

# EXPERIMENTAL INVESTIGATION OF HIGH TEMPERATURE BEHAVIOUR OF AN ASPHALT BINDER MODIFIED WITH LAVAL UNIVERSITY SILICA BASED ON MULTIPLE STRESS CREEP AND RECOVERY TEST

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REZA FALLAH<sup>1</sup>, GHOLAMALI SHAFABAKHSH<sup>1\*</sup>,  
ZOHREH BAHRAMI<sup>2</sup>

<sup>1</sup>*Department of Road and Transportation Engineering, Faculty of Civil Engineering, Semnan University, Semnan, Iran*

<sup>2</sup>*Department of Nanotechnology, Faculty of New Sciences and Technologies, Semnan University, Semnan, Iran*

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**Abstract.** For experimental investigation of the high temperature behaviour of an asphalt binder modified with Laval university silica (LUS-1) nanostructured particle, four different asphalt binders were produced using a mixture of 2, 4, 6 and 8 wt% of this additive and a neat bitumen at 170 °C. The neat bitumen was yielded from crude oil refining and had a penetration grade of 85–100. After a 20 min vibration, the produced mixtures were mechanically mixed for 30 min in a high-shear homogenizer mixer with an angular velocity of 4500 rpm. Then, the modified binders and neat bitumen were subjected to multiple stress creep and recovery (MSCR) test, after the aging process in rolling thin film oven (RTFOT) test. The results of this study, which were in agreement with the results of the

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\* Corresponding author. E-mail: ghshafabakhsh@semnan.ac.ir

Reza FALLAH (ORCID ID 0000-0002-5083-2396)  
Gholamali SHAFABAKHSH (ORCID ID 0000-0002-9127-8379)  
Zohreh BAHRAMI (ORCID ID 0000-0002-3990-9292)

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dynamic shear rheometer (DSR) test, indicated that LUS-1 could improve the high temperature behaviour of binders. The greatest improvement occurred using 4 wt% of LUS-1, where this improvement was more pronounced at high stress levels. Elevating the levels of stress and temperature led to diminished traffic grade and more viscous behaviour in asphalt binders modified with LUS-1.

**Keywords:** high temperature behaviour, modified asphalt binder, multiple stress creep and recovery test, nanostructured silica, non-recoverable creep compliance, recovery percent, rutting.

## Introduction

Rutting is one of the most important types of distress in asphalt pavement. This distress is more pronounced in pavements which are subjected to heavy and slow traffic. Use of efficient aggregates as well as selecting asphalt binders with appropriate elasticity can be considered as two strategies in improving resistance to pavement rutting. Application of an efficient modified binder has been the top priority of researchers for reducing rutting, extending pavement lifespan, and saving money in pavement maintenance and repair due to the high cost of upgrading local aggregates or transporting efficient aggregates from out of the project site. Ensuring the quality of asphalt binders to resist rutting has been possible by conducting tests on superior pavement (Superpave) specifications. However, these tests have been performed only in the linear viscoelastic region, while the stress level will be different in an asphalt mixture, based on the applied load, contact area of the tire, time of loading, and temperature. Further, the magnitude of asphalt binder strain in the asphalt mixture, which is subjected to loading at high temperatures, can be hundreds of times greater than the overall bulk strain of the asphalt mixture. Therefore, rutting of asphalt mixtures is a phenomenon affected by the stress level and is nonlinear viscoelastic. Thus, the linear viscoelastic characteristics of Superpave specifications, appearing in the form of dynamic shear rheometer (DSR) test, will not be able to directly describe the rutting of asphalt mixtures, and this can only be achieved by multiple stress creep and recovery (MSCR) test (D'Angelo, 2009). Different values of the optimum content can be achieved for the modifier, given the type of binder response, in terms of being linear or non-linear in the tests. This highlights the importance and difference of applying MSCR test as compared to DSR test in analyzing the high temperature performance of binders (Mansourian et al., 2019). Also, the parameters of MSCR test had a good capability for predicting the rutting of asphalt mixtures compared to other rheological indicators of asphalt binders (Laukkanen et al., 2014; Lv et al., 2019).

Numerous studies have been conducted on the ability of the MSCR test in determining the high temperature behaviour of binders modified by different modifiers. In this regard, the binder modified by titanium dioxide (TiO<sub>2</sub>) nano-particles had less value of  $J_{nr}$  than neat bitumen and it was more sensitive to unexpected stress changes that might occur on the surface of pavement (Marinho et al., 2019). The results of the MSCR test also illustrated that the accumulated strain would be significantly reduced by using lower rubber fineness and lower grade neat asphalt in producing asphalt binders modified with rubber, and it was proved that  $J_{nr}$  and  $R$  indicators could be used in order to determine the high temperature properties of asphalt binders modified by rubber powder (Lei et al., 2016). Combining anti-stripping agents (ASAs) with crumb rubber modified asphalt (CRMA) reduced  $J_{nr}$  increased  $R$  and, thereby, increased resistance to deformation, which was in agreement with the results of  $G^*/\sin(\delta)$  obtained in DSR test (Tang et al., 2019). Increasing  $R$  and decreasing  $J_{nr}$  by modifying neat bitumen and binders containing a high percentage of polymer with SBS were some of the achievements of other researchers (Babagoli et al., 2018; Lin et al., 2019). In one study, it was observed that asphalt binder response shifted from linear viscoelastic region to non-linear viscoelastic region by increasing modifier dosage in long creep periods compared to short creep periods (Ashish & Singh, 2019). In other studies, using different additives, such as SBS, crumb rubber, and carbon nano-tubes, resulted in increased  $R$ , decreased  $J_{nr}$  and, consequently, increased asphalt binder resistance to rutting based on MSCR test (Zhou et al., 2019, Dong et al., 2019; Yang et al., 2019). In laboratory research, the non-recoverable creep compliance was increased for both neat and modified binders at the stress level of 3.2 kPa, by increasing aromatic compounds and small asphalt molecules (Wang C. & Wang Y., 2019).

Modification of asphalt binders with nanosilica has been the topic of some recent research. Studies on bitumen modified by a combination of crumb rubber, microcrystalline synthetic wax, and nano-silica have shown improvements in high temperature performance of asphalt binders, such that the effect of crumb rubber and nano-silica has been significant while the impact of microcrystalline synthetic wax has been trivial (Al-Omari et al., 2020). Studies on asphalt binders modified with a mixture of nano-silica and crumb rubber in a wet process have revealed the role of nano-silica in improving the high temperature properties and very minor reduction of low temperature ductility of the asphalt binder (Han et al., 2017). The use of nano-silica in a modified asphalt binder showed a significant rise in the complex shear modulus when compared to using neat bitumen (Villacorta & Nordbeck, 2019). Modification of asphalt binder with nanosilica showed significant improvements in permanent deformation of asphalt binder (Villacorta & Nordbeck, 2019).

The rutting performance of micro silica modified asphalt mixtures was discussed and improvement in permanent deformation of hot mix asphalt was reported (Shafabakhsh et al., 2015). It was also reported that the nano  $\text{TiO}_2/\text{SiO}_2$  modified bitumen improved the rutting performance of asphalt mixtures containing steel slag aggregates (Shafabakhsh & Ani, 2015). Researchers reported that enhanced asphalt performance at high and low temperature is achieved using nano- $\text{SiO}_2$  and polymers as composite modifiers (Sun et al., 2017). Another study concluded that the addition of nano-silica augmented the stiffness of the binder and reduced its ductility and temperature sensitivity (Taherkhani & Afroozi, 2016). Doping of multi-walled carbon nanotubes with nano-silica and adding them to the bitumen improved the high temperature performance of the binder (Saltan et al., 2018). It was also indicated that nanosilica-modified asphalt binders showed improved rutting performance (Saltan et al., 2017). The improvements in rheological properties of nanosilica and rock asphalt modified bitumen were observed (Shi et al., 2018).

Researchers showed tendency to use different nanomaterials for modification of asphalt binder, based on the fact that mechanical behaviour of bituminous materials depends to a high extent on microstructure of these materials on a micro and nano-scale (Fischer & Cernescu, 2015; Li et al., 2017; Yang & Tighe, 2013). In this regard, the results of MSCR test which was performed only at high temperature performance grade of neat bitumen, revealed that there was more modification effects on the permanent deformation of asphalt binders in nano-silica with a high specific surface area and fibrous morphology, than other custom nanosilica particles with spherical morphology (Moeini et al., 2019).

In order to better evaluate the high temperature behaviour of asphalt binders modified with nanosilica particles with a high specific surface area and fibrous morphology, this study was conducted not only at high temperature performance of neat bitumen but also at higher temperatures and more numbers of asphalt binders. Due to higher capability of MSCR than DSR test in evaluating the high temperature behaviour of asphalt binders, this study was performed using MSCR test. The modifier used in this research was a new generation of nanosilica materials with a high specific surface area and fibrous morphology called mesoporous silica compounds, made at Laval University of Canada in 2003, and known under the scientific name of LUS-1 in international papers (Bonnevot et al., 2003). The LUS-1 has been a topic of interest for researchers in different fields due to its non-toxicity, biocompatibility, high mechanical and chemical stability, high surface area, as well as pore volume (Wanyika, 2013; Bahrami et al., 2015). Thus, it has the potential to be added to neat bitumen as a modifier. Good thermal and

hydrothermal stability of this modifier can help increase the durability of hot mix asphalt pavements modified with it in warm-arid or warm-humid areas. The most important property of the LUS-1 nanostructured additive is that its morphological structure is in the form of micro-sized fibres, while these fibres contain nano-sized pores with 3 nm dimensions. This means that its agglomeration ability in mixing with bitumen is significantly reduced by the outer micrometric fibrous structure of this nanoparticle, compared to custom nano particles. On the other hand, because of its numerous nanometric inner pores, it also enjoys all the specific properties of nano particles and its performance can be modified for different executive purposes by embedding other chemicals into its channels (Bonnevot et al., 2003; Beck et al., 1992).

## 1. Materials and methods

### 1.1. Materials

The research materials, as shown in Fig. 1, include neat bitumen and LUS-1 mesoporous silica as bitumen modifier. The base bitumen used in this study is a neat bitumen with Marshall Specification of 85–100, obtained from crude oil refining (see Table 1). The additive, used for bitumen modification in this research, is LUS-1.

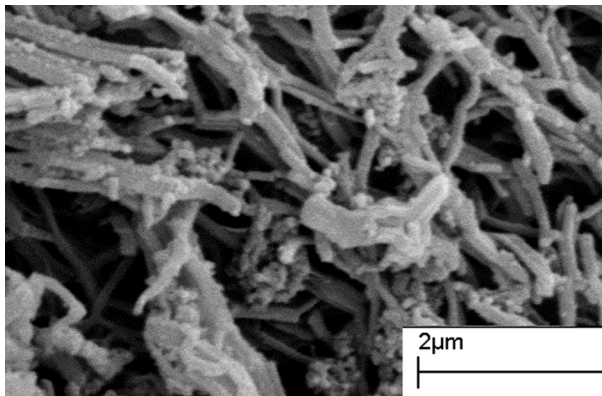


**Figure 1.** The neat bitumen and its modifier (LUS-1)

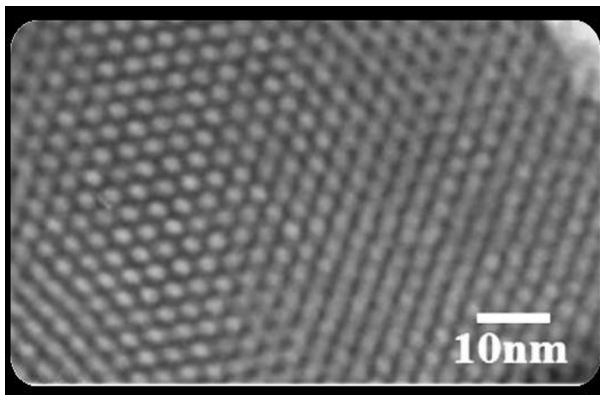
Table 1. Properties of neat bitumen

Marshall classification of bitumen	Softening point, °C	Penetration, 0.1 mm	Density, gr/cm <sup>3</sup>	Flash point, °C	Ductility at 25 °C, cm	Mass loss, %	Brookfield viscosity at 135 °C, Pa·s
85–100	48.6	96	1.015	294	100 <	0.07	0.336

Fig. 2 presents its morphological structure, as scanned by scanning electron microscopy (SEM). Fig. 3 depicts its structure of pores as scanned by transmission electron microscopy (TEM). Table 2 reports the properties of this modifier. As can be seen, the LUS-1 has regular lines of channels and pores made of SiO<sub>2</sub> units with a fibrous morphological structure.



**Figure 2.** Morphological structure (SEM) of LUS-1



**Figure 3.** Hexagonal configuration (TEM) of LUS-1

Table 2. Properties of LUS-1 (Bonnevot et al., 2003)

Line formula	Appearance	Structure	Specific area, m <sup>2</sup> /g	Pore diameter, nm	Pore volume, cm <sup>3</sup> /g	Density, g/cm <sup>3</sup>	Supportable pressure, t/cm <sup>2</sup>
SiO <sub>2</sub>	White Powder	Hexagonal	800–1300	3	0.841	0.09	5

## 1.2. Methods

Studies have suggested that the behaviour of asphalt binders in the MSCR test is different based on the type and weight percentage of modifier, and the test temperature (Delgadillo et al., 2006; Navarro et al., 2019; Shafabakhsh et al., 2019; Ghanoon & Tanzadeh, 2019). This is an experimental study, in which samples of experimental asphalt binders were prepared and tested at different weight percentages of modifier, and test temperatures by MSCR, through mixing base bitumen and LUS-1 nano-silica. The modifier content affects the level of binder performance. Specifically, the addition of 3 wt% of nano-silica caused the rise in high temperature performance but had no effect on the performance grade of asphalt binders in previous study, while the addition of 7 wt% led to the maximum high temperature performance of the asphalt binders up to two grades (Shafabakhsh et al., 2019). Based on previous studies, the maximum content of common nano-silica added to neat bitumen was 7 wt% of bitumen (Shafabakhsh et al., 2019; Ghanoon & Tanzadeh, 2019; Moeni et al., 2019; Villacorta & Nordbeck, 2019; Arshad et al., 2017). Thus, in this research, the maximum modifier content was limited to 8 wt% of the bitumen, given the parity of modifier increasing steps and the necessity of recognizing its peak effect on bitumen.

Accordingly, first, the base neat bitumen was heated to 170 °C, and then by separately adding LUS-1 nano-silica with 2, 4, 6 and 8 wt% of bitumen respectively, and 20-min vibration of the compound to maximize the separation of the nano-silica particles and to help more homogenous dispersion of them in the compound, the obtained mixture was mechanically mixed in a high shear homogenizer mixer at 4500 rpm for 30 min. Finally, considering the primary base neat bitumen and four other asphalt binders, five types of asphalt binder were produced, with MSCR test at high temperature being performed on them after aging process in rolling thin film oven test (RTFOT). Because the asphalt binders were liquid, a small droplet of each of them was spread on a microscope slide as a thin layer and then they were imaged by scanning electron microscopy. The reason for using droplets of binder and spreading them as a thin layer on the microscope slide was that suction of liquid binders in vacuum conditions would not damage the SEM device. Finally, the five samples of asphalt binders were imaged with appropriate magnification through SEM. These images were compared to evaluate the dispersion quality of nanostructured particles in four samples with different modifier contents. The dispersive quality of mesoporous nanostructured particles of silica reflects their homogeneous distribution in bitumen and their agglomeration.



In this research, MSCR test was conducted according to ASTM D: 7405 standard. First, for 10 cycles of 10 s at the stress level of 0.1 kPa, followed by immediately another 10 cycles of 10 s at the stress level of 3.2 kPa. During the 10 s cycles, the specimens were loaded at a constant creep load of mentioned stress for 1 s duration creep and followed by a zero-stress recovery of 9 s duration (ASTM D7405, 2015). In the experimental process of the MSCR test, there was an obvious relationship between the recovery percentage ( $R$ ), indicating delay elasticity of binders, and the strain percentage in the creep section of the test and  $J_{nr}$  representing the stiffness of the binder. It normalizes the binder response to the applied stress and clearly indicates the difference between various modified asphalt binders. Also,  $J_{nr,diff}$  denotes the asphalt binder sensitivity to the value of increased stress and, in some cases, may indicate the efficiency of the network formed by the modifier inside bitumen against augmented stress. The maximum non-recoverable creep compliance is limited by the standard performance grade of asphalt binders to the values of 4, 2, 1 and 0.5 respectively, for standard (S), heavy (H), very heavy (V), and extreme (E) traffic loads (AASHTO M 332, 2019).

In this study, given that the high temperature performance of the five binders produced based on the results of the strategic highway research program (SHRP) rheological tests was already determined, the MSCR test for each of the binders was performed from the lowest high temperature performance among the five binders to the high temperature performance of that binder itself, in order to save costs according to Table 3. An additional test was performed at 70 °C for binders containing 6 and 8% LUS-1 to allow for comparing some indicators and trends by achieving at least three points for three binders. In SHRP method, the design temperatures used to select the grade and type of bitumen are not air temperatures but pavement temperatures. Also, the Superpave system determines the high temperature of the pavement design at a depth of 20 mm below the pavement surface and

**Table 3. MSCR test temperatures for different asphalt binders**

<b>LUS-1, wt%</b>	<b>High temperature performance grade, °C</b>	<b>Experiment temperature at MSCR test, °C</b>
0.0	58	58
2	64	58, 64
4	70	58, 64, 70
6	64	58, 64, 70
8	64	58, 64, 70



uses it to select asphalt binder. Therefore, in some areas with low latitudes, where the air temperature reaches 50 °C in summer, it is not far from the mind that the temperature of asphalt pavement at a depth of 20 mm can even reach 64 °C and 70 °C. However, this is not the only reason for performing the MSCR test at 64 °C and 70 °C in this study. Considering that the high temperature performance of neat bitumen used in this study was 58 °C, in order to ensure the effectiveness of the modifier against rutting, an attempt was made to perform the MSCR test in at least two performance grades more than 58 °C.

## 2. Results and discussion

### 2.1. The effect of temperature

Figs. 4 and 5 demonstrate the effect of the modifier content and temperature levels on the elastic recovery and non-recoverable creep compliance, respectively. It is observed that upon temperature rise whether at low or high stress levels, the non-recoverable creep compliance increased, while the elastic recovery decreased. The maximum value of non-recoverable creep compliance and the minimum value of elastic recovery occurred at the temperature level of 70 °C. On the other hand, the lowest value of non-recoverable creep compliance and the highest value of elasticity occurred at 58 °C. As expected, at the temperature level of 58 °C, the neat bitumen has the highest  $J_{nr}$  and lowest  $R$  value, whereas all modified binders exhibit lower  $J_{nr}$  and higher  $R$  values. The modified binders represent lower non-recoverable creep compliance values at lower temperatures in comparison with the higher ones at the same dosages. Since the non-recoverable creep compliance is defined as the ratio of the residual strain in an asphalt binder (after a creep and recovery cycle) to the applied stress, a lower value of compliance describes higher deformation resistance. Experimental results indicate that the resistance to deformation of asphalt binder can effectively be improved by LUS-1 due to more reduction of non-recoverable creep compliance value. This beneficial effect could be related to the specific porous and fibrous structure of LUS-1, which provides the ability of coupling and binding to the asphalt binder matrix leading to enhanced mechanical and rheological properties of asphalt binder.

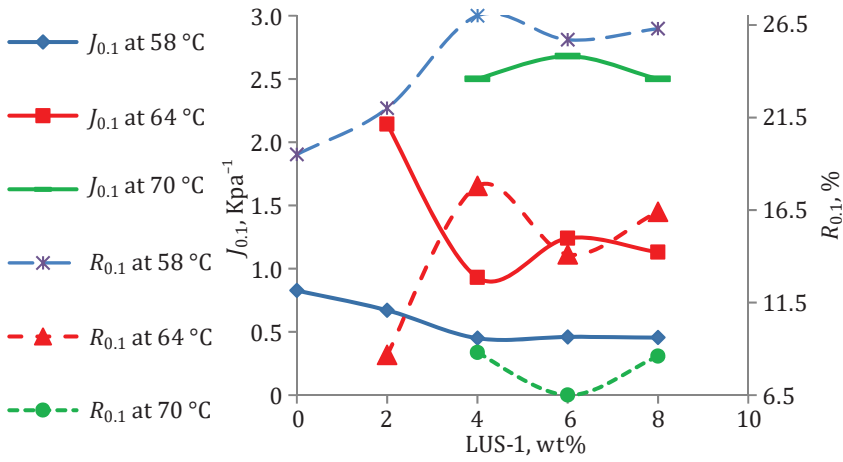


Figure 4. Effect of modifier content and temperature levels on  $J_{0.1}$  and  $R_{0.1}$

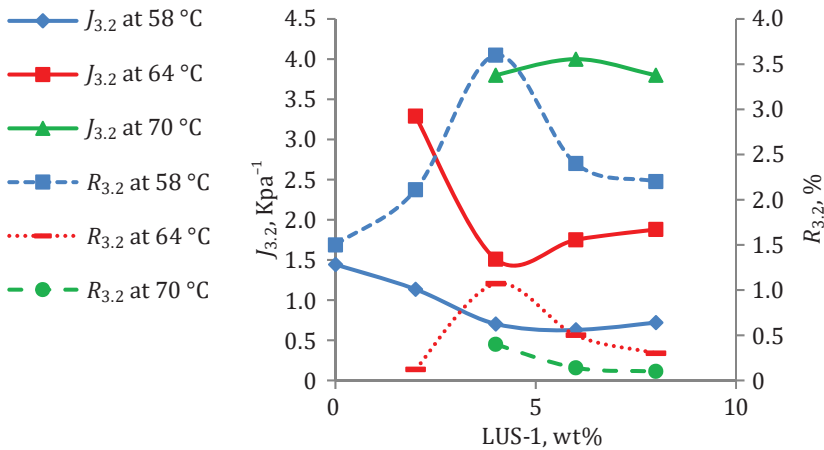


Figure 5. Effect of modifier content and temperature levels on  $J_{3.2}$  and  $R_{3.2}$

▲ Neat bitumen + 4 wt% LUS-1    ◆ Neat bitumen + 6 wt% LUS-1    ● Neat bitumen + 8 wt% LUS-1

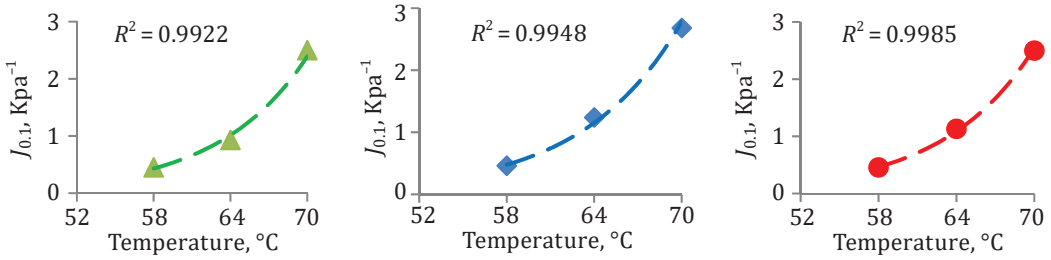


Figure 6. The correlation between temperature and  $J_{0,1}$

▲ Neat bitumen + 4 wt% LUS-1    ◆ Neat bitumen + 6 wt% LUS-1    ● Neat bitumen + 8 wt% LUS-1

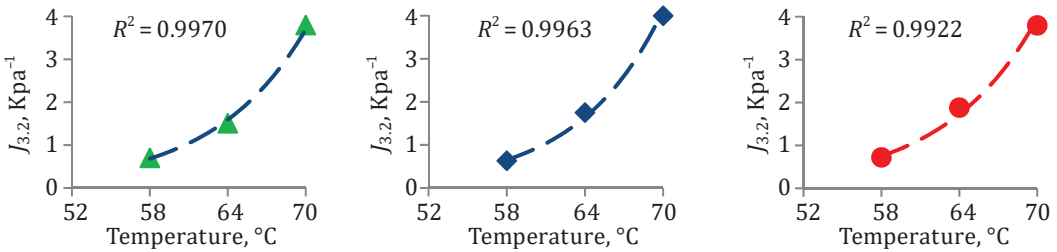


Figure 7. The correlation between temperature and  $J_{3,2}$

▲ Neat bitumen + 4 wt% LUS-1    ◆ Neat bitumen + 6 wt% LUS-1    ● Neat bitumen + 8 wt% LUS-1

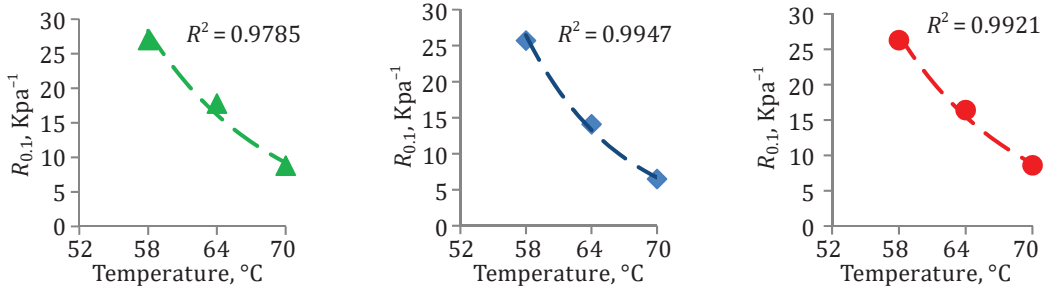


Figure 8. The correlation between temperature and  $R_{0,1}$

▲ Neat bitumen + 4 wt% LUS-1    ◆ Neat bitumen + 6 wt% LUS-1    ● Neat bitumen + 8 wt% LUS-1

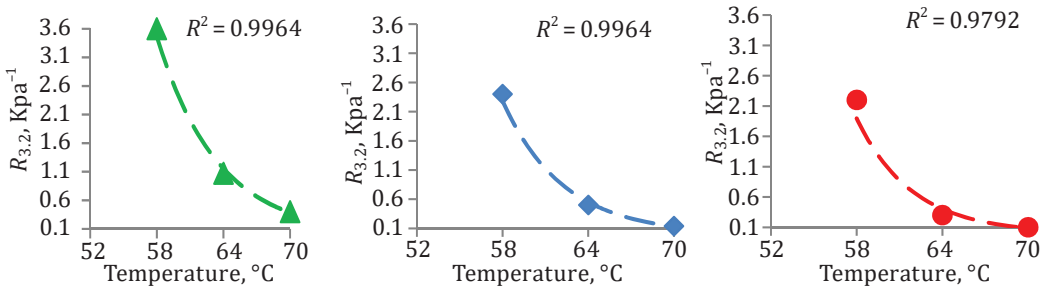


Figure 9. The correlation between temperature and  $R_{3,2}$

Figs. 6–9 illustrate a very good correlation with an exponential function between behavioural parameters of the MSCR test asphalt binders,  $J_{nr}$ ,  $R$ , and temperature.

Table 4. Effect of stress and temperature levels on traffic-adjusted binder grade

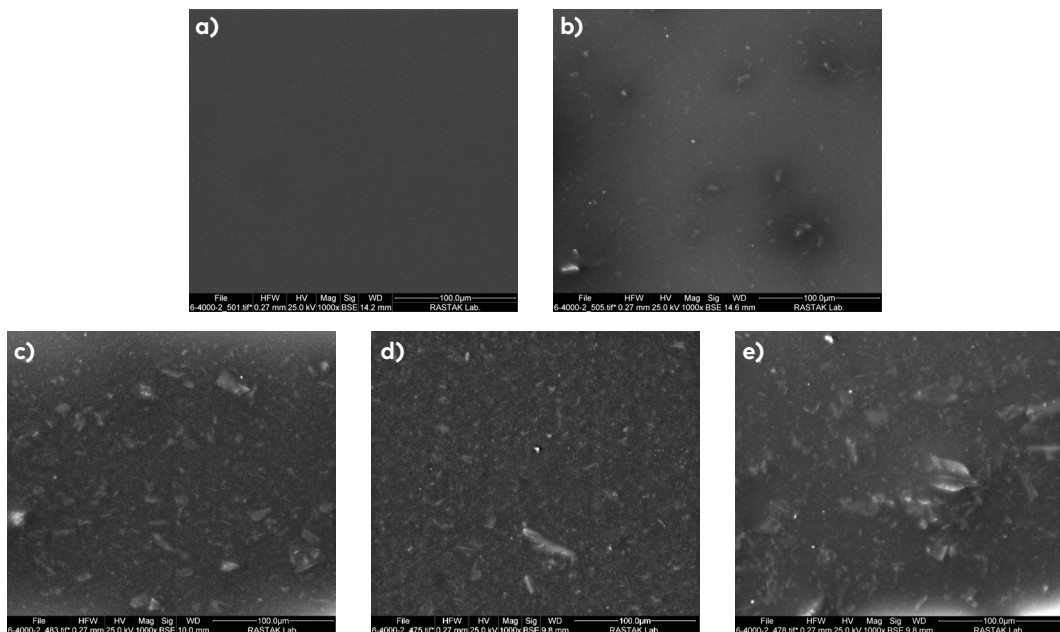
LUS-1 Content, wt%	Test temperature					
	58 °C		64 °C		70 °C	
	Stress levels		Stress levels		Stress levels	
	0.1 kPa	3.2 kPa	0.1 kPa	3.2 kPa	0.1 kPa	3.2 kPa
0.0	V	H	–	–	–	–
2	V	H	S	S	–	–
4	E	V	V	H	S	S
6	E	V	H	H	S	S
8	E	V	H	H	S	S

According to Table 4, the rise in temperature also decreased the traffic grade of the binders, such that this reduction was the one grade for all binders at the stress level of 3.2 kPa. This decline was more severe at the stress level of 0.1 kPa and for some binders it reached two grades. This suggests, as the stress level diminished, the effect of temperature on changing the traffic grade of the binders increased.

## 2.2. The effect of modifier content

According to Figs. 4 and 5, it can be observed that at a constant temperature level (58 °C), the neat bitumen has the highest  $J_{nr}$  and lowest  $R$  value, whereas all modified binders exhibit lower  $J_{nr}$  and higher  $R$  values. This means that with increasing in LUS-1 content, an increase in the stiffness of modified binders occurred due to the enhanced consistency which could result in a more decrease in the ratio of the residual strain in modified asphalt binders to the applied stress in repetitive creep test. The results indicate that modified binders show lower  $J_{nr}$  values than neat bitumen. This could have resulted from a more intense reinforcing effect of LUS-1 arising from its fibrous and nano-sized porous structure which could lead to a more binding efficiency in a network like manner throughout the binder. Figs. 4 and 5 also show that at all triple temperatures and dual stress levels in this study the highest value of  $R$  and the lowest value of  $J_{nr}$  relate to asphalt binder containing 4% of LUS-1 nano-particles.

According to Fig. 10, the fair dispersion of LUS-1 nanostructured particles in the asphalt binders containing 2, 4 and 6 wt% of nanostructured particles can be seen. It seems that small LUS-1 nanostructured particles, with a very low weight and size, may bind the coarse bitumen molecules with inter-particle bonding forces and cause an increase in creep stiffness and shear stiffness of asphalt binders at high temperatures. The density of LUS-1 chains that connect two infinite parts of the bitumen network is defined as cross-linking density, which is proportional to the stiffness of the asphalt binder. Thus, the LUS-1 chains may increase the cross-linking density of bitumen. In the binder containing 8 wt% of the modifier, the heterogeneous dispersion of LUS-1 nanostructured particles can be seen. This heterogeneous dispersion may be due to the existence of higher amounts of nano-modifiers compared to other binders, which has led to the formation of agglomerates in this asphalt binder. Coarse agglomerates may reduce the bonding force between the nanostructured particles and decreasing the specific surface area and their activity. This ultimately can reduce the efficacy of modifier in this binder.

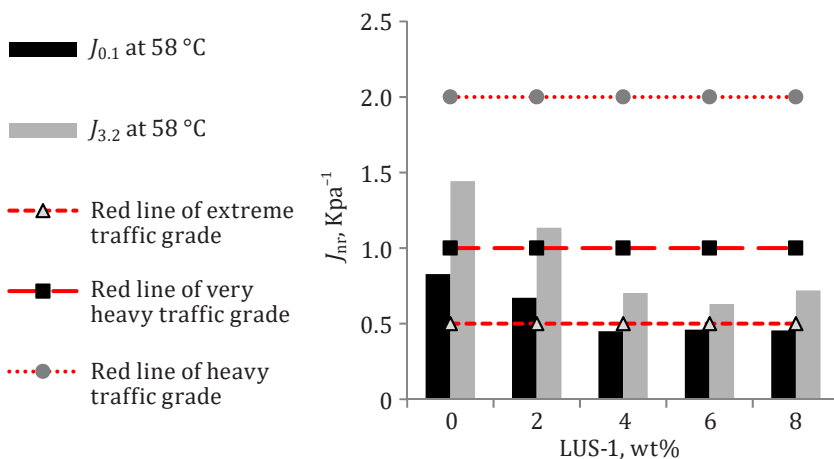


**Figure 10.** SEM image of asphalt binders: a) Neat bitumen, b) Neat bitumen + 2 wt%LUS-1, c) Neat bitumen + 4 wt%LUS-1, d) Neat bitumen + 6 wt%LUS-1, e) Neat bitumen + 8 wt %LUS-1

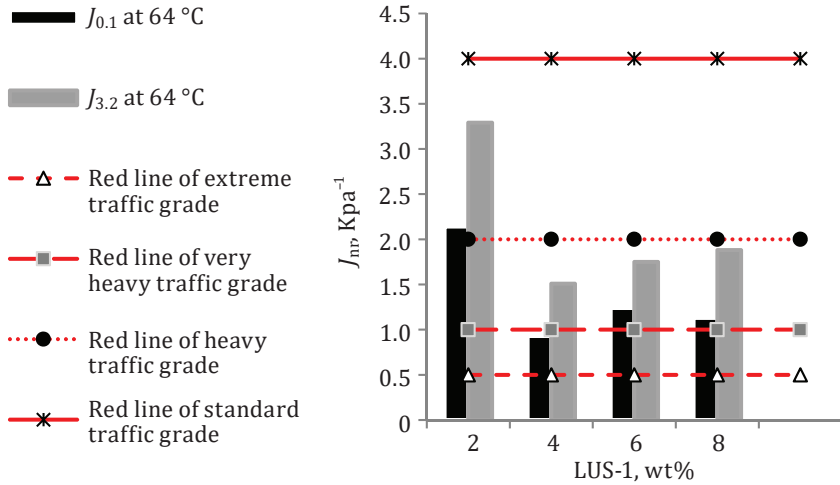
The LUS-1 nano-modifier also improved the bitumen delay elasticity by helping create an integrated network among the bitumen components, as observed by maximization of  $R$  in 4 wt%. Based on Fig. 14, the addition of research modifier did not lead to an absolute descending trend in the binder sensitivity to stress increase and it was fluctuating. The minimum stress sensitivity occurred in binders containing 6 wt% of the modifier. On the other hand, the binders containing 4 wt% of the modifier did not have any problem and showed an appropriate distance with the plotted red line. Elevation of the modifier content according to Table 4 increased the bitumen traffic grade, especially at temperatures of 58 °C and 64 °C. However, it was ineffective at 70 °C, suggesting the reduced effect of the modifier content on the traffic grade improvement of the binders at high temperatures.

### 2.3. The effect of stress level

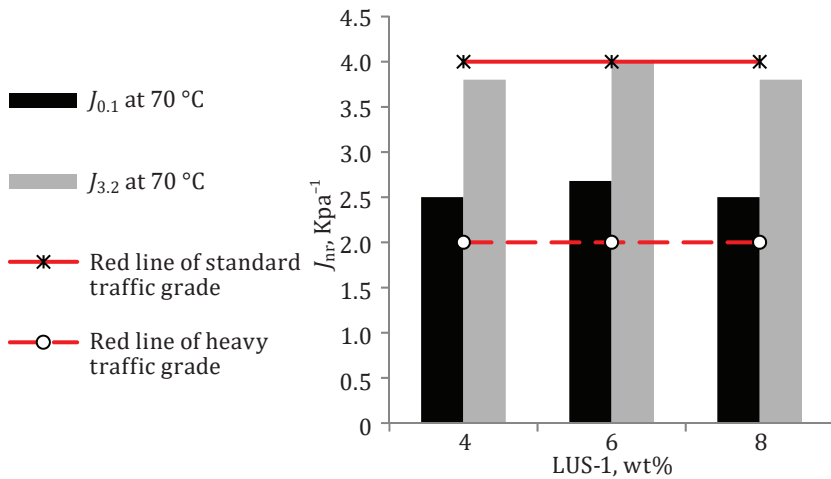
Figs. 11–13 depict the effect of stress level on the non-recoverable creep compliance of asphalt binders at different temperature levels. As can be seen, upon elevation of the stress level applied to the asphalt binders from 0.1 kPa to 3.2 kPa, the creep compliance increased at all temperature levels; in other words, the resistance to rutting decreased. By 3100% increase in the stress level, the percentage of rise in the value of non-recoverable creep compliance was fluctuating among the five binders within the [37%, 74.5%] interval, with the minimum interval being related to the binder containing 6% additive and the maximum to neat bitumen, both of which were recorded at 58 °C.



**Figure 11.** Effect of stress levels on non-recoverable creep compliance of asphalt binders at 58 °C



**Figure 12.** Effect of stress levels on non-recoverable creep compliance of asphalt binders at 64 °C



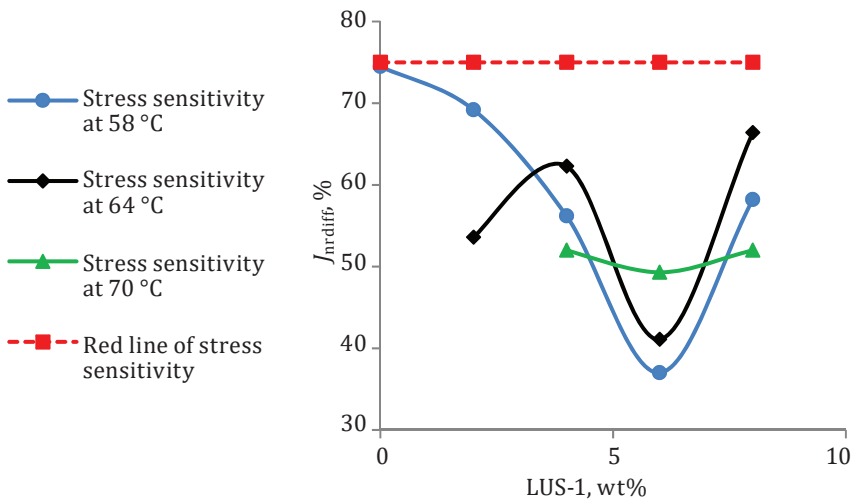
**Figure 13.** Effect of stress levels on non-recoverable creep compliance of asphalt binders at 70 °C



According to Fig. 14, it is observed that the increase in temperature has different effects on the stress sensitivity in asphalt binders, but their stress sensitivity did not exceed 75 % as the permitted value specified by AASHTO M332. Since the  $J_{nr\text{diff}}$  is calculated by Eq (1), the results of  $J_{nr-0.1}$  and  $J_{nr-3.2}$  in asphalt binder modified with 6 wt% of LUS-1 reveal that the decreasing changes in  $J_{nr-3.2}$  are more dominant than  $J_{nr-0.1}$ , so that its  $J_{nr\text{diff}}$  is the lowest value among all binders. This means that with an increase in stress level, the asphalt binder modified with 6 wt% had the best performance at all temperature levels due to the enhanced consistency, which could result in a more decrease in the ratio of the residual strain in modified asphalt binders to the applied stress in repetitive creep test at the higher stress level of 3.2 kPa, comparing to that at the lower stress level of 0.1 kPa. As can be seen from Fig. 6, in terms of stress sensitivity index, the modified binder with 4 wt% of LUS-1 is ranked after the binder containing 6 wt% of LUS-1.

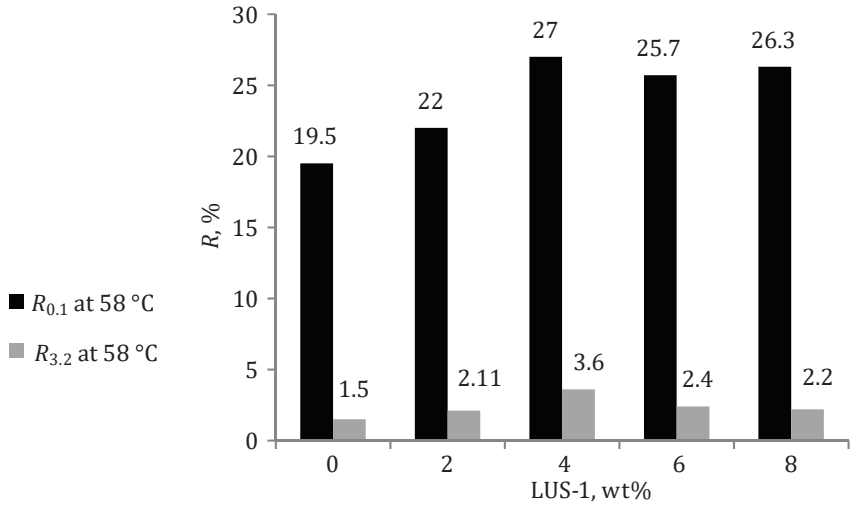
$$J_{nr\text{diff}} = \frac{J_{nr-3.2} - J_{nr-0.1}}{J_{nr-0.1}} (100) \quad (1)$$

The data in Table 4 reveal that elevation of the stress level applied to the asphalt binders was effective in changing their traffic grade, such that it caused a grade reduction for all binders at 58 °C. At 64 °C, except for the binder containing 4% modifiers, it was ineffective in all other cases and at 70 °C, there was no change in the traffic grade of the binders. This demonstrates that the effect of stress level elevation on the traffic grade of the binders diminished upon temperature rise.

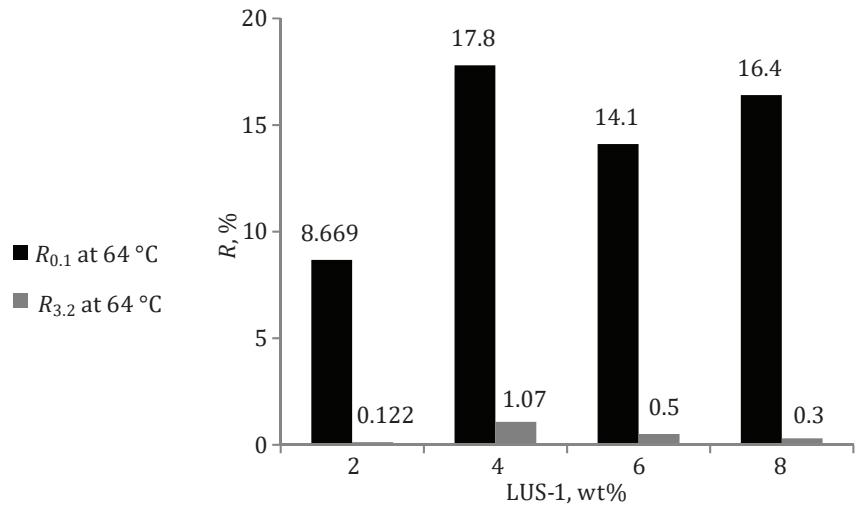


**Figure 14.** Effect of modifier content and levels of temperatures on stress sensitivity

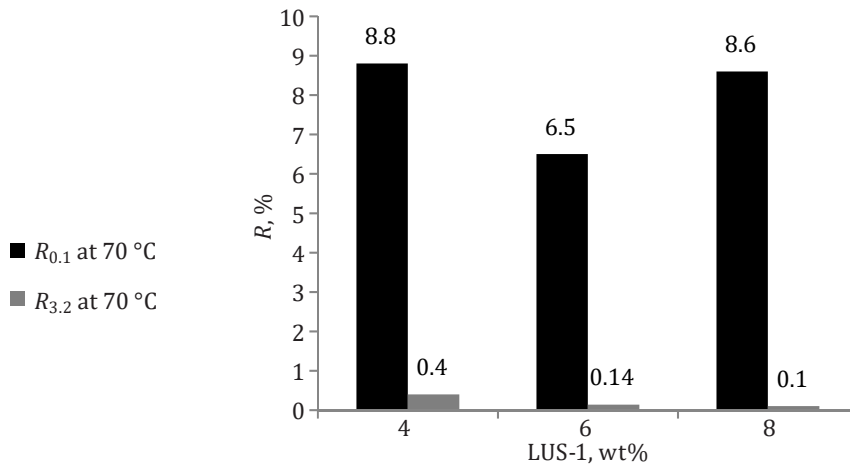
Figs. 15–17 indicate that the increase in stress level in the MSCR test was strongly effective on the delay elasticity of the asphalt binders. The percentage of this effect lied within the [86.7%, 98.8%] interval, which was significantly greater than the impact of increasing stress level on the value of non-recoverable creep compliance. The lower limit was related to the asphalt binder containing 4% additive at 58 °C, while the upper interval limit to the asphalt binder containing 8% additive at 70 °C.



**Figure 15.** Effect of stress levels on recovery percent of asphalt binders at 58 °C



**Figure 16.** Effect of stress levels on recovery percent of asphalt binders at 64 °C



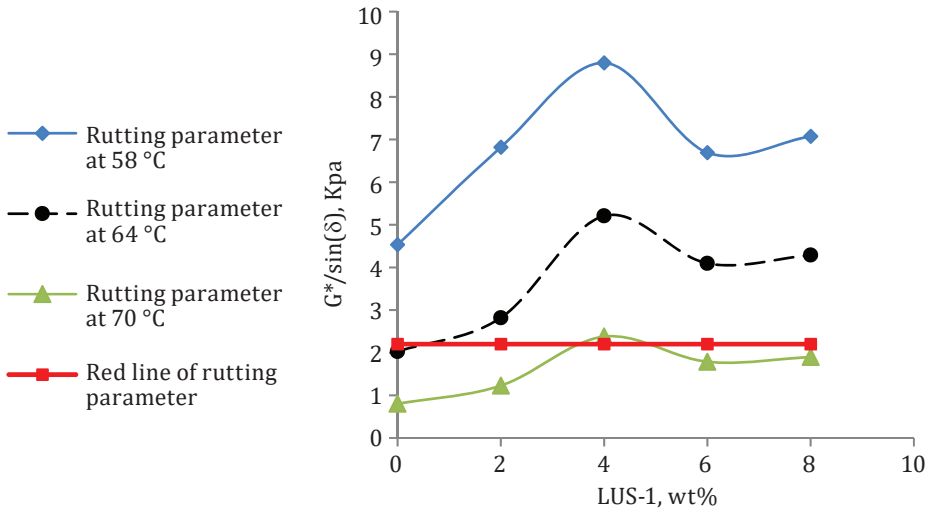
**Figure 17.** Effect of stress levels on recovery percent of asphalt binders at 70 °C

## 2.4. Comparing the results of complex modulus (DSR) tests and MSCR tests

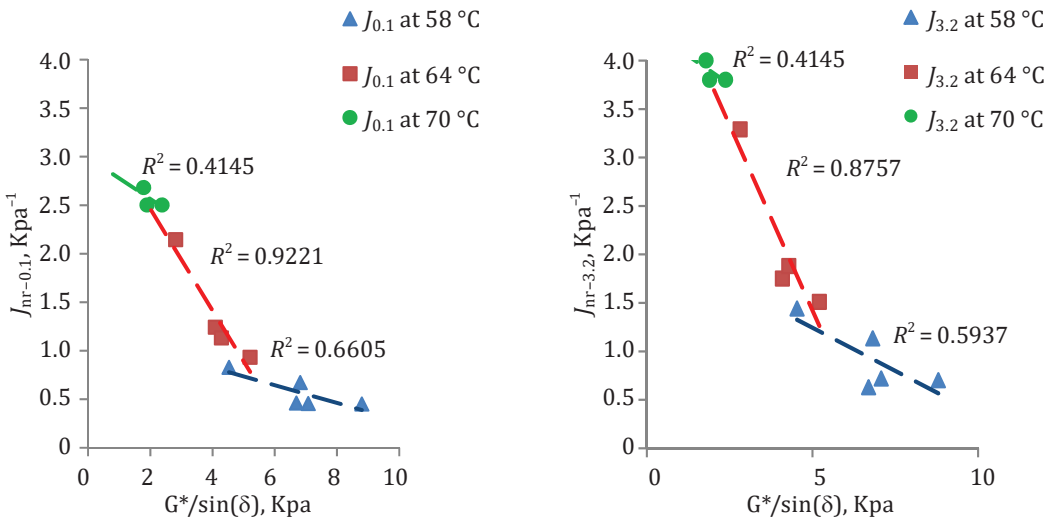
The comparison between Fig. 18 (part of the results of another study conducted by the authors of this article) and Figs. 4–5 indicates that the measured values of the rheological performance-related parameters concerning high temperature evaluations, including rutting parameter ( $G^*/\sin(\delta)$ ), non-recoverable creep compliance ( $J_{nr}$ ) and the recovery percent ( $R$ ), demonstrate that LUS-1 provides high modification effects on asphalt binder. This could be attributed to the nano-sized pore structure and extremely high specific surface area of LUS-1 particles and its fibrous morphology, which could greatly facilitate the formation of a network of nanostructured silica with a strong reinforcing effect throughout the binder leading to an improved resistance to permanent deformation of asphalt binder and enhancement of rutting resistance at higher in-service temperatures.

Figs. 4, 5 and 18 also show that the results of complex modulus (DSR) tests and MSCR tests are consistent, as the variation trends of  $R$  and  $G^*/\sin(\delta)$  are the same at different temperature levels. On the other hand, the variation trend of  $J_{nr}$  is opposite, and in both tests the maximum value of  $R$  and  $G^*/\sin(\delta)$  and the minimum value of  $J_{nr}$  occur in the 4% of bitumen modifier. This suggests that the greatest potential resistance to rutting is observed in the binder containing 4 wt% of LUS-1. Also, in both complex modulus tests and MSCR tests, the potential resistance to rutting decreases upon temperature level elevation. Figs. 19–20 also

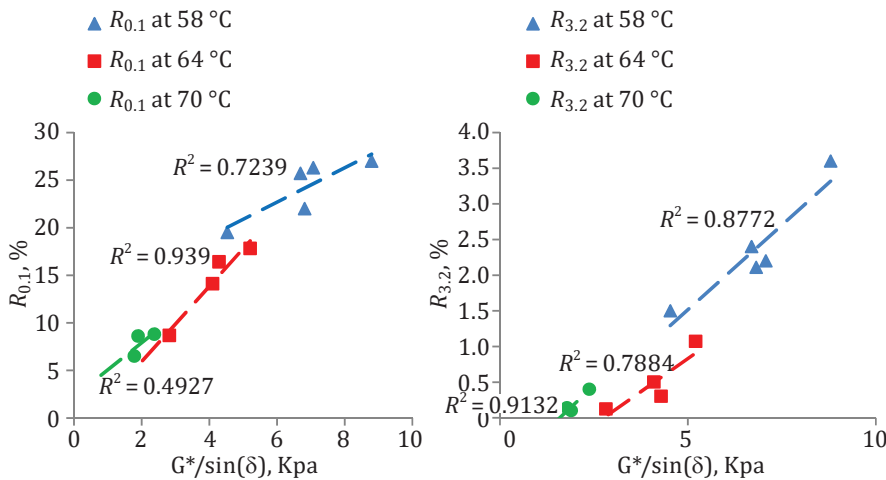
show a relative linear correlation between,  $G^*/\sin(\delta)$  as the rutting parameter in the complex modulus test along with  $R$  and  $J_{nr}$  as the MSCR test parameters.



**Figure 18.** Effect of modifier content and temperature on results of complex modulus test (aged at RTFOT)



**Figure 19.** Linear correlation between  $G^*/\sin(\delta)$  and  $J_{nr}$



**Figure 20.** Linear correlation between  $G^*/\sin(\delta)$  and recovery

## 2.5. The most effective of modifier content

According to Figs. 4, 5, 11–13, 15–17, it is observed that the binder modified with 4 wt% of MNS had the lowest non-recoverable creep compliance and the highest amount of elastic recovery among all the asphalt binders. Fig. 18 also confirms that this binder has the highest value of rutting parameter. Thus, all values of the rheological performance-related parameters concerning high temperature evaluations indicate that 4 wt% of MNS provides higher modification effects on asphalt binder than those established by the use of other contents of MNS. This could be attributed to the nano-sized pore structure and extremely high specific surface area of LUS-1 particles and its fibrous morphology, which could greatly facilitate the formation of a network of nanostructured silica with a strong reinforcing effect throughout the binder leading to an improved resistance to permanent deformation of asphalt binder and enhancement of rutting resistance at higher in-service temperatures.

## Conclusion

This study has been conducted in order to experimentally investigate the high temperature behaviour of asphalt binders modified with LUS-1 nanostructured particles based on the MSCR test. The following conclusions can be drawn:

Since all values of the rheological performance-related parameters concerning high temperature behaviour, including non-recoverable creep compliance ( $J_{nr}$ ), recovery percent ( $R$ ) and stress sensitivity ( $J_{nr\text{diff}}$ ), indicate that LUS-1 provides high modification effects on asphalt binder, the use of LUS-1 is recommended to improve bitumen resistance to rutting.

The binder modified with 4 wt% of LUS-1 has the lowest non-recoverable creep compliance and the highest amount of elastic recovery among all the asphalt binders. Thus, the use of 4 wt% of LUS-1 nanostructured particles leads to the greatest improvement in the high temperature behaviour of bitumen.

Elevating the levels of stress and temperature leads to diminished traffic grade and more viscous behaviour in asphalt binders modified with LUS-1.

The high temperature behaviour-related parameters, including non-recoverable creep compliance ( $J_{nr}$ ) and recovery percent ( $R$ ) obtained from MSCR test, agree with the rutting parameter ( $G^*/\sin(\delta)$ ) obtained from the DSR (complex modulus) test.

## Conflict of interest

The authors declare that they have not any competing financial, professional, or personal interests from other parties.

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