



## Research Article

# Investigating the high-temperature performance and activation energy of carbon black-modified asphalt binder

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## Abstract

This study set out to evaluate the influence of carbon black (CB) on the rheological properties and activation energy of asphalt binder at high service temperatures. The rheological performance of virgin and modified asphalt binders are investigated using three evaluation approaches: (1) Superpave specification parameter ( $G^*/\sin\delta$ ), (2) multiple stress creep recovery (MSCR) test, and (3) interaction model for computing zero shear viscosity (ZSV). Moreover, the Arrhenius model was used to quantify the activation energy ( $E_f$ ) of virgin and CB-modified asphalt binders. The result of this study reveals that modifying asphalt binder with up to 10% of CB, by weight of the total asphalt binder, enhances the elastic behavior ( $R\%$ ) and decreases the non-recoverable creep compliance ( $J_{nr}$ ) of asphalt binder at high temperatures. Moreover, according to this study, the ZSV index can describe successfully rutting resistance of asphalt binder when compared with MSCR and Superpave rutting specification parameter. Besides, it was indicated that CB-modified asphalt binder has a high fluid resistance as more thermal energy was required for overcoming intermolecular force between molecules.

**Keywords** Rheology · Asphalt binder · Modification · Carbon black · Rutting resistance · Activation energy · Arrhenius model

## 1 Introduction

Dating back to many years ago, asphalt binder is still used for producing asphalt concrete, and it has been known as the most common construction material for paving the roadways [1]. However, researchers are still continuing to develop higher-quality asphalt binders resulting in more sustainable and resilient flexible pavements [2–5]. The overall performance of asphalt mixture is highly dependent upon the asphalt binder's intrinsic properties [6, 7]. Whereby, a high-quality asphalt binder can prevent or delay the environmental-and-traffic-related distresses in flexible pavements and hence increase the longevity of

these pavements. Asphalt pavement distresses, i.e., rutting and cracking, occur before the time that flexible pavements reach their service lives [8, 9]. These distresses result in huge amounts of money spent on rehabilitation or even reconstruction of flexible pavements; hence, it would be wiser to prevent such expenditures when it is possible to develop more sustainable and more resilient asphalt binders [10, 11].

High service temperature, high traffic load, asphalt mixture mechanical properties, and pavement structure are the main reasons collectively resulting in the occurrence of permanent deformation in the surface of flexible pavements [12]. Such factors contribute to cause permanent

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deformation/depression on wheel paths resulting in a poor serviceability [13, 14]. Although unrecoverable deformations in pavement layers and subgrade can collectively result in the occurrence of rutting on the pavement surface, the main reason for the occurrence of rutting is the nonlinear plastic deformation of asphalt concrete layer [15, 16]. Sousa et al. [17] concluded that the rutting in the asphalt mixture layer could be caused by both densification and plastic deformation due to traffic loading. According to Sousa et al. [17], the plastic deformation occurs with no volume change in the asphalt mixture in both loading area and upheaval zone. The viscoelastic behavior of asphalt mixture is only because of the presence of the asphalt binder. The aggregate skeleton has no contribution to the viscoelastic behavior, although the aggregate carries the majority of traffic loading. Therefore, the rheological properties of asphalt binder affect the plastic flow in the asphalt mixture [18]. Eisemmann and Hilmer [19] presented that the rutting, due to densification, is attributed to shear deformation without volume change. Enhancing the shear resistance of asphalt binder, through modification, would be an efficient method to mitigate this type of distress, given that there are no issues related to the mix design of asphalt concrete. For instance, numerous polymer-based modifiers can enhance the shear resistance of asphalt binder [11, 20, 21]. Although these polymer-based modifiers are added in small amounts, they can double the price of modified asphalt binder [22, 23]. To make the polymer modification method more economically viable, it is possible to use waste polymers [24, 25], or carbon-based by-products such as carbon black (CB) that is obtained from the incomplete combustion of heavy petroleum products [26]. CB not only is a good candidate for improving the rutting resistance, but also it imparts anti-aging properties to asphalt binder [27–29] and improves the physical and rheological behavior of asphalt binder [30, 31].

Furthermore, CB has a physical and chemical affinity with asphaltene and maltene that are the constituents of asphalt binder [32–34]. It has been proved that carbon atoms in the graphite-like sheets presented in the carbon black fortify carbonyl functional group (C=O) and C–O bonds of the asphalt binder [35]. It has been also claimed that the lighter portion of asphalt binder is absorbed by carbon black resulting a higher viscosity asphalt binder [36, 37]. In another way, CB incorporated in crumb rubber can transform into asphalt binder by absorbing paraffin and maltenes of asphalt binder [38]. This superior performance can be attributed to the particular surface property of CB having various functional groups that facilitate the interaction with hydrocarbon fluids like asphalt binder [32]. This feature may alter the bond strength between asphalt binder's hydrocarbon chains. There has been

increasing interest in the use of CB in asphalt mixture, because this material can be used as a conductive additive to enhance the anti-aging and thermal conductivity of asphalt mixtures [35].

In addition to experimental approaches, there has been many numerical methods investigating the permanent deformation in asphalt concrete resulting in the development of specification parameters capable of measuring as well as accounting for the rutting resistance of virgin and modified asphalt binders [39–43]. These numerical research efforts can help identify the influence of CB on changing the rheological behavior of asphalt binder. Besides, such a numerical identification approach facilitates the investigation of the effectiveness of CB on the high-temperature performance of asphalt binder. Moreover, CB particles interact with the light portion of asphalt binder that may change the intermolecular force due to the interaction of CB particles with maltene phase of asphalt binder. This is important, because the modified asphalt binder should be fluid enough for mixing and compaction purpose. Therefore, the required activation energy, the amount of thermal energy to overcome an energy barrier of asphalt binder's atoms and molecules to move to an adjacent vacant place, is still unclear for such modified asphalt binders. Although a few studies are dedicated to the mechanical properties of CB, the influence of CB on the rheological, permanent deformation resistance, and activation energy of asphalt binder is still a matter of concern and dispute.

This study investigates the effect of carbon black on the rheological and rutting performance as well as the activation energy of asphalt binder. For this purpose, the objective of this study is, first, to characterize the viscoelastic behavior of CB-modified asphalt binder through using different approaches: (1) measuring the Superpave specification parameter, (2) calculating the average recovery percentage, (3) calculating the non-recoverable creep compliance, and (4) calculating the zero shear viscosity (ZSV). Second, evaluate the effect of CB on asphalt binder's activation energy based on Arrhenius model.

## 2 Material and test methods

### 2.1 Materials and modification procedure

In this study, a virgin (neat) asphalt binder with an 85/100 penetration grade (with a performance grade of 58-22), obtained from Akam Binder Inc., was used as a base binder, and then it was modified by CB N375 obtained from Pars Carbon Black Company, Tehran, Iran. Table 1 presents the physical properties of CB used in this study. Also, Fig. 1

**Table 1** Physical properties of Carbon Black

Properties	Value	Standard
Iodine adsorption number (mg/g)	95–85	ASTM D1510
Absorption number (cm <sup>3</sup> /100 g)	119–109	ASTM D2414
Pelleted—pour density (g/L)	365–25	ASTM D1513

shows the flowchart of the modification procedure and executed tests.

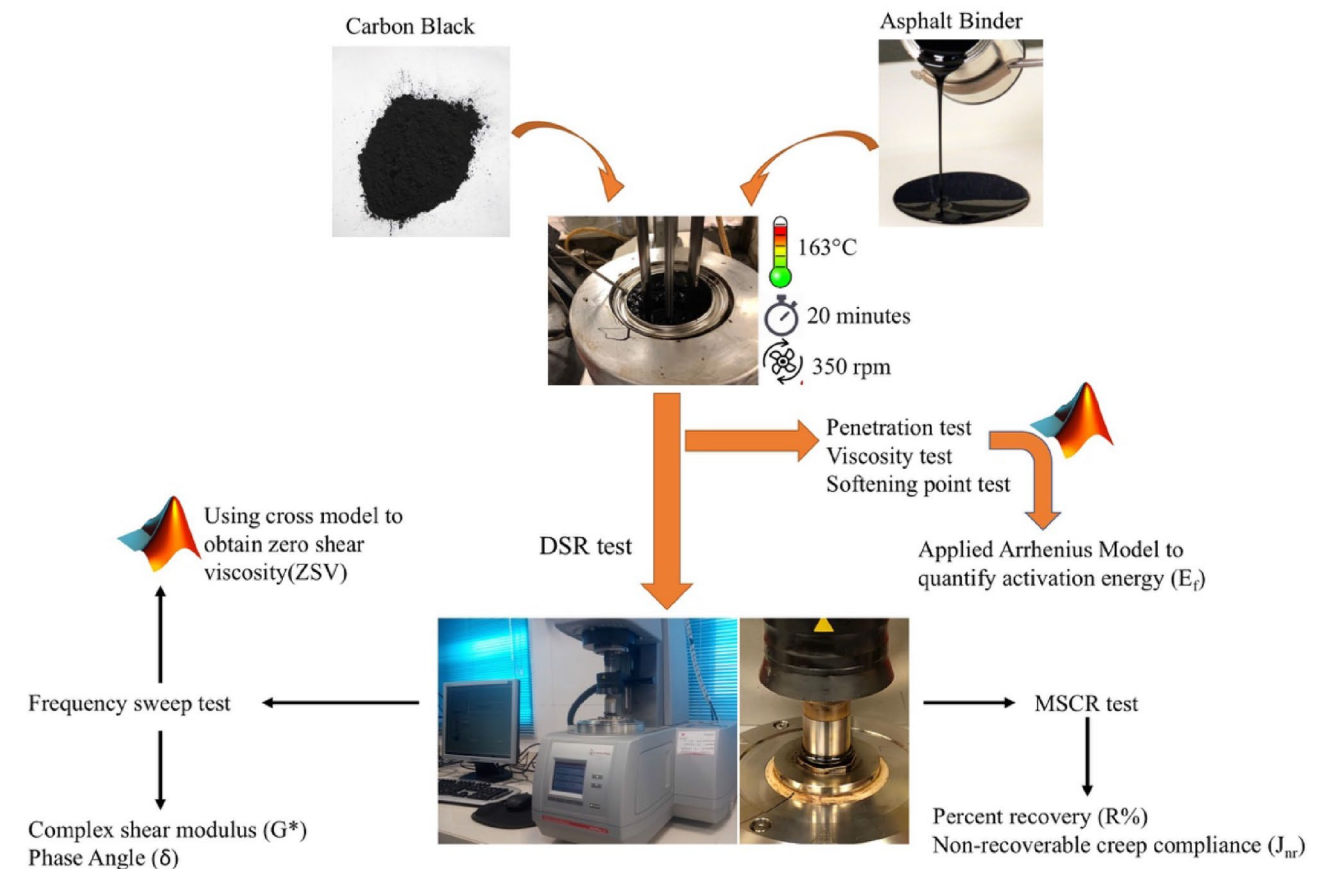
The CB was added to virgin asphalt binder at variable rates of 5%, 10%, and 15% by weight of total asphalt binder. As shown in Fig. 1, virgin asphalt binder and CB were blended using medium-shear radial flow mixer at a 350 revolution per minute (RPM) for 20 min at 163 °C. For simplicity, the virgin binder and the three-modified asphalt binders were coded as VB, CB 5%, CB 10%, and CB15%.

**2.2 Test methods**

In 1993, the Strategic Highway Research Program (SHRP) introduced the dynamic shear rheometer (DSR) apparatus

to characterize the rheological and viscoelastic behavior of asphalt binder. This characterization method has been inspired by the concept of torsional flow between parallel plates applying oscillatory and rotationally stresses/strains over a range of temperatures and loading frequencies [44]. The DSR applies rotational and oscillating stresses/strains to a slim film of binder located between two plates of DSR apparatus to obtain the viscoelastic parameters of asphalt binder at different temperatures.

As the foremost step, Anderson [45] introduced  $G^*/\sin\delta$  to characterize the rutting resistance of asphalt binder that is calculated based on the two main viscoelastic parameters: complex shear modulus ( $G^*$ ) and phase angle ( $\delta$ ). It is supposed that permanent deformation is caused by the total dissipated energy relating this parameter to rutting behavior in asphalt binder. To evaluate the required amount of thermal energy to overcome the intermolecular force was calculated using the Arrhenius model based on viscosity data captured from viscosity test. The rotational viscosity is determined by measuring the required torque to maintain the cylindrical spindle rotational speed while submerging in a steel mold filled with binder at a constant rate (AASHTO TP4). It is wise to note that viscosity value is



**Fig. 1** A graphical schematic of modification and testing process

measured at constant speed of 20 RPM. Moreover, it was decided to measure the viscosity of virgin and CB-modified asphalt binders at 135, 150, 165°C.

### 2.2.1 Superpave specification parameter

Bahia and Anderson [46] and Robert et al. [47] have reported that complex shear modulus ( $G^*$ ) and phase angle ( $\delta$ ) obtained from DSR determine the relationship between asphalt binder stiffness and its deformation type. It is justifiable to regard rutting resistance at high service temperatures in the early age of pavement serviceability, because, at these temperatures, the behavior of asphalt binder is more viscous rather than elastic. The higher the  $G^*$ , and lower the  $\delta$ , the greater the rutting resistance of asphalt binder. An asphalt binder with higher value of  $G^*$  is stiffer and provides increased resistance against deformation. Therefore, SHRP proposed to use  $G^*/\sin\delta$  as the Superpave rutting specification parameter. It is wise to note that after emerging polymer-modified asphalt binders, the multi stress creep and recovery test has been introduced to describe the stress dependency and anti-rutting resistance of modified asphalt binders.

To provide robust reasoning about the influence of CB on the high-temperature viscoelastic behavior of binder, the Superpave parameter ( $G^*/\sin\delta$ ) was measured for both virgin and modified specimens at various test temperatures. Moreover, to observe the interaction of temperature and the CB-dosage effects on the Superpave high-temperature parameter, it was decided to define a rutting resistance improvement ratio for the modified binder which was obtained by dividing  $G^*/\sin\delta$  of CB-modified binder to that of the control sample (i.e., virgin binder). To this end, the result of the frequency sweep test at 10 rad/s was selected to calculate this ratio.

### 2.2.2 Zero shear viscosity

At high temperatures, virgin asphalt binder behaves like a Newtonian fluid making asphalt mixtures more prone to rutting. When the rate of loading is very low, the molecular transition rate of asphalt binder is very slow that makes its behavior like Newtonian fluids, so acclimation microstructure is done uniformly, on the other word; the asphalt binder, in this case, is in equilibrium condition. In this condition, the viscosity of binder is called zero shear viscosity (ZSV). However, asphalt binder demonstrates pseudoplasticity, or shear-thinning fluid behavior, due to which binder's viscosity decreases with an increase in shear rate. At very low shear rates, the pseudoplastic liquids act like Newtonian fluids, and their viscosity is not dependent on shear rate or loading frequency. This viscosity, i.e.,  $\eta_0$ , is referred to as zero shear viscosity (ZSV) [48]. Sybilski [49]

illustrated that ZSV can be used for evaluating the rutting resistance of asphalt mixture. There are several techniques to compute the ZSV value for asphalt binders. In this study, the frequency sweep test was used to compute the ZSV—assuming that the asphalt binder is a cross fluid. A cross fluid is a Newtonian liquid whose viscosity depends on the applied shear rate [49–51]. As a result, the flow curves were fitted to the cross model (Eq. 1).

$$\frac{\eta^* - \eta_\infty^*}{\eta_0^* - \eta_\infty^*} = \frac{1}{1 + (k\omega)^m} \quad (1)$$

where  $\eta^*$  is complex viscosity,  $\eta_0^*$  is zero shear viscosity (ZSV),  $\eta_\infty^*$  is limiting viscosity,  $\omega$  is angular frequency (rad/s), and  $m$  and  $k$  are constant coefficients. The frequency sweep test is commonly implemented between 0.1 and 100 rad/s. Within this range, it is assumed that  $\eta^* \gg \eta_\infty^*$ . As a result, the cross model can be rewritten as (Eq. 2):

$$\eta^* = \frac{\eta_0^*}{1 + (k\omega)^m} \quad (2)$$

Using the curve-fitting tool in Matlab<sup>®</sup>, it would be possible to fit Eq. (2) to complex viscosity versus angular frequency data point for each temperature to determine the ZSV value. The ZSV value is straightly proportional to permanent deformation resistance, and a greater ZSV is representative of higher anti-rutting resistance in asphalt binder [14]. To capture the viscoelastic properties of virgin and modified asphalt binders, the frequency sweep test was performed under a controlled-strain mode at frequencies ranging from 0.1 to 100 rad/s. The shear strain of 1% was selected to ensure that the asphalt binder response remains in the linear viscoelastic zone. Also, to capture the rutting behavior of asphalt binders, the frequency sweep test was conducted at temperatures above 46 °C—46, 52, 58, and 64 °C—which the rutting is most likely to happen.

### 2.2.3 Average recovery percentage and non-recoverable creep compliance

Multi stress creep and recovery (MSCR) test has been alternatively used for examining the rutting resistance of asphalt binders when Superpave specification parameter fails to determine a justifiable correlation between loading type, loading level, and stress level. Average recovery percentage ( $R\%$ ), which is calculated based on the data acquired from MSCR test, can directly examine the elastic response of asphalt binders under creep and recovery loading [52]. The non-recoverable creep compliance ( $J_{nr}$ ) that is also obtained from the MSCR test is a useful parameter to examine the rutting behavior of asphalt binder. In this study, the multi stress creep and recovery (MSCR) test

was conducted following the ASTM D7405-15 standard. The loading and recovery durations, respectively, were 1 and 9 s. Ten creep–recovery cycles were applied for each stress levels: 0.1 and 3.2 kPa. It should be noted that the frequency sweep and MSCR tests were performed by an Anton Paar rheometer A101 with 25-mm diameter parallel plates spaced with a 1-mm gap opening at 46, 52, 58, and 64 °C, to capture viscoelastic properties of virgin and CB-modified asphalt binders.

### 2.2.4 Activation energy investigation method

Since asphalt binder is a viscoelastic material, its viscous portion can be considered as a thermally activated process in which the molecules of binder should overcome an energy barrier to move. In other words, when asphalt binder flows, the layer of molecules slide on each other, and as a result, intermolecular forces cause resistance against flow [49]. The amount of energy required to overcome this resistance called activation energy, which can be obtained by employing the Arrhenius model as follows:

$$\eta = Ae^{\frac{E_f}{RT}} \quad (3)$$

where  $\eta$  is the viscosity of asphalt binder,  $T$  is the temperature in Kelvin,  $A$  is a model constant,  $E_f$  is the activation energy, and finally,  $R$  is the universal gas constant equal to  $8.314 \text{ J mol}^{-1} \text{ K}^{-1}$ .

To simplify this equation for obtaining  $E_f$ , Eq. (3) can be rewritten as follows:

$$\ln \eta = + \ln A \frac{E_f}{RT} \quad (4)$$

Equation 4 shows that the slope of  $\ln \eta$  versus  $1/T$  equals to  $E_f/R$ , because the trend is linear. Therefore, it is possible to determine the activation energy by considering the slope of  $\ln \eta$  versus  $1/T$ . The slope of this plot gives us the  $E_f/R$ . As a result, considering the universal gas constant,  $E_f$  can be calculated easily. For instance, the trend for obtaining the  $E_f$  value for the virgin binder is shown in Fig. 2.

## 3 Result and discussion

### 3.1 Physical properties

According to Table 2, increasing CB content decreases the penetration of asphalt binder, making it stiffer, and the lowest penetration value is obtained at the CB of 15%. Such behavior can be attributed to the consistency of the modified asphalt binder due to the interaction of CB particles with the asphalt binder components. Generally, increasing CB content in the modified blend increases the

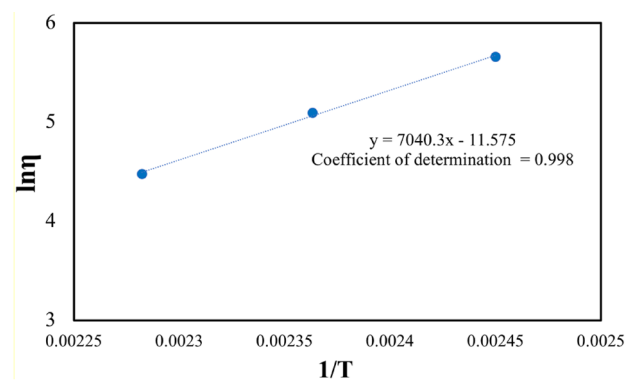


Fig. 2  $\ln \eta$  versus  $1/T$  for virgin asphalt binder

Table 2 Physical properties of virgin and CB-modified asphalt binders

Sample	Viscosity (135 °C) pa s	Penetration (25 °C) 0.1 mm	Softening point (°C)
OB	0.287	98	45
CB 5%	0.350	81	47.3
CB 10%	0.427	72	48.4
CB 15%	0.436	69	48.6

ratio of asphaltene to maltene, because the light portion of asphalt binder is absorbed by CB particles, which transform it high viscosity asphalt binder. The softening point, however, increases with the increase in CB content, which can be related to change in the ratio of maltene/asphaltene as well.

CB particles absorb the light portion of the binder, increasing the ratio of asphaltene/maltene, and as therefore, the fluidity (the ability of an asphalt binder to flow easily) of the binder decreases. Consistently, the results of the viscosity of virgin and modified asphalt binder illustrate a good agreement with binder fluidity as the great viscosity related to CB 15% (Table 2).

### 3.2 Viscoelastic properties

Figure 3 presents two main viscoelastic parameters for virgin and CB-modified asphalt binders. As can be seen from Fig. 3a, increasing CB content in the blend increases the complex shear modulus in which the highest value attributed to CB 15%. With an explorer in Fig. 3, there is clear trend of decreasing phase angle with adding CB in which the lowest phase angle attributed to the CB 15% that implies the idea that it has more rutting resistance compared to others. It can be related to the high surface area of CB particles that absorbs the aromatic and light

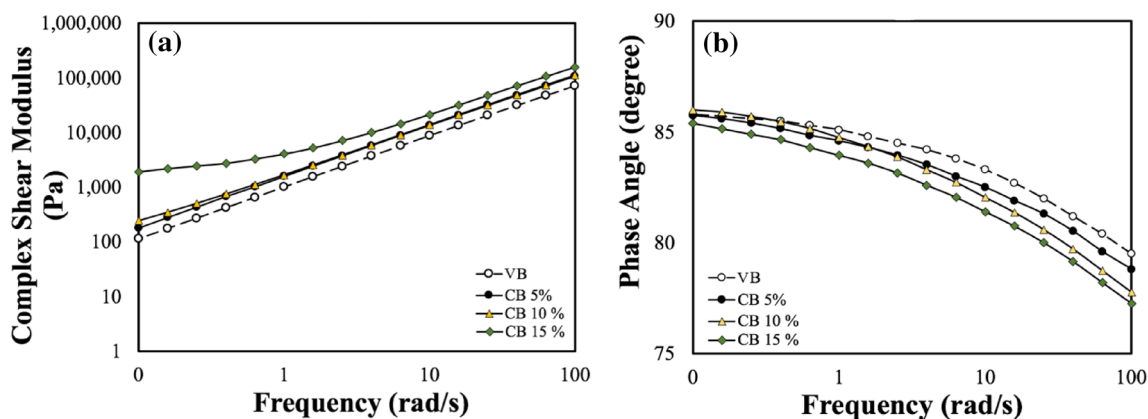


Fig. 3 a Complex shear modulus ( $G^*$ ) and phase angle ( $\delta$ ) versus loading frequency ( $\omega$ ) for virgin and CB-modified asphalt binders at 46 °C

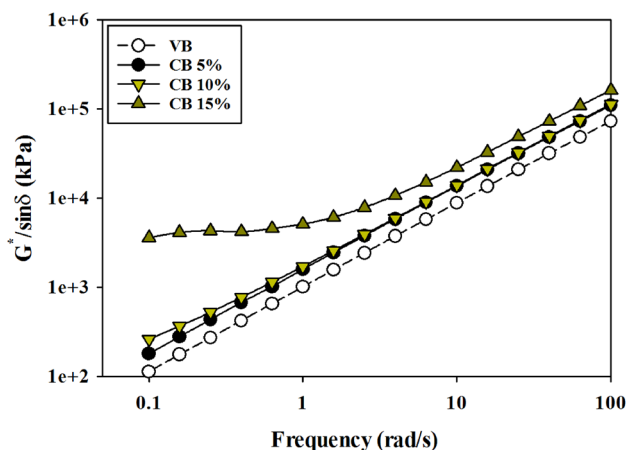


Fig. 4  $G^*/\sin\delta$  for virgin and CB-modified binders at 46 °C

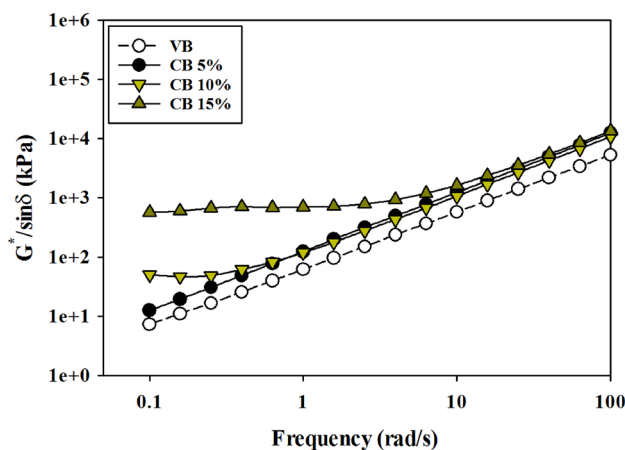


Fig. 5  $G^*/\sin\delta$  for virgin and CB-modified binders at 64 °C

portion of the asphalt binder. As a result, the binder will be stiffer compared to the virgin asphalt binder.

Figures 4 and 5 present the Superpave rutting parameter for both virgin and CB-modified binders. It can be seen that modifying binder with CB increases the value of  $G^*/\sin\delta$ . At low frequencies, i.e., the frequencies smaller than 3.98 rad/sec, the difference between the value of the  $G^*/\sin\delta$  for the virgin binder and the modified binders increases gradually with a decrease in frequency. It is worth noting that, at both temperatures, the difference between the  $G^*/\sin\delta$  values is the largest when comparing virgin binder and the binder modified with 15% CB. Modification of asphalt binder with CB increases the  $G^*/\sin\delta$  value, which leads to improved rutting resistance compared with the virgin asphalt binder.

### 3.3 Superpave rutting specification parameter

Comparing Figs. 4 and 5 proves the key role of temperature on influencing the rutting resistance of asphalt

binder. At 64 °C, the  $G^*/\sin\delta$  values are smaller than the values calculated for binders tested at 46 °C. Also, raising the temperature to 64 °C makes the influence of CB more distinguishable at lower frequencies due to the fact that at higher temperature or lower frequencies, CB-modified binder presents a high ratio of asphaltene to maltene enhancing binder temperature susceptibility, where the asphalt binder is the most prone to rutting.

Figure 6 presents the rutting improvement ratio based on data obtained from the frequency sweep test at 10 rad/s. Whereby this ratio can be calculated by dividing  $G^*/\sin\delta$  of CB-modified binder to  $G^*/\sin\delta$  of the virgin binder at the loading frequency of 10 rad/s. The rutting resistance ratio of the binder modified with CB increases with increase in temperature. This improvement can be justified by the increased ratio of asphaltene/maltene due to absorption of a portion of maltenes by CB particles. Moreover, when the specimens are subjected to temperature increases, the increased rate of  $G^*/\sin\delta$  for asphalt binder modified with 10% CB is greater than those

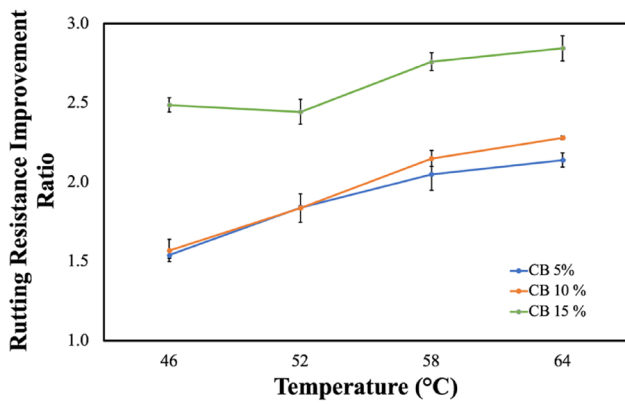


Fig. 6 Rutting resistant improvement ratio at all test temperatures at 10 rad/s

of asphalt binders modified with 5% and 15% CB. In other words, the asphalt binders modified with 5% and 15% CB become less temperature susceptible, in the context of rutting, which can be attributed to the presence of very small amounts of light portion in these modified binders.

According to Fig. 6, by increasing CB content in the blend, the rutting resistance improvement ratio raises, essentially the highest improvement attributed to the CB 15%. It can be justified in terms of the interaction between CB particles and asphaltene in binder colloidal structure. Due to the high absorption percentage of CB particles, modifying binder with CB causes CB particles covered with a layer of asphaltene leading the binder to low mobility. As a result, the viscosity of the modified asphalt binder is increased. To support it, the viscosity value given in Table 2 presents a good agreement to this explanation.

### 3.4 Zero shear viscosity

As it was mentioned in the test method section, the cross model is a suitable means for computing the ZSV value of asphalt binder. This model consists of three parameters; zero shear viscosity, complex viscosity, and two coefficients that can be obtained using the fit-curve technique performed on the data acquired from the frequency sweep test at each temperature. A nonlinear regression analysis was used in Matlab® to fit Eq. (2) to the acquired data. Figure 7 represents the complex viscosity versus angular frequency data obtained from a frequency sweep test at the test temperature of 64 °C. As it can be observed CB 10% and 15% have a high complex viscosity at low frequencies, because binder modification improves permanent deformation resistance.

To characterize the rutting performance of CB-modified binders at high temperatures, the ZSV values were calculated for both modified and virgin binders. Table 3 shows

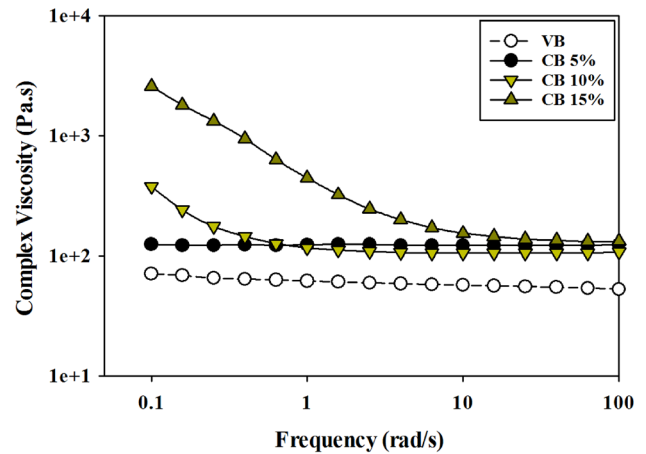


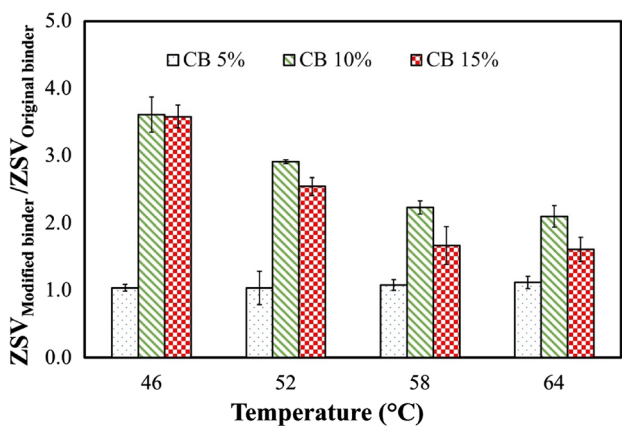
Fig. 7 Complex viscosity versus angular frequency at 64 °C

the computed ZSV for both virgin and CB-modified asphalt binder. With an explore in this table, it is obvious that increasing test temperature reduces ZSV value. Moreover, extending the CB content in the modified blend up to 10% by weight of total binder asphalt binder increases the ZSV value, and for further content, it is reduced.

To have a better understanding, the ZSV values calculated for modified binders were divided by that of virgin binder to obtain indices. These indices were used for making a comparison between the CB-modified binders in terms of rutting resistance enhancement. Note that in Fig. 8, increasing the amount of CB in the blend increases the ZSV value. At all the temperatures, CB 10% has the highest indices, followed by CB 15% and then CB 5%. The calculated index for CB 5% remains constant at all the test temperatures. CB 10% and CB 15% have the highest indices at the test temperature of 46 °C, and their indices, with a slight reduction at the test temperature of 52 °C, remain virtually constant at higher tests temperatures. This stable behavior can be attributed to the fact that the binder softens and loses its stiffness at these high temperatures (i.e., 52 °C ≤ T ≤ 64 °C). It is worth highlighting that the results obtained from the analysis performed on ZSV values are in contradiction with those obtained from the analysis

Table 3 Value for virgin and CB-modified asphalt binders at different temperatures

Zero shear viscosity (Pa s)				
Temperature (°C)	46	52	58	64
Virgin binder	8880	3440	1380	573
CB 5%	9217	3563	1495	642
CB 10%	32,057	10,010	3077	1203
CB 15%	31,790	8744	2305	923



**Fig. 8** Rutting resistant improvement at all test temperature based on ZSV

performed on Superpave high-temperature specification parameter.

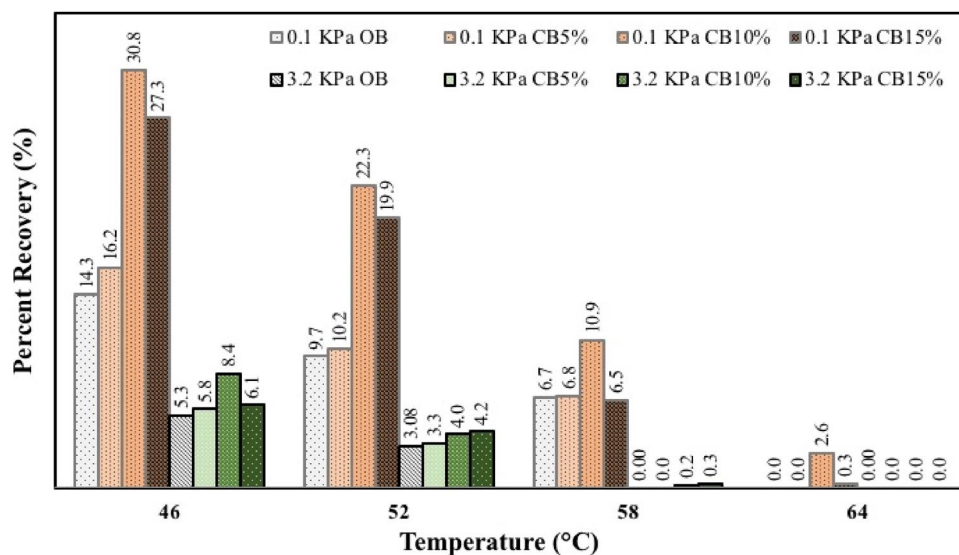
### 3.5 Average recovery percentage and non-recoverable creep compliance

The average recovery percent ( $R\%$ ) obtained from the MSCR test is representative of asphalt binder’s elasticity under cyclic loading, and the higher the  $R\%$ , the higher the deformation recovery. As a result, it can be stated that a binder with a higher  $R\%$  has more elasticity. Figure 9 shows the recovery percentage of the virgin binder and CB-modified asphalt binder at two stress levels: 0.1 and 3.2 kPa, respectively. The highest  $R\%$  is obtained from the binder modified with 10% CB. Therefore, the addition of CB at content about 10% of binder weight results in enhancement of elastic recovery as well as rutting resistance in

asphalt binder. Moreover, increasing the CB to a percentage beyond 10% (e.g., 15%) results in a reduction in elastic recovery. At all the selected test temperatures, both virgin and CB-modified asphalt binders show sensitivity to change in stress level, i.e., the  $R\%$  values calculated for all the binder types are greater at the low-stress level of 0.1 kPa than those calculated for the stress level of 3.2 kPa. The reason for the reduction in  $R\%$  at higher stress levels can be attributed to the stress dependency of asphalt binder in which higher stress level making the viscous behavior more dominant. Also, increasing the temperature causes reduction in the recovery percentage of asphalt binder which can be attributed to increasing of maltene in asphalt binder at higher temperatures making the viscous behavior more dominant [50]. At the stress level of 0.1 kPa, CB 10% and CB15% show a better performance in terms of having higher  $R\%$  values at all the test temperatures in comparison with virgin asphalt binder. Such improvement may be attributed to the reduction in maltene absorbed by CB particles making the asphaltenes’ role more dominant. Moreover, the highest recovery percentage is related to the modified asphalt binder containing 10% CB.

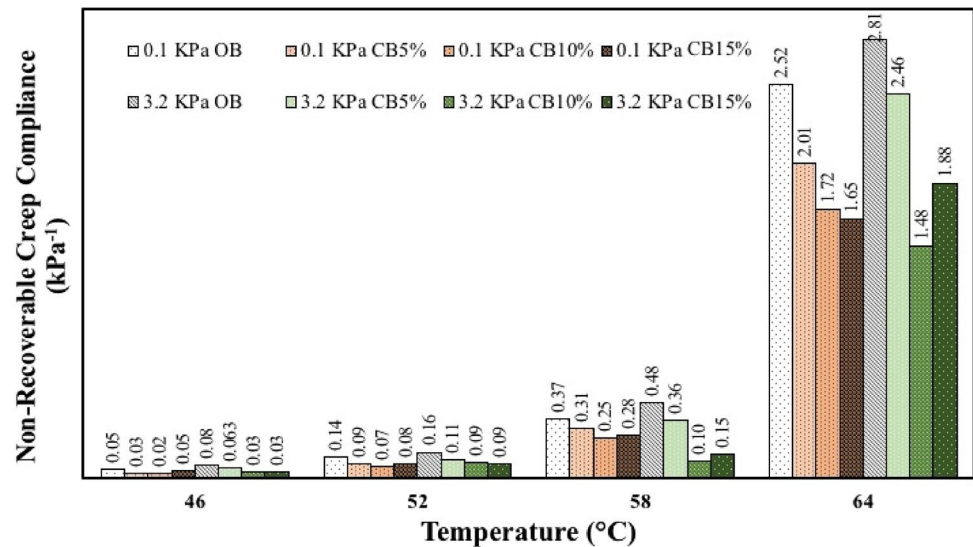
The non-recoverable creep compliance ( $J_{nr}$ ) is also obtained from the MSCR test and is used for characterizing the viscoelastic response of asphalt binder. The lower the  $J_{nr}$  value, the higher the rutting resistance. According to the findings shown in Fig. 10, the presence of CB results in the reduction in  $J_{nr}$  value at both stress levels, and this reduction is the greatest at the test temperature of 64 °C, followed by 58 °C. Moreover, at the high-stress level of 3.2 kPa, the influence of CB on the viscoelastic response is greater than the low-stress level of 0.1 kPa. Note that in Fig. 10, at the test temperature of 64 °C, the  $J_{nr}$  value of asphalt binder modified with 10% CB has decreased for

**Fig. 9** Recovery percentage of virgin and CB-modified asphalt binders





**Fig. 10** Non-recoverable creep compliances of virgin and CB-modified asphalt binders

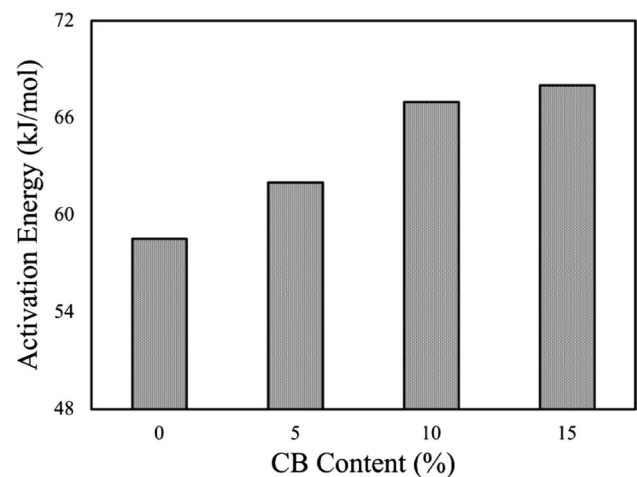


about 32% and 48%, respectively, at the stress levels of 0.1 kPa and 3.2 kPa.

After considering all analysis approaches, it can be concluded that the ZSV analysis can be more representative of high-temperature performance compared with the Superpave rutting specification parameter when comparing those to the MSCR result analysis. In other words, according to Superpave rutting specification, for a high dosage of CB (i.e., 15%), the rutting resistance was increased, while, according to MSCR test result, the elastic recovery of asphalt binder decreased. This discrepancy can be explained in part by the idea that the presence of high content of CB in the modified blend increases the probability of phase segregation in the asphalt binder. As a result, deposition of heavy molecules might take place, which can disturb the stability of maltene and asphaltene phases in the CB-modified asphalt binder. Moreover, the ZSV analysis placed in a good agreement with the MSCR test result. Simply put, it can be stated that ZSV can be used as an excellent rutting resistance representative approach. Based on such result and discussion, it can be concluded that the CB content of 10% is the maximum recommended value for increasing the resistance of asphalt binder against rutting.

### 3.6 Activation energy

Figure 11 presents the activation energy that requires to overcome the barrier force between modified asphalt binder's molecules to move. This feature is significant from energy consumption at the hot mix asphalt (HMA) production process, in which, higher activation energy implies the idea that the molecules of binder need more kinematic energy to overcome the intermolecular force to the fluid. As can be seen from Fig. 11, modifying asphalt binder



**Fig. 11** Required activation energy for virgin and CB-modified asphalt binders

with CB increases activation energy as the highest value related to modified asphalt binder containing 15% of CB. It implies the fact that CB interacts with asphalt binder maltene portion that results in larger hydrocarbon chains. Since the high activation energy implies the fact that in the HMA production process, more heat energy required to make the binder fluid enough for pumping and mixing with aggregate. As a result, it increases the energy consumption and then increases the final cost of the material. Moreover, it increases greenhouse gas emission, especially in the production process of HMA.

It was claimed that the extra thermal energy generated for modified asphalt binders, make asphalt binder more susceptible to short-term aging [53]. From this standpoint, it is recommended to evaluate the short-term aging of CB-modified asphalt binder for future works.

## 4 Conclusion

This study set out to evaluate the effect of CB on asphalt binder rheological performance and fluid resistance at high service temperatures. To capture such objectives, three approaches were adopted to assess CB on rutting and rheological performance of asphalt binders: Superpave rutting specification parameter ( $G^*/\sin\delta$ ), zero shear viscosity (ZSV), and multiple stress creep and recovery (MSCR) test as well as a cross model were used to quantify of fluid resistance by introducing activation energy concept. This study led to the following outcomes:

- Increasing CB content in the modified asphalt binder up to 10% amplifies the enhancement trend of asphalt binder's rutting resistance.
- CB-modified asphalt binder indicates low non-recoverable creep compliance compared to the virgin asphalt binder, especially for 10% CB content.
- Extending carbon black content in the modified asphalt binder results in stiffer asphalt binder in which the highest stiffer asphalt binder containing 15% CB content.
- Addition of CB beyond 10% either maintains or degrades the elastic recovery and non-recoverable creep compliance.
- The effect of CB on the elastic properties of asphalt binder at the high-stress level is more representative than lower loading stress level.
- Binder modified with 10% CB performs better in terms of ZSV and MSCR values on  $J_{nr}$  either in 0.1 or 3.2 kPa stress level.
- Comparing different rutting assessment approaches shows that the zero shear viscosity (ZSV) can be used as an excellent representative approach for evaluating the elastic behavior of asphalt binder for high service temperature performance.
- CB-modified asphalt binders indicated higher activation energy in comparison with the virgin binder that increases the total material cost of HMA during the plant production process.

This study focused on the effect of CB on rutting resistance of asphalt binder in which CB content of 10 percent was concluded as the optimum percentage that valid for high-temperature performance based on the analysis discussed in this study. However, for determining the optimum content of CB, it is recommended to evaluate the low and intermediate temperature performance of CB-modified asphalt binder. Since there is no comprehensive state of the art on the performance of

CB-modified asphalt binder and mixture, it is recommended to investigate the physicochemical properties of CB-modified asphalt binder in terms of chemical functional groups (i.e., carbonyl and sulphoxide functional groups) and the short/long term aging resistance of CB-modified asphalt binder. Moreover, it is suggested to extend the analysis level from asphalt binder to asphalt mixture and then validate the ZSV rutting evaluation approach with rutting performance of asphalt mixture to establish a promising correlation between them.

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**Conflict of interest** The authors declare that there is no any conflict of interest regarding this study.

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