageing plays a significant role as the anticipated design life of these pavements may get affected due to ageing [27]. In this regard, antiageing and antioxidants are gaining importance to reduce the ageing in bitumen. However, some commonly used antioxidants, such as furfural, lead diamyldithiocarbamate, and dilauryl thiodipropionate, are known to cause dermal and respiratory irritations and are also known to affect water quality [28–30].

On the other hand, WCO is abundantly available (around 50 million tons globally [31]) and disposed of without proper treatment. It is gaining attention as a rejuvenator in bituminous mixtures with high recycled asphalt content. Meanwhile, the antiageing effect of WCO in bituminous mixture is not significantly explored and needs immediate attention to explore its antiageing properties [14]. This study adds to the state-of-the-art, investigating the antiageing properties of WCO in bitumen by conducting rheological and chemical analyses.

3. Materials and Methods

3.1. Materials

3.1.1. Bitumen Binder. In this study, two binder grades were used: performance graded PG 64-10 and viscosity graded VG30. PG 64-10 binder was a polymer-modified binder,

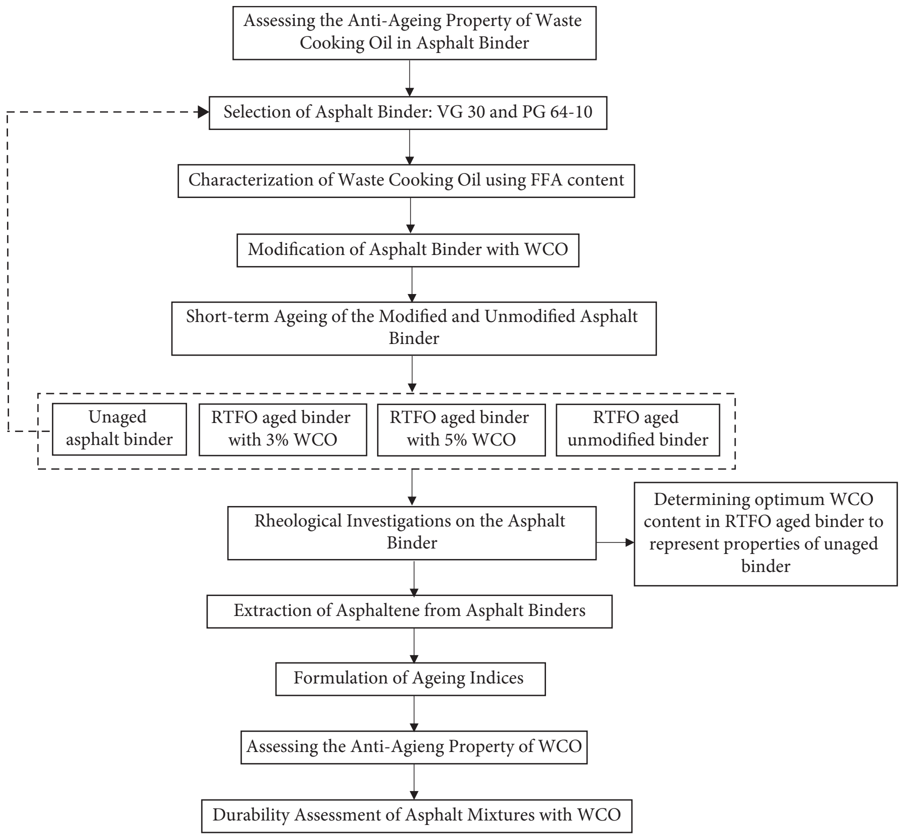


Figure 2: Research outline.

while VG30 was an unmodified binder. These binders were compared in this study because both the binders showed similar higher temperature performance of 64°C. The properties of the binders are shown in Results.

3.1.2. Waste Cooking Oil (WCO). WCO was collected from a public hostel mess. The WCO was then filtered with a filter paper having a diameter of 125 microns to extract the impurities. The quality of WCO was assessed by determining its free fatty acid (FFA) content as per ASTM D5555 [32] by a titration method. The FFA of WCO was found to be 1.39%.

3.2. Methods

3.2.1. Ageing of the Binder and WCO Addition. The control binders were first mixed with 3.0 and 5.0% (by weight) of WCO using a mechanical stirrer at a constant speed of 1000 rpm for 1 hour at 160°C [33]. Then, the unmodified and modified binders were short-term aged as per ASTM D2872 [34] to assess the antiageing properties of different binders. The short-term ageing of modified binders (short-term aged modified binder) is shown as the "aged binder with % of WCO" throughout the study.

3.2.2. Rheological Test. The rheological tests were conducted using a dynamic shear rheometer (DSR) with a parallel plate configuration. The oscillation test was conducted to determine the complex shear modulus at three temperatures: 25, 35, 45, 55, and 64°C and a frequency of 1.59 Hz. The specimen size used was 25 mm in diameter and 1.0 mm thick.

The frequency sweep test was conducted at a strain rate of 0.1%, with frequencies ranging from 20 to 0.1 Hz. The temperature for frequency sweep was 25, 35, 45, 55, and 64°C. The specimen size for temperatures above 45°C was 25 mm in diameter and 1.0 mm thick. On the other hand, for temperatures below 45°C, the specimen size was 8.0 mm in diameter and 2.0 mm thick. These sample sizes of binder were selected as per ASTM D7175 [35].

The multiple stress creep recovery (MSCR) test was conducted to investigate the creep-recovery characteristics of the binder. The test was conducted at a temperature of 64°C, representing the high pavement surface temperature. The stress levels, specimen dimension, and number of creep-recovery cycles were selected according to ASTM D7405 [36].

3.2.3. Asphaltene Extraction. The asphaltene extraction was conducted as per ASTM D6560 [37] with *n*-heptane as the solvent. In this procedure, bitumen at a rate of 1.0 g in 30 ml of *n*-heptane was mixed. The weight of the sample and the volume of the *n*-heptane may vary depending on the expected asphaltene content of the binder. The mixture is heated under reflux for about an hour. This leads to the precipitation of asphaltenes, waxy substances, and inorganic materials, which are then collected on a filter paper. The filtrate is then treated with hot heptane in an extractor to remove the waxy substances. Once the waxy substances are removed, hot toluene is used to dissolve asphaltene, as a result of which inorganic materials are separated. The extraction solvent is then evaporated, and asphaltenes are weighed. Asphaltene content is finally reported as the percentage by weight of asphaltenes extracted to the total weight of the bitumen sample tested.

3.2.4. Durability Test. The durability test was performed as per ASTM D3625 [38]. The test was conducted to assess the stripping in the bitumen-coated aggregates. In this test, 200 grams of aggregates that passed through a 20 mm sieve and were retained on a 12.5 mm sieve were mixed with a 5% binder at a temperature of 160°C. After a uniform coating, the mixture was allowed to cool at room temperature for about two hours. Distilled water was heated to boiling, and the mixture at room temperature was added to the boiling water. The mixture was left undisturbed in the boiling water for 10 minutes. The mixture was removed from the boiling water and allowed to cool to room temperature. The degree of the coating was established visually, which was classified as well, moderate and poor, as shown in Figure 3.

3.2.5. Formulation Ageing Index. Ageing Index (AI) is a parameter that quantifies ageing by comparing numerical magnitudes associated with any of the bitumen characteristics for two successive ageing conditions, namely viscosity, rutting resistance, asphaltene/maltene ratio, etc. Asphaltene content, which is obtained from the asphaltene extraction process as explained in Section 3.2.3, can be used to calculate the maltene content of the corresponding sample by subtracting the asphaltene content from 100 as maltenes collectively represent aromatics, resins, and saturates. The ratio of the asphaltene-to-maltenes content could form a parameter A/M ratio, which could be used to evaluate the ageing indices of the bitumen samples. In this study, two ageing indices were formulated as shown in equations (1) and (2). Equation (1) evaluates the ageing in terms of the asphaltene-to-maltenes ratio, which considers the chemical nature of the bitumen. On the other hand, equation (2) evaluates the ageing in terms of the SHRP rutting parameter, where the rheological property is considered

$$AI_{A/MRatio} = \frac{A/M\ \textbf{ratio of the aged binder}}{A/M\ \textbf{ration of unaged binder}}, \quad (1)$$

$$AI_{G^*/\sin(\Delta)} = \frac{G^*/\sin(\Delta)\ \textbf{of aged binder}}{G^*/\sin(\Delta)\ \textbf{of unaged binder}}. \quad (2)$$

4. Results and Analysis

4.1. Penetration and Softening Point. The penetration and softening point temperatures of all eight bitumens tested in this study are shown in Table 1. It can be observed that the short-term aged modified binders showed a higher penetration value and a lower softening point as compared to the short-term aged unmodified binders. The rate of change of penetration value or softening point temperature due to the addition of WCO did not show any particular trend, which necessitates rheological investigations.

4.2. Effect of WCO Addition on Complex Shear Modulus. The rutting parameter ($G^*/\sin(\delta)$) was determined for the binders at 64°C and is shown in Figure 4. It can be seen that the unaged binders had a rutting parameter value of more than 1.0 kPa at 64°C, and in short-term aged conditions, a value of more than 2.2 kPa was observed. VG30 binder depicted a relatively greater change in the rutting parameter between unaged and aged conditions compared to PG64-10. This can be attributed mainly to the presence of polymers in the PG64-10, which might have reduced the tendency to age.

The WCO addition had a significant effect on both unaged and aged binders. In an unaged condition, VG30 almost depicted a 50% drop in the rutting parameter when 5.0% WCO was added. On the other hand, a 60% drop in the rutting parameter was observed in PG 64-10 with a 5.0% WCO for unaged binder. This indicates that the addition of WCO to the unaged binder will have detrimental effects on the permanent deformation characteristics of the bitumen. In the short-term aged condition of modified binders, adding 3.0% and 5.0% WCO reduced the rutting parameter. However, in each case, the aged binder showed a rutting parameter higher than 1.0 kPa. The main objective of adding WCO in unaged binders was to maintain the unaged binder's characteristics during mixing, transportation, and construction, as these activities result in significant ageing due to the loss of volatile components.

The variation of the rutting parameter as a function of frequency at 64°C is shown in Figure 5 for three binder types. It can be seen that for both the binders (PG 64-10 and VG30), short-term aged modified binders tend to approach unaged binders' characteristics when WCO is added. This could be attributed to the fact that the addition of WCO restores the asphaltene-to-maltene ratio of the binders by increasing the maltene content of the bitumen.

Master curves were fitted using the Christensen-Anderson-Marasteanu (CAM) model as shown in equation (3) and using the frequency sweep test data. The shift factors were determined using the William-Landel-Ferry (WLF) equation as shown in equation (4):

$$G^*(\omega) = G_g * \left[1 + \left(\frac{\omega_c}{\omega_r}\right)^K\right]^{-m/K}, \quad (3)$$

$$\log a(T) = \frac{-C_1 * (T - T_{ref})}{C_2 + T - T_{ref}}, \quad (4)$$

where $G^*(\omega)$ is the complex shear modulus at that frequency, Pa. G_g is the glass modulus, Pa. ω_r is the reduced frequency at the reference temperature, rad/s. ω_c is the crossover frequency at the reference temperature, rad/s. m, and K are the shape parameters, which collectively form the

Well Coated (a) | Moderately Coated (b) | Poorly Coated (c)

Figure 3: Degree of coating in aggregates. (a) Well coated, (b) moderately coated, and (c) poorly coated.

Table 1: Penetration and softening point temperature.

Binder	Penetration	Softening point (°C)
Unaged PG64-10	70	47.5
Aged PG64	35	56.5
Aged PG64 + 5%	80	44.0
Aged PG64 + 3%	47	52.0
Unaged VG30	60	49.0
Aged VG30	34	56.0
Aged VG30 + 5%	67	48.0
Aged VG30 + 3%	38	54.0

rheological index. T is the test temperature (°C). T_{ref} is the reference temperature set at 35°C. C_1 and C_2 are material constants.

The master curves for PG 64-10 and VG30 are shown in Figure 6. The shift factors and master curve fitting parameters for PG 64-10 and VG30 binder are shown in Table 2. It can be observed from Table 2 that the crossover frequency (ω_c) of the aged binder is lower than the unaged binder for both PG 64-10 and VG30. However, the addition of WCO to the aged binder increases the crossover frequency, which indicates that for a given temperature, the storage modulus will dominate the loss modulus at a higher frequency.

The master curve shows that as loading time increases, the $|G^*|$ magnitude of the binder decreases dramatically. It was observed that the modified samples of both VG30 and PG 64-10 showed lower $|G^*|$ values than their respective unaged binder samples. However, the short-term aged binder of VG30 with 5% WCO showed results similar to that of the control binder, which indicates that antiageing is achieved. On the other hand, in Figure 6, it can be observed that the aged binder of PG64-10 modified with 3% WCO showed properties similar to those of the control binder (unaged PG64-10).

4.3. Effect of WCO Addition of Creep-Recovery Behavior. The MSCR test was conducted to assess the creep-recovery behavior of bitumen binders at 0.1 and 3.2 kPa stress levels at a temperature of 64°C. The percent recovery of the binders is shown in Table 3. It can be seen that the addition of WCO reduced the recovery in the bitumen to a certain extent. In the case of PG 64-10, up to 3.0% addition of WCO, no significant drop in the % recovery was observed. However, at a WCO addition of 5.0%, a significant drop was observed, masking the effect of the polymer in PG 64-10. This indicates that the addition of WCO beyond the optimum (3%) will result in recovery properties of the binder being compromised. A similar observation was also made in the study [39]. This can be mainly attributed to the decrease in the asphaltene-to-maltene ratio, where the increased content of saturates lubricates the polymer chains, reducing their ability to recover due to slippage between the chains. The VG30 binder depicted significantly less recovery, irrespective of the presence of WCO.

The creep of the bitumen subjected to a load of 0.1 kPa was modeled using Burger's model, which consists of the Maxwell and Kelvin–Voigt models in a series configuration. In this study, one Maxwell model and one Kelvin–Voigt model were used to represent the creep behavior of the bitumen using strain as the parameter. In the Maxwell model, the spring and dashpot are in a series configuration. While in the case of the Kelvin–Voigt model, the spring and dashpot are configured in parallel. The constitutive equation for the creep response of Burger's model is shown in equation (5) [40].

$$\varepsilon_{tc} = \frac{\sigma_0}{E_M} + \frac{\sigma_0 * t}{\eta_M} + \frac{\sigma_0}{E_k} * \left(1 - e^{\left(-E_k/\eta_k\right) * t}\right), \quad (5)$$

where ε_{tc} = strain in the creep phase. σ_0 = constant creep stress, kPa. E_M and E_k = moduli of spring elements in Maxwell and Kelvin–Voigt models, respectively, kPa. η_M

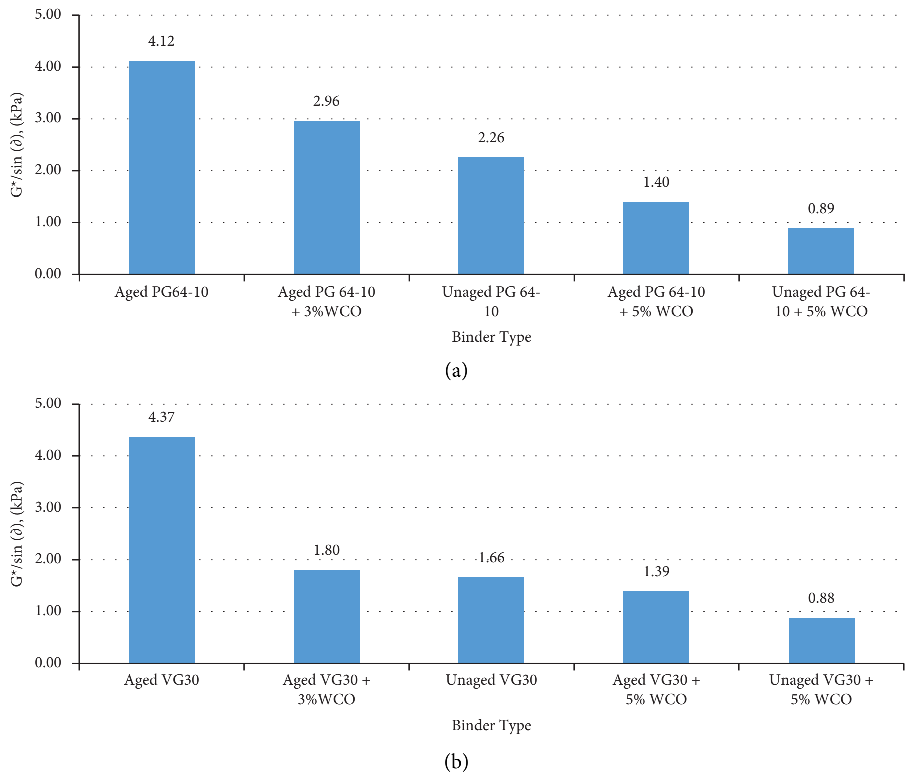


Figure 4: Effect of WCO addition on rutting parameters: (a) PG 64-10; (b) VG30.

and η_k = viscosity of dashpot elements in Maxwell and Kelvin–Voigt Model, respectively, kPa-s. t = creep time, seconds.

In equation (5), the first term on the right-hand side of the equation represents the instantaneous elastic strain (ε_e) due to the spring element in the Maxwell model. The second and third terms represent the viscous strain (ε_v) and viscoelastic strain or delayed elasticity (ε_d) due to the dashpot in the Maxwell model and Kelvin–Voigt model elements, respectively. The total strain is the sum of all the above three strains. The MSCR test data of PG 64-10 bitumen at 0.1 kPa creep stress was used to determine Burger's model parameters using equation (5). The fitted parameters are shown in Table 4. The fit was found to have an R^2 of at least 0.95 in all the cases. Using the parameters shown in Table 4, the total creep strain was divided into three components as discussed in the above section and is shown in Table 5. The elastic strain is constant for a given binder during the creep loading, while the viscous and viscoelastic strains increase continuously. The strain values shown in Table 5 were used to calculate the strain ratio, which is the ratio of viscous strain to the sum of elastic and delayed elastic strain. Figure 7 shows the trend in strain ratio for PG 64-10 binder.

It can be seen from Figure 8 that the unaged PG 64-10 depicted the lowest strain ratio during creep loading, indicating it possesses a very low viscosity. On the other hand, aged PG 64-10 with 5% WCO depicted the highest strain ratio, indicating higher viscous components compared to the elastic and viscoelastic components. Such binders will tend to undergo permanent deformation upon load application. Interestingly, aged PG 64-10 with 3% WCO depicted a similar strain ratio or a lower strain ratio to be specific than the aged PG 64-10 binder. This indicates that the addition of WCO imparts some amount of elasticity to the binder, which can be considered optimal. A similar observation was also made based on the master curves, as shown in Figure 6.

4.4. Effect of WCO on Asphaltene Content in Bitumen Binders. The asphaltene in the different bitumens was extracted as per ASTM D6560. Figure 9 shows the asphaltene content in percentage for the binders. It can be seen from Figure 9 that the aged binder possesses the highest percentage of asphaltene compared to other binders. In the case of aged PG 64-10, the asphaltene content increased by 7.4% compared to the unaged binder. However, in the case of aged VG 30, an increase of 30.15% was observed compared to the unaged state, indicating that VG30 is more age-susceptible. In support of this, there was a steep increase in $G^*/\sin(\delta)$ of aged VG 30 compared to unaged VG 30, as shown in Figure 4. When the WCO was added to the aged binder as an antiageing additive, the asphaltene content decreased, mainly due to the peptizing ability of the WCO, which dissociates the asphaltene molecules. The higher the WCO content added, the more reduction was seen in the asphaltene. In the case of VG 30 binder, a 3% addition of WCO resulted in an almost 25.0% decrease in the asphaltene, while a 5% addition of WCO depicted a 28.0% decrease in the asphaltene. It can be observed that the lower the asphaltene content in the aged condition, the lower WCO

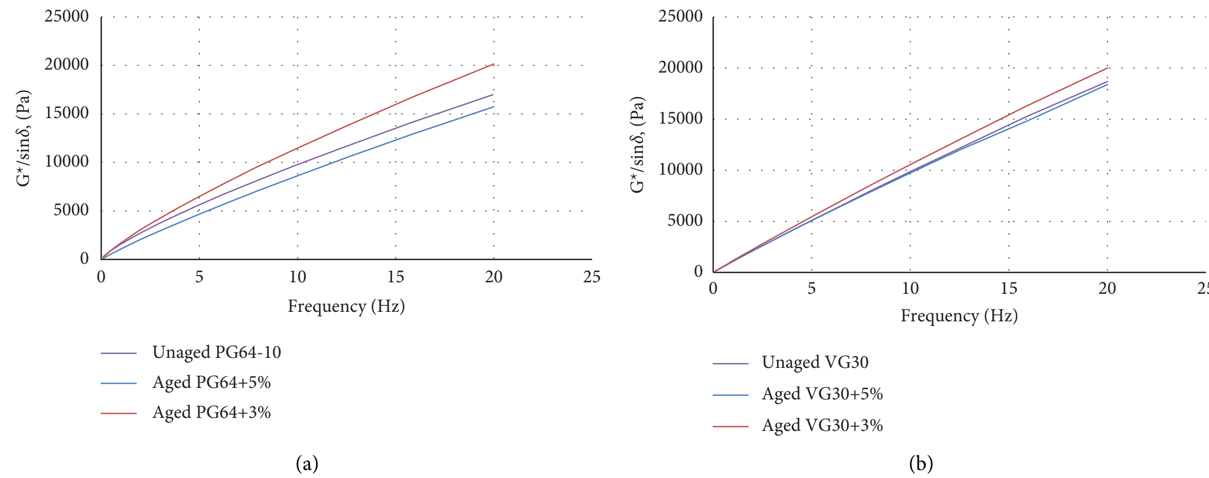


Figure 5: Variation of rutting parameters as a function of frequency: (a) PG 64-10; (b) VG30.

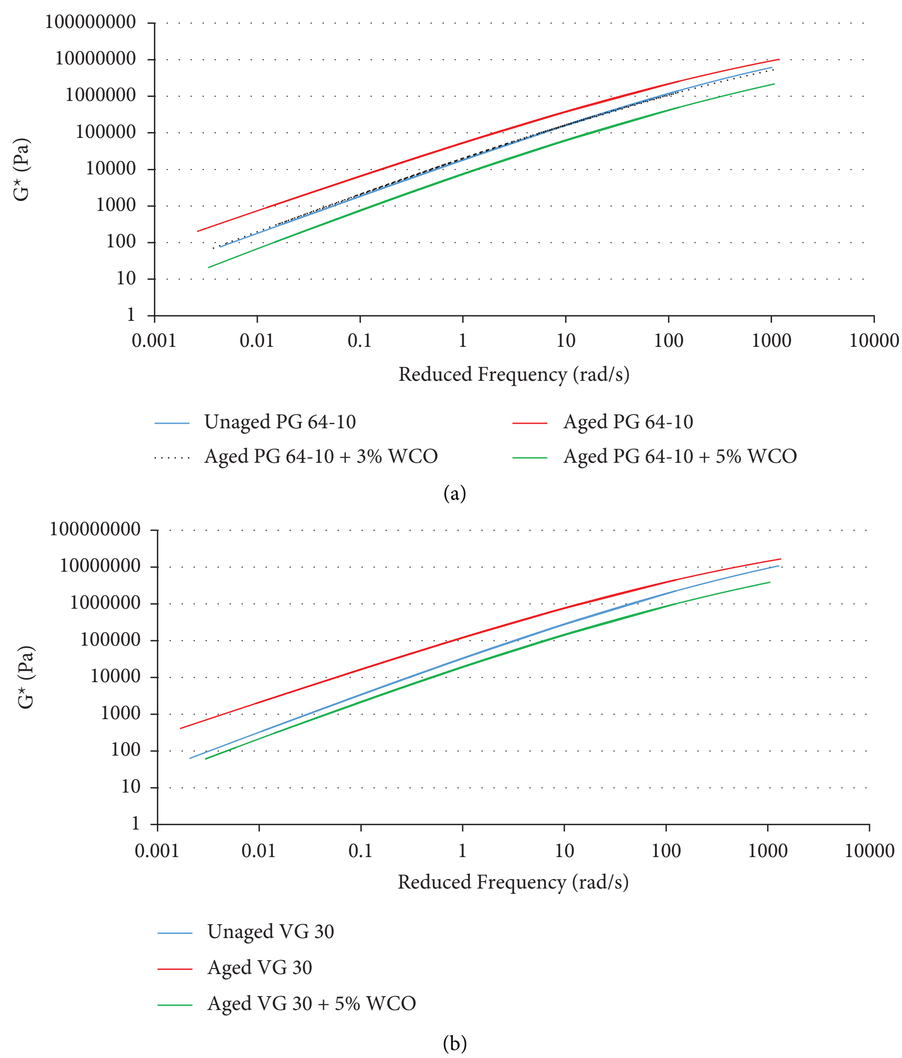


Figure 6: Master curve for bitumen binders. (a) PG 64-10; (b) VG30.

Table 2: WLF and CAM model parameters for master curves.

Binders	WLF equation parameter		CAM model parameters			
	C1	C2	ω_c (rad/s)	m	k	G_g (Pa)
Unaged PG 64-10	16.04	186.08	2857.24	1.032	0.366	$7.837E+07$
Aged PG 64-10	20.00	214.25	2347.08	0.992	0.276	$1.764E+08$
Aged PG 64-10 + 3% WCO	19.44	220.85	2405.38	1.089	0.251	$1.735E+08$
Aged PG 64-10 + 5% WCO	19.38	217.79	2453.09	1.153	0.208	$1.680E+08$
Unaged VG30	21.67	223.92	2760.38	1.066	0.283	$2.256E+08$
Aged VG30	24.99	252.25	1371.88	0.938	0.295	$1.508E+08$
Aged VG30 + 5% WCO	23.60	264.81	1424.86	1.100	0.223	$1.390E+08$

Table 3: %Recovery of the binders.

Binders	%Recovery	
	0.1 kPa	3.2 kPa
Unaged PG 64-10	52.82	32.17
Aged PG64-10	55.46	29.09
Aged PG 64-10 + 3%WCO	43.94	17.36
Aged PG 64-10 + 5% WCO	5.40	0.400
Unaged VG30	0.0088	0.003
Aged VG30	5.21	0.0089
Aged VG30 + 3% WCO	0.011	0.0010
Aged VG30 + 5% WCO	0.010	0.0014

Table 4: Burger's model parameters for PG 64-10.

Burger's model parameters	Unaged PG 64-10	Aged PG 64-10	Aged PG 64-10 + 3% WCO	Aged PG 64-10 + 5% WCO
E_M (kPa)	36.156	24.913	11.682	19.087
N_M (kPa-s)	0.510	0.678	0.429	0.141
E_K (kPa)	2.185	3.951	6.88 ∗ 10ˆ−7	3.344
N_K (kPa-s)	0.755	3.389	2.173	4.062

content will be required to initiate the antiageing effect or restore the aged binder's properties to the unaged binder. Any further addition of WCO will hamper the permanent deformation characteristics of the binder. This observation confirms that for PG 64-10 binder, the optimum WCO was found to be 3.0%, and in the case of VG 30 binder, it was 5.0%, as observed from the master curve and strain ratio curve in the case of PG 64-10. The higher asphaltene content in the aged binder will need a higher addition of WCO to cause sufficient peptizing action to cause antiageing effect.

The rheological properties (Viscosity and rutting parameter) were correlated with the asphaltene content in aged, aged modified binder with 3% WCO, and aged modified binder with 5% WCO binders, as shown in Figure 10. The correlation between the asphaltene content and rheological parameters was found to be excellent. It can be seen that the aged bitumen, which had the highest asphaltene content, had the highest viscosity and rutting parameters. However, when WCO was added, the content of asphaltene was reduced, owing to the peptizing power of WCO. The addition of WCO also decreases the asphaltene-to-maltene (A/M) ratio, which contributes to the reduction in viscosity and rutting parameters. Further, in both PG 64-10 and VG30 binders, the rutting parameter was not less than 1.0 kPa when WCO was added to the short-term aged binder. In the case of the VG 30 binder, it can be seen that there was a large drop in the asphaltene content when 3.0% WCO was added, unlike the PG 64-10 binder.

4.5. Effect of WCO on Ageing Indices. Ageing indices are indices that quantify the extent of ageing in the bitumen compared to unaged bitumen. The ageing indices can be expressed using rheological and basic properties of bitumen, such as penetration. In this study, ageing indices are defined using rheological and chemical parameters. The ageing indices for PG 64-10 and VG30 based on rheological and chemical parameters are shown in Figures 7 and 11, respectively.

The threshold line is shown in Figures 7 and 11, which represents the case of no ageing. It can be observed that for both the binders, the ageing index of the short-term aged binder was highest. Adding WCO reduced the ageing index and tended to approach a value of 1.0, representing the case of no ageing. In the case of VG30, almost at 5.0% WCO, the ageing indices based on chemical and rheological parameters both approached a value of 1.0, indicating antiageing effect. Furthermore, in the case of PG 64-10, the WCO content to

Table 5: Elastic, viscous, and viscoelastic strains for the PG 64-10 binder.

Binder	Time, seconds	0.10	0.20	0.30	0.40	0.50	0.60	0.70	0.80	0.90	1.00
Unaged PG 64-10	(ε_e), %	0.28	0.28	0.28	0.28	0.28	0.28	0.28	0.28	0.28	0.28
	(ε_v), %	1.96	3.92	5.88	7.84	9.80	11.76	13.73	15.69	17.65	19.61
	(ε_d), %	1.15	2.01	2.66	3.14	3.50	3.77	3.97	4.12	4.24	4.32
Aged PG 64-10	(ε_e), %	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40
	(ε_v), %	1.47	2.95	4.42	5.90	7.37	8.85	10.32	11.80	13.27	14.75
	(ε_d), %	0.28	0.53	0.75	0.94	1.12	1.27	1.41	1.54	1.64	1.74
Aged PG 64-10 + 3% WCO	(ε_e), %	0.86	0.86	0.86	0.86	0.86	0.86	0.86	0.86	0.86	0.86
	(ε_v), %	2.33	4.66	6.99	9.32	11.66	13.99	16.32	18.65	20.98	23.31
	(ε_d), %	0.46	0.92	1.38	1.84	2.29	2.75	3.21	3.67	4.12	4.58
Aged PG 64-10 + 5% WCO	(ε_e), %	0.52	0.52	0.52	0.52	0.52	0.52	0.52	0.52	0.52	0.52
	(ε_v), %	7.09	14.18	21.28	28.37	35.46	42.55	49.65	56.74	63.83	70.92
	(ε_d), %	0.22	0.43	0.61	0.79	0.95	1.08	1.22	1.34	1.45	1.54

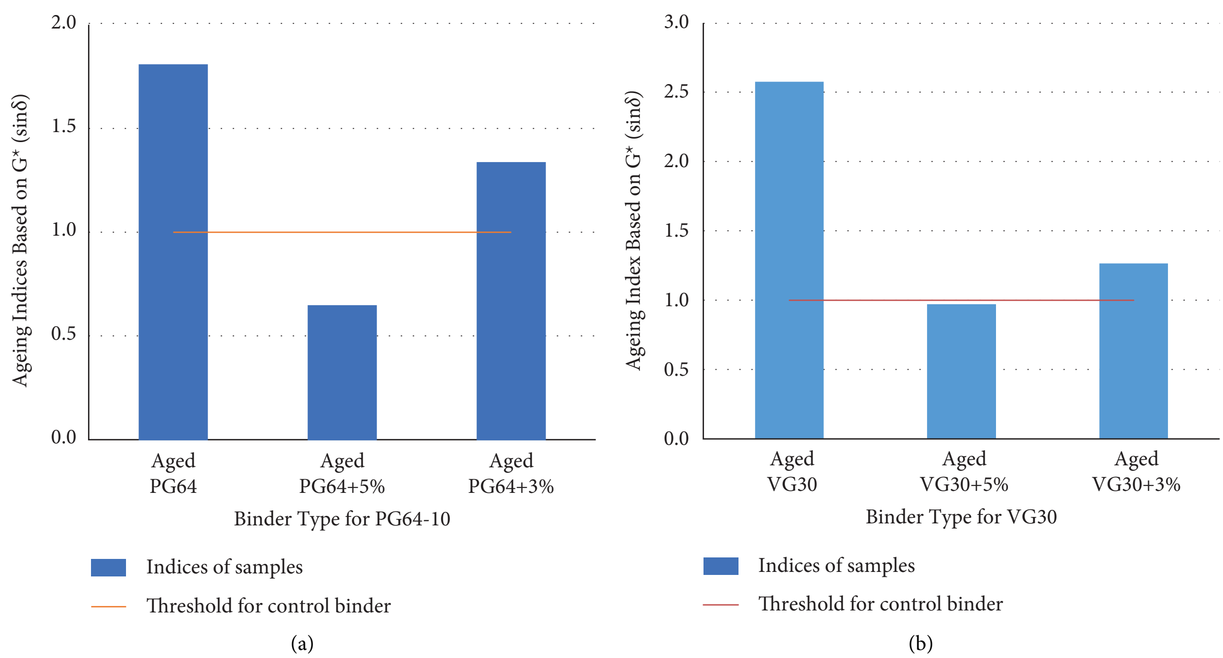


Figure 7: Ageing indices ratio for bitumen binders based on rheological parameter. (a) Binder type for PG64-10. (b) Binder type for VG30.

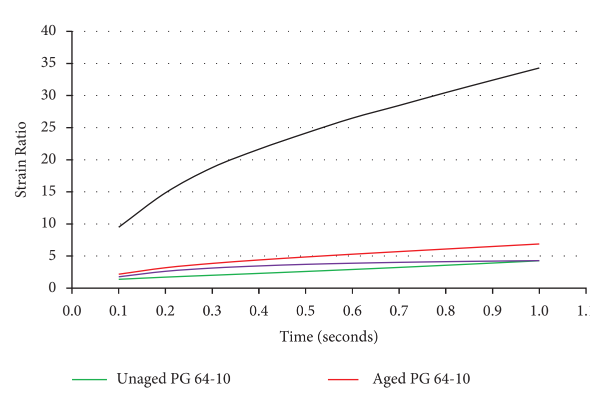


Figure 8: Strain ratio for PG 64-10 bitumen.

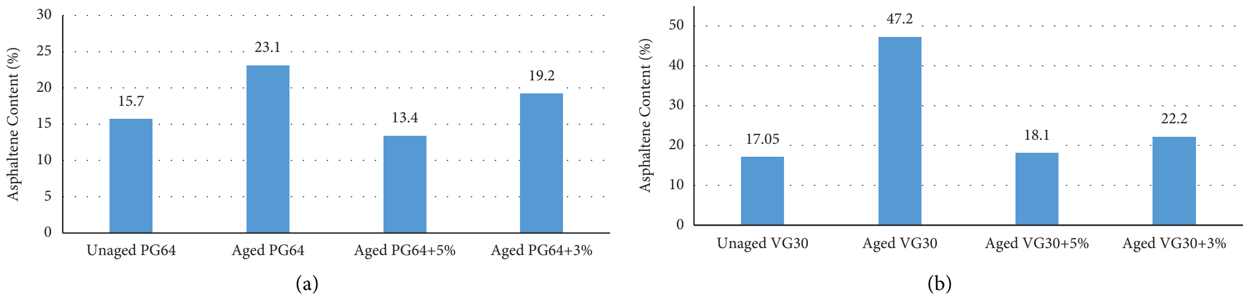


Figure 9: Variation in asphaltene content in different bitumens: (a) PG 64-10; (b) VG30.

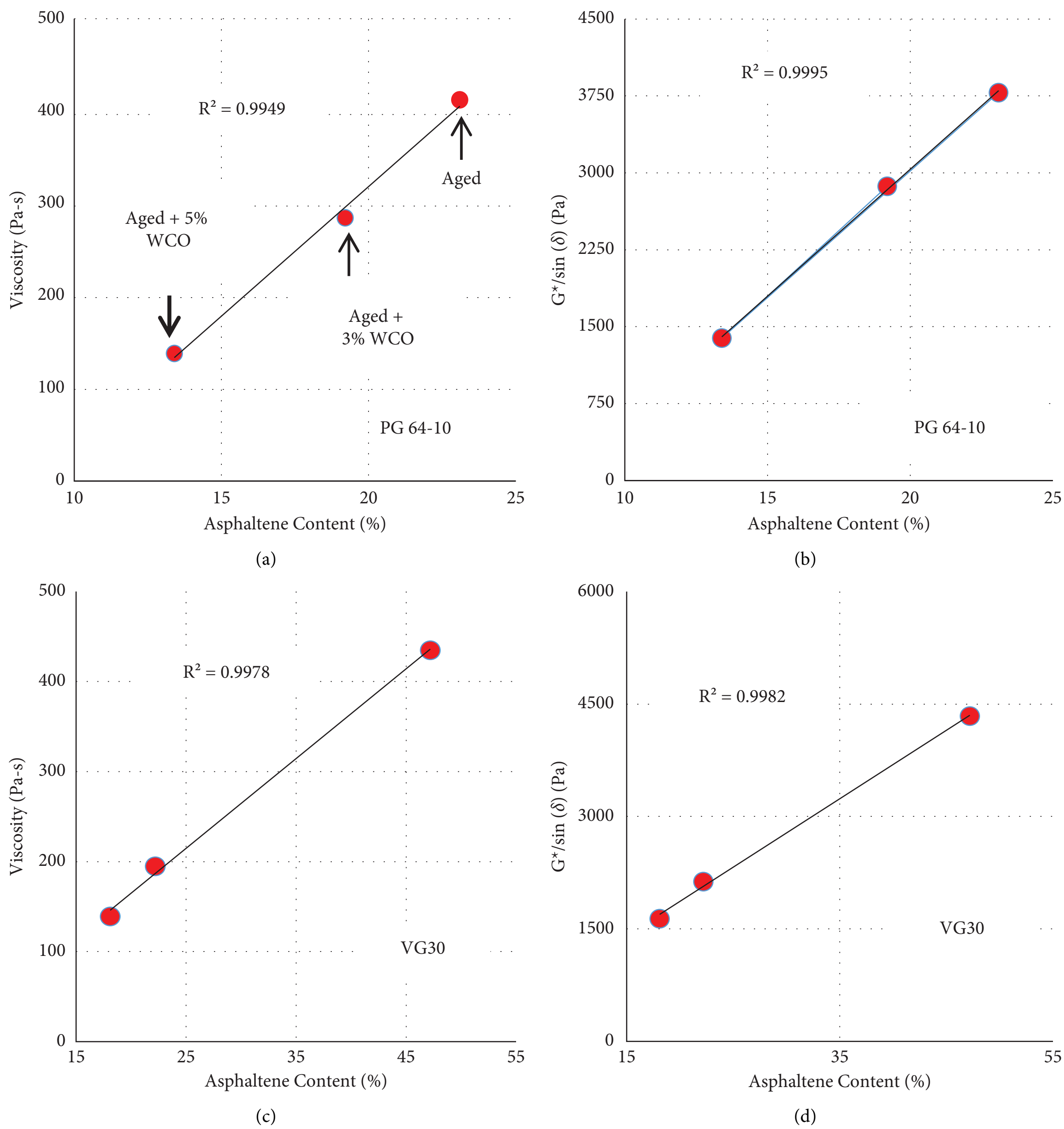


Figure 10: Relationship between asphaltene content and rheological properties determined at 64°C and 1.59 Hz frequency.

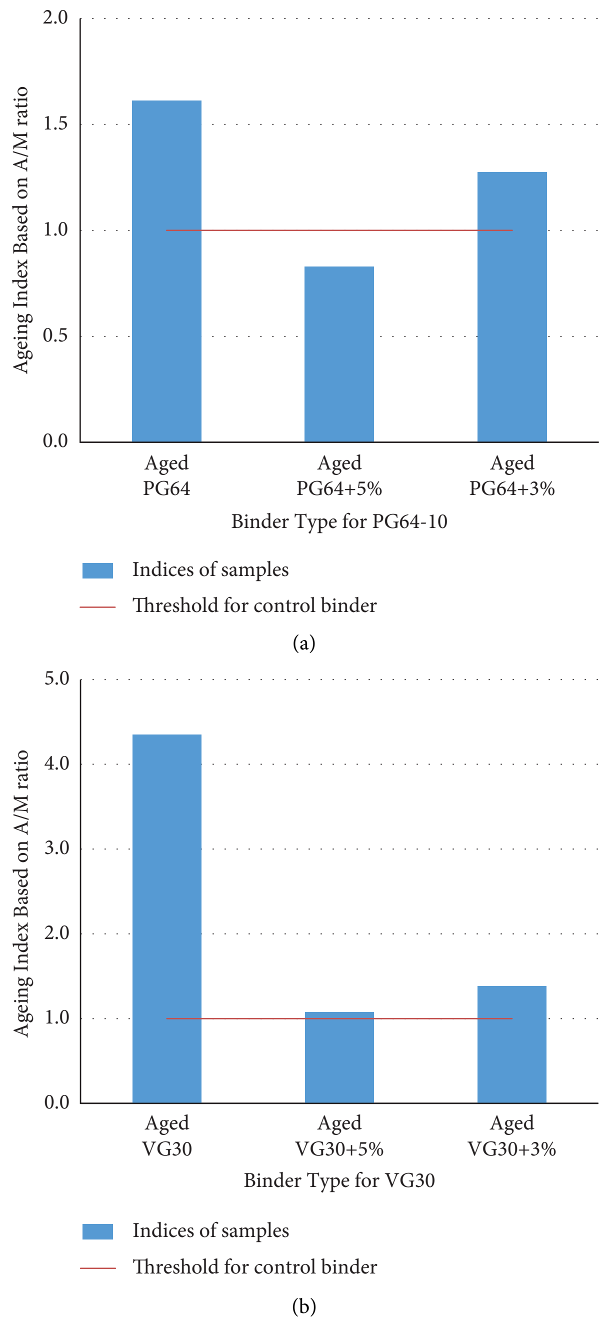
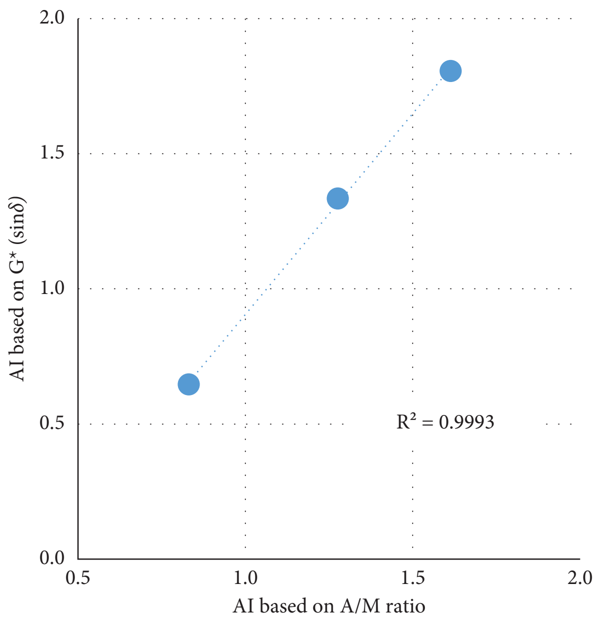


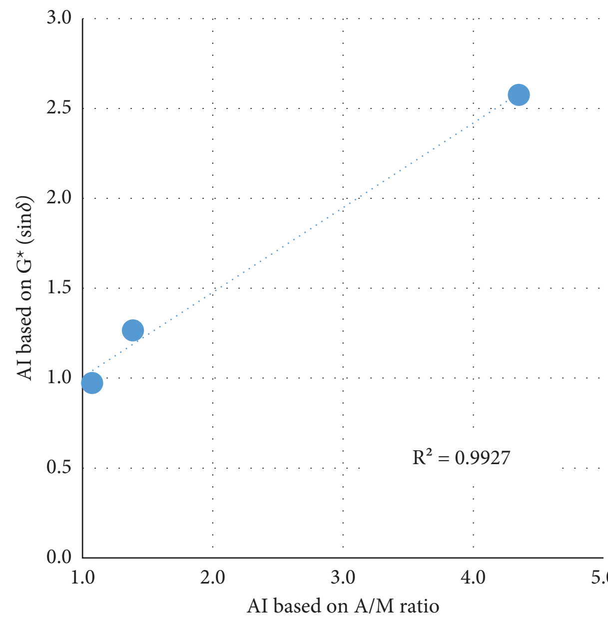
Figure 11: Ageing indices ratio for bitumen binders based on chemical parameters. (a) Binder type for PG64-10. (b) Binder type for VG30.

achieve an ageing index of 1.0 was between 3.0 and 5.0%. This observation clearly indicates that adding WCO during the mix production stage will reduce the ageing tendency of the binder and mitigates the short-term ageing of bitumen. The reduced short-term ageing will lead to significant benefits such as increased durability, endurance, and reduced cracking. The correlation between ageing indices based on the rheological and chemical parameters is shown in Figure 12. It can be clearly seen that the ageing indices based on the rheological and chemical parameters are excellently correlated, indicating the correctness of the methodology of testing.

4.6. Effect of WCO on Bitumen Stripping. The effect of the addition of WCO on the durability of the mixtures was investigated by conducting a bitumen stripping test. The total number of aggregate particles was counted for each binder and were classified as well, moderate, and poor based on the degree of coating by visual appearance. The percentage of aggregates under each category of coating is shown in Table 6. It can be observed that the unaged binder depicted the "well" degree of coating for the highest percentage of aggregates. However, after adding WCO, there was a slight increase in the aggregate particles in the moderate and poor coating categories compared to the unaged binder. Further, as the WCO content



(a)



(b)

Figure 12: Correlation between ageing indices based on rheological and chemical parameters.

Table 6: Percent of the total number of aggregate particles with different degrees of coating.

Coating	*% of coating on aggregates*					
	Unaged PG64-10	Aged PG64 + 3%	Aged PG64 + 5%	Unaged VG30	Aged VG30 + 3%	Aged VG30 + 5%
Well	96.67	93.33	91.67	98.67	96.33	95.67
Moderate	3.33	3.34	5.55	1.33	3.34	1.16
Poor	0.00	3.33	2.78	0.00	0.33	3.17

increases, there seems to be a probability of an increase in the stripping of bitumen. A similar observation was made in a recent study by Yan et al. [19], where it was found that WCO additions between 4.0 and 6.0% could reach moisture damage resistance similar to that of the unaged binder. These results indicate that WCO can be used as an antiageing additive up to a certain limit, which should be identified depending on the unaged binder type.

5. Conclusions

The main objective of this study was to investigate the antiageing effect of WCO, considering rheological and chemical aspects. Based on the laboratory studies, the following conclusions are made:

(i) The addition of WCO to the unaged binder resulted in an increase in penetration and a decrease in softening point. However, no particular rate/trend was observed related to change in penetration or softening point.

(ii) The master curve using the CAM model clearly depicted the presence of an optimum WCO content for a given binder, where one can expect an overlapping with the master curve of the unaged binder. For PG 64-10 and VG30 binders, the optimum WCO was found to be 3.0 and 5.0%, respectively.

(iii) The higher the extent of short-term ageing in bitumen, the higher will be the value of optimum WCO as more WCO is needed to peptize the higher asphaltene content.

(iv) Adding WCO improves the creep-recovery characteristics up to a certain percentage, which can be considered optimum. The strain ratio based on Burger's model depicted a significant increase in the viscous strain as the WCO content increased. This indicates that the optimum WCO should be carefully selected, as even a slight increase in WCO beyond the optimum may lead to permanent deformation in mixtures.

(v) The ageing indices based on rheological and chemical parameters had an excellent correlation. The presence of optimum WCO was also confirmed based on ageing indices, which agreed with the rheological studies.

(vi) The addition of WCO may slightly increase the striping of bitumen in the mixtures beyond the optimum. Future studies may investigate the effect of WCO on durability aspects of bituminous mixtures in the presence of antistripping agents.

Data Availability

All data used to support the findings of the study are available in the paper.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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