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Performance-Related and Rheological Characterisation of Natural Rubber Modified Bitumen

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Abstract:	<p>This paper is motivated by the limited research available evaluating the comprehensive effects of natural rubber bitumen modification (NR) for pavement applications. A detailed laboratory programme was carried out to evaluate the influence of NR on the high-intermediate-low temperature performance, including rheological characterisations and performance-related tests (fatigue cracking, thermal cracking, fracture and permanent deformation resistance). The main findings indicate some beneficial effects of NR on the performance-related response for fatigue and permanent deformation. Additional benefits were observed regarding the fracture strength and the thermal cracking resistance (ΔT_c). The viscoelastic effects of NR produced significant changes in the rheological properties of the base bitumen. Overall, the use of NR at a concentration of up to 7% by weight of bitumen proved to be a reasonable option for use in pavement applications, having more substantial benefits in tropical regions in agreement with its predominant effects on the bitumen modification at intermediate-to-high temperature conditions.</p>

September 8th 2020

Respected
Dr. Dimitrios Aggelis
Editor
Construction and Building Materials, ELSEVIER

Dear Dr. Aggelis:

After the comments raised by the reviewers, I kindly resubmit for evaluation the updated version of the scientific article entitled: "Performance-Related and Rheological Characterisation of Natural Rubber Modified Bitumen" by Jarurat Wititanapanit, Juan S. Carvajal and Gordon Airey. Likewise, through this communication I would like to inform you that this article is original in nature and that it has not been and will not be presented to any other journal or academic event. In the same way I allow myself to confirm that the authors of this article assume collective responsibility for the information contained in the manuscript.

The following is the information of the authors of the article:

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It's worth mentioning that both of my affiliations (University of Nottingham (UK), and Universidad del Norte (Colombia)) should appear on the manuscript for all stages of review and for publication if the manuscript is accepted.

I appreciate your kind attention to this letter and the attached manuscript.

Sincerely,



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Nottingham, April 9th 2020

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Performance-Related and Rheological Characterisation of Natural Rubber Modified Bitumen

Abstract

This paper is motivated by the limited research available evaluating the comprehensive effects of natural rubber bitumen modification (NR) for pavement applications. A detailed laboratory programme was carried out to evaluate the influence of NR on the high-intermediate-low temperature performance, including rheological characterisations and performance-related tests (fatigue cracking, thermal cracking, fracture and permanent deformation resistance). The main findings indicate some beneficial effects of NR on the performance-related response for fatigue and permanent deformation. Additional benefits were observed regarding the fracture strength and the thermal cracking resistance (ΔT_c). The viscoelastic effects of NR produced significant changes in the rheological properties of the base bitumen. Overall, the use of NR at a concentration of up to 7% by weight of bitumen proved to be a reasonable option for use in pavement applications, having more substantial benefits in tropical regions in agreement with its predominant effects on the bitumen modification at intermediate-to-high temperature conditions.

Keywords: Natural Rubber; Bitumen Modification; Fatigue; Rutting; Fracture; Performance; Rheology.

1. Introduction

Natural rubber (NR) is a material produced from native trees (*Hevea brasiliensis*) in South Asia, which is commonly used as a manufactured product either solely or combined with other materials in various industries (e.g., automobile, textile, insulating membranes, adhesives, footwear and construction). NR is widely used as a consequence of its high stretch ratio (i.e., high ductile response or elasticity), its ability to withstand external loading (i.e., permanent deformation resistance) and the high water resistance. In spite of being extensively produced and commercialised from the early 70's, the current bulging stocks of NR in Southeast Asian countries and the trends of decreasing trade price are gradually encouraging governments and environmental agencies to sought for alternative applications in road infrastructure projects [1, 2]. Similarly, the pressing environmental needs for recycling demands the selection of environmentally friendly alternatives as surrogates of finite natural resources, which supposes the incorporation of waste materials such as recycled tyre rubber [3-6] for road infrastructure applications,. At this respect, from the late 80's to the early 90's, research studies have assessed the effects of waste rubber (typically reclaimed from tyres) on bituminous mixtures incorporating multiple approaches and techniques [7-9]. For instance, [10] provided practical guidelines for the design, construction and maintenance of tyre-rubber modified mixtures. More recently, [5] presented a comprehensive

1 literature review of the various benefits associated with the use of recycled tyre-rubber (RTR) with a critical
2 analysis of the existing technologies and specifications that apply to the production process (i.e., production,
3 handling and storage). Nevertheless, most investigations of using rubber for road infrastructure applications
4 relate to RTR effects on bituminous mixtures [11-14] but only limited information is available on the virgin
5 rubber (NR) in terms of its multi-scale effects on bituminous mixtures. For instance, [15] studied the effects of
6 Natural Rubber on Modified Blends (NRMB) based on penetration, softening point and toughness, which
7 substantiated significant improvement of the permanent deformation resistance. Similarly, [16] concluded that
8 NR improves the durability of polymer modified asphalt (PMA) mixtures, and this effect is more prominent
9 when combined with peroxy formic acid to produce epoxidised natural rubber.

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19 The need to attain further experimental evidence that aids understanding the effects of NR on bitumen
20 modification and performance of bituminous mixtures supports this research study. Subsequently, this paper
21 aims to investigate the influence exerted by NR on the rheological properties and performance-related
22 characteristics of NRMB (i.e., fatigue cracking, thermal cracking, fracture, and permanent deformation). The
23 paper incorporates a range of tests and analytical theories to study the performance of NR modified bitumen.
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27
28
29 The paper structure includes a detailed description of the materials and experimental techniques used for
30 assessing the NRMBs. The laboratory data is further analysed in agreement with relevant analytical approaches
31 and current scientific literature. The paper is completed with a list of the major findings, and recommendations
32 for further research.
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37 **2. Materials and Testing Methods**

38 **2.1. Experimental Programme**

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42 A comprehensive laboratory programme was carried out as shown in Figure 1. The natural rubber latex used for
43 bitumen modification is in liquid form and containing high ammonia (HA), with a chemical structure of cis-1, 4-
44 polyisoprene (also called isoprene) with a chemical formula of C_5H_8 with a molecular weight distribution of 10^5
45 – 10^7 g/mol. The main properties of the NR used include those presented in **Table 1**. A 40/60-penetration grade
46 bitumen was used as the base bitumen (referred to as “control” in the manuscript). NR contents of 0%, 3% and
47 7% by total weight of modified bitumen were used. The laboratory tests included in the experimental
48 programme are listed in Figure 1. Each tests result correspond to the average of three test replicates, with the
49 exception of the DENT test that was conducted on 2 replicates as per standard guidelines.
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Table 1. Natural rubber properties.

Parameters	Unit	Specification	Results
Total solids content (TSC)	% by mass	61.50 min.	61.80
Dry rubber content	% by mass	60.00 min.	60.21
pH value at 25.4°C	--	11 max.	10.48
Pottassium Hidroxiide Number (KOH)	% by mass	0.70 max.	0.69
Alkalinity (as ammonia on total weight)	% by mass	0.65-0.75	0.68
Non rubber content	% by mass	1.70 max.	1.59
Volatile fatty acid number	--	0.05 max.	0.0229
Mechanical stability time at 55% TSC	Sec.	1000 - 1200	1080
Specific gravity at 25°C	--	0.94	0.9448
Magnesium content	ppm	40 max.	36.75
Viscosity at 60% TSC, spindle no.1 at 60 rpm	cps	--	59.50
Coagulum content, 80 mesh	ppm	--	20

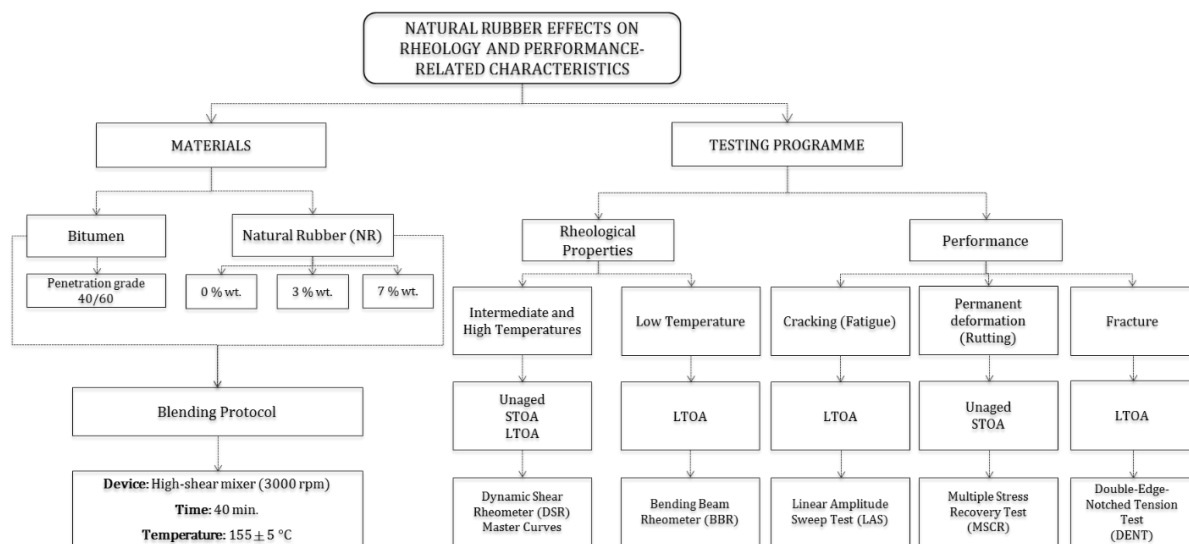


Figure 1. Testing programme for NRMB characterisation. **Note:** STOA refers to samples subjected to RTFO,

LTOA refers to samples subjected to RTFO + PAV.

2.2. Blending Protocol

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2 A high-shear mixing approach was followed to guarantee sufficient material dispersion and interaction time
3
4 between the base bitumen and NR. For this purpose, a high-shear mixer with double-head was used. The
5
6 procedure for NRMB manufacture included the following steps: (1) The base bitumen (1500g) was preheated in
7
8 an oven at 150°C for 120 minutes. Simultaneously, NR was weighed for mixing at the desired concentration. (2)
9
10 The base bitumen was extracted from the oven and transferred to a hotplate with the high-shear mixer setup and
11
12 the ability to keep the temperature at 150°C. (3) The required amount of NR (45g and 105g for 3% and 7%
13
14 blend, respectively) were gradually added to the neat bitumen in order to minimize formation of lumps and air
15
16 bubbles (due to the presence of water-base liquid) and thereby maximize blending efficiency. (4) The bitumen
17
18 was manually stirred with a glass rod during NR addition until a fairly homogeneous blend was obtained with
19
20 minimal formation of air bubbles. (5) Once the blend reached the target temperature on the hotplate (150 °C),
21
22 checked periodically with a hand thermometer, mechanical blending commenced from low speed (800 rpm) up
23
24 to a maximum of 2,400 rpm (for 3% NR content) and 4,200 rpm (for 7 % NR content).
25

2.3. Linear Viscoelastic Rheological Characterisation

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27
28 Strain-controlled frequency sweep tests were conducted using a Bohlin Gemini® Dynamic Shear Rheometer
29
30 (DSR) to determine the viscoelastic properties of the base bitumen and to assess the level of modification
31
32 induced by NR. For this, parallel-plate geometry was used (8 mm and 25 mm) with 2 mm and 1 mm gap,
33
34 respectively. The bitumen was heated up in the oven at 150 °C and then poured into the 8 mm and 25 mm
35
36 moulds and samples were mounted in the DSR after the samples had cooled down and could be removed from
37
38 the moulds. Low to intermediate temperatures (5 °C to 35 °C) were assessed with the 8 mm geometry, whereas
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40 high temperatures (35 °C to 75 °C) used the 25 mm geometry. Frequencies from 0.1 Hz to 10 Hz were used with
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42 a strain level of 0.5% (within the Linear Viscoelastic Region (LVE), based on amplitude sweep tests). The time-
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44 temperature superposition principle (TTSP) was applied for shifting of the rheological data (i.e., $|G^*|$ and δ) to
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46 produce smooth master curves. The shifting of the data was done using a 25 °C reference temperature with the
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48 application of the Williams-Landel-Ferry (WLF) model. In addition to the $|G^*|$ and δ master curves, a
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50 comparison of these parameters was made through Black Space Diagrams due to its simplicity as it does not
51
52 require shifting of rheological data. These rheological parameters were used to study the relationship between
53
54 stiffness and viscoelastic properties of the base bitumen and NRMBs [17].
55

2.4. Performance-Related Tests

1 A DSR was used to characterise the rheological and performance properties of the base bitumen and NRMB
2 blends. In addition to standard oscillatory linear viscoelastic (LVE) frequency sweep tests, the DSR was also
3 used to study the fatigue and permanent deformation performance-related characteristics of the materials.
4 Fatigue cracking was assessed through the Linear Amplitude Sweep (LAS) test and permanent deformation
5 (rutting) by means of the Multiple Stress Creep Recovery (MSCR) test. The fracture characteristics of the
6 materials were determined using the Double-Edged Notched Tension (DENT) test. These are considered
7 performance-related tests due to their ability to correlate material response to those of real pavement structures
8 subjected to various loading regimes [18]. The detailed description of these tests is presented in subsequent
9 sections.

18 **2.4.1. Linear Amplitude Sweep (LAS)**

20 The LAS test (AASHTO TP101) [19] was used to study the fatigue resistance of the base bitumen and NRMBs
21 as a function of ageing and NR content. The test method is intended for accelerated damage and involves cyclic
22 loading of bitumen samples that are subjected to increasing strain amplitudes. This leads to damage
23 accumulation, which is used to assess fatigue performance in terms of the number of cycles required to failure
24 (i.e., a decrease of the initial $|G^*|$ value at the peak shear stress, as per [19]). Aged bitumen samples of 8 mm
25 diameter and 2 mm thickness are tested in a strain-controlled mode using the DSR parallel plate geometry. Two
26 test stages are conducted: (1) The initial stage is non-destructive and involves assessing the undamaged
27 rheological properties of the bitumen samples in terms of the complex modulus (G^*) and phase angle (δ). This
28 stage is done via a frequency sweep test (i.e., oscillatory loading from 0.2 Hz to 30 Hz at 0.1% strain
29 amplitude). (2) Subsequently, the bitumen sample is subjected to a rest period of 10s before the onset of the
30 secondary stage. This secondary stage is destructive in nature through applying increasing strain amplitudes
31 (from 1% to 30% strain) at a constant frequency of 10 Hz and is referred to as the continuous oscillatory strain
32 sweep test. In brief, the test parameters include: (1) 100 loading cycles at 0.1% strain to obtain undamaged
33 material response, and (2) 30 subsequent rounds of 100 cycles from 1%-30% strain, increasing linearly by 1%,
34 for a total of 3,100 cycles of loading [20].

35 Considering previous literature recommendations for minimizing the risk of delamination (i.e., poor
36 bitumen/plate bonding), caution was taken to select the testing temperature [21]. To control delamination issues,
37 the failure plane of the sample was checked at the end of the test to identify the typical torsional-fatigue failure
38 of each specimen (i.e., circumferential crack) [22]. The temperature for LAS test was 15 °C and 25 °C for all
39 specimens.

materials, based on previous literature studying fatigue at intermediate temperatures [23]. The selection of two test temperatures was considered to assess the effects of this factor on the materials' fatigue response. In this study, two strain levels were considered to take into account both "strong" and "weak" pavement structures. For a "strong" layer with thickness higher than 100 mm, a low strain ($\gamma = 2.5\%$) was selected. Similarly, for a "weak" layer with thickness lower than 100 mm, a higher strain ($\gamma = 5.0\%$) was considered [24].

Viscoelastic continuum damage (VECD) mechanics, adopting Schapery's theory, was used to simulate damage growth and thus predict fatigue life as a function of strain (Equation 3). It is based on the simplified classic fatigue law (Equation 1) with A and B being obtained from the mathematical relationship between strain (γ) and cycles to failure (N_f) as shown in Equation 2,

$$N_f = A(\gamma_{max})^B \quad \text{Equation 1}$$

$$A = \frac{f(D_f)^k}{k \left(\pi \frac{I_D}{|G^*|} C_1 C_2 \right)^\alpha} |G^*|^{-\alpha} \quad \text{Equation 2}$$

$$B = -2\alpha$$

$$N_f = \frac{f(D_f)^k}{k \left(\pi \frac{I_D}{|G^*|} C_1 C_2 \right)^\alpha} |G^*|^{-\alpha} (\gamma_{max})^{-2\alpha} \quad \text{Equation 3}$$

Where,

$$k = 1 + (1 - C_2)\alpha,$$

f is the loading frequency, Hz,

$|G^*|$ is the undamaged complex shear modulus,

γ_{max} is the maximum expected binder strain for a given pavement structure in %,

D_f is the damage accumulation at failure, defined as the $D(t)$ corresponding to the reduction in initial $|G^*|$ at the peak shear stress.

2.4.2. Double-Edged Notched Tension (DENT)

The fracture characteristics in the ductile state of bitumen/NRMBs were studied following the Canadian Standard (Test Method LS-299) [25]. The essential work of fracture required to generate new surfaces was studied through the DENT test, which was used to determine the ductile failure resistance under horizontal tensile load application, at a specified temperature (15°C). Initially, as per standard guidelines, a 15 °C test temperature was considered, but at this condition the samples exhibited a premature brittle-failure (i.e., sample breakage). For this reason, the selection of a higher testing temperature of 25 °C was required in order to

facilitate a more ductile material response. The fracture properties studied included: (1) The essential work of fracture (W_e), (2) plastic work of fracture (W_p), and (3) the approximated critical crack-tip opening displacement ($CTOD$). These properties were calculated to study the resistance of the bitumen/NRMBs to ductile fracture. The DENT test was conducted after conditioning of the samples in a water bath (i.e., the specimens were submerged 25 mm below water surface) at 25 °C for 180 minutes. The deformation load was applied at a controlled rate of 50 mm/min and it stopped when all the samples reached the ductile failure. During testing, data of the elongation length (in mm), and force (in Newtons) was recorded and subsequently analysed to determine the fracture parameters (i.e., W_e , W_p and $CTOD$).

The significance of the measured properties for the comparison of the materials' fracture performance relate to: (1) the total work of fracture (W_t), which refers to the area under the load versus load-line displacement curve, kJ (Equation 4); (2) the specific total work of fracture (w_t), related to each replicate sample tested, kJ/m² (Equation 5); (3) the specific essential work of fracture (w_e), the energy required to fracture or break the sample without plastic deformation away from the fracture zone, kJ/m²; (4) the specific plastic work of fracture (w_p), the non-essential work dissipated during the deformation of a volume of bitumen around the fracture zone, MJ/m³; (5) the geometric constant of the plastic zone (β), and (6) the critical tip opening displacement ($CTOD$), mm (Equation 6 and Equation 7).

$$W_t = \int_0^{t_f} P \times d \quad \text{Equation 4}$$

Where,

P is the load in Newtons,

d is the displacement in the test in m,

t_f is the time when the maximum stroke is reached or ductile failure, whichever is attained first.

$$w_t = \frac{(\text{average } W_t)}{(B \times l)} \quad \text{Equation 5}$$

Where,

B is the sample thickness in m, and

l is the ligament length (the space between the notches) in m.

$$COTD = \frac{w_e}{\sigma_n} \quad \text{Equation 6}$$

Where,

σ_n is the net section stress of specimen in N/m², calculated using

$$\sigma_n = \frac{P_{peak}}{(B \times l)} \quad \text{Equation 7}$$

Where,

P_{peak} is the average peak load of the specimen tested with the smallest ligament length (i.e., the average maximum load for the 5 mm ligament specimens).

Determination of w_e and βw_p was done either graphically by plotting w_t for the three ligament lengths or mathematically using the least squares fitting method. w_e refers to the specific essential work of fracture (i.e., w_t value when $l = 0.0$) and βw_p is the slope of the best fit straight line (for $w_t = w_e + \beta w_p l$).

2.4.3. Multiple Stress Creep Recovery (MSCR)

The MSCR test was used to determine the permanent deformation susceptibility of the base bitumen and NRMBs. This test was conducted on unaged and RTFO-aged materials to study their elastic recovery properties. A DSR parallel-plate geometry with spindle diameter of 25 mm and 1 mm testing gap was used to undertake the test, using a Bohlin CVO® equipment. Test temperature of 60°C was selected based upon typical ranges reported in literature for pavement surface temperature in Thailand (65-68 °C [26]; and 53-59 °C [27]). The test followed the AASHTO TP 70 Standard method [28]. In brief, the sample was loaded for 20 cycles of creep-recovery at 0.1 kPa stress levels and 10 cycles for the 3.2 kPa stress condition. First, a 1 second creep load is applied and subsequently released for 9 seconds. Data was recorded by the DSR equipment and subsequently analysed in terms of the % recovery (*R-Value*), the non-recoverable creep compliance (J_{nr}), and the stress sensitivity parameter ($J_{nr-diff}$). Equations as per standard were used to calculate the parameters for both stress conditions (0.1 kPa and 3.2 kPa). The *R-value* refers to the ability of bitumen to recover (i.e., dissipate stresses) after repeated loading and higher values reflect better material response in terms of permanent deformation. The J_{nr} is an indicator of permanent deformation resistance that for higher values indicates a higher susceptibility to rutting distress of a given bitumen. The $J_{nr-diff}$ is used to distinguish the stress sensitivity of the bitumen due to the change of low stress (0.1 kPa) to high stress (3.2 kPa) conditions. Higher values of this parameter indicate higher sensitivity to the change in the loading stress to which the bitumen is subjected to under specific temperature conditions.

2.5. Low-temperature Rheological Characterisation

The low-temperature rheological properties for the bitumen/NRMBs were studied by means of the Bending Beam Rheometer (BBR) in accordance with the AASHTO T313-12 Standard [29]. In brief, the test method

1 involved the application of a 980 mN load to the beam specimen during 60 seconds of loading using the three-
2 point-bending approach. During the loading period, the low-temperature creep stiffness (S) and relaxation
3 properties (m -values) were recorded and later analysed for assessing the temperature/concentration effects and
4 determination of critical low-temperature values ($T_c(S)$ and $T_c(m)$) and the differential of these values, Delta T_c
5 (ΔT_c). The measured stiffness was calculated using the loading, deflection values and geometrical features of
6 the beams manufactured by using the beam theory. The estimated stiffness was calculated from a mathematical
7 fitting of the data. The m -value refers to the slope of the relationship between the logarithmic of measured
8 stiffness and is obtained at the logarithm of time for the total of 240 seconds of loading. Test temperatures
9 included for the low-temperature characterisation included -18 °C and -24 °C.

10 ΔT_c values were calculated using Equation 8. This parameter is a measure of the closeness of the critical low
11 temperatures measured from the Stiffness and m -value with respect to their influence on the low temperature.
12 Previous research has suggested this parameter is valid for more accurate prediction of field performance of
13 bitumen (fatigue cracking response) as a result of its relation to bitumen ductility [30]. This research proposed
14 two limits for ΔT_c that refer to a cracking warning level and a limiting value of -2.5 °C and -5.0 °C. These
15 criteria were incorporated for the discussion of results. The critical low-temperature values ($T_c(S)$ and $T_c(m)$)
16 were determined as per standard. From these values, the higher temperature is selected as the critical one for a
17 given material [31].

$$18 \Delta T_c = T_c(\text{Stiffness}) - T_c(m - \text{value}) \quad \text{Equation 8}$$

19 Where: T_c is the critical temperature for each of the BBR parameters (S , m -value).

20 **2.6. Ageing Protocols**

21 To simulate ageing occurring during manufacturing and laying of bituminous mixtures, Short-Term Oven
22 Ageing (STOA) and Long-Term Oven Ageing (LTOA) protocols were conducted on the base bitumen and
23 NRMBs manufactured through the blending approach previously described. The STOA was carried out with the
24 Rolling Thin Film Oven (RTFO) test [32] and LTOA with the Pressure Ageing Vessel (PAV) [33].

25 The effects of NR on NRMBs are analysed in terms of: (i) rheological characteristics at low-intermediate-high
26 temperatures, and (ii) performance-related responses (fatigue performance, permanent deformation resistance
27 and fracture properties). The effects of short- and long-term oven ageing on the rheology and performance-
28 related characteristics of NRMBs are also quantified.

3. Rheological Changes Induced by NR

3.1. Intermediate-High Temperature Effects (Master Curves)

The effects of NR in terms of the rheological changes induced to the base bitumen at the three NR concentrations were assessed in terms of complex modulus (G^*), phase angle (δ) master curves and the Black Diagrams. For the sake of brevity, only the results of the unaged materials are shown taking into consideration that similar observations were attained for the short-term (RTFO) and long-term (RTFO + PAV) ageing conditions in terms of G^* and δ master curves and Black Space diagrams. The rheological results in Figure 2 show that the effects of NR on the rheological parameters G^* and δ are more pronounced at higher temperatures and lower loading frequencies as demonstrated in Figure 2a, and Figure 2b. These results indicate that NR produces NRMBs with an increased elastic response (i.e., lower phase angles) and increased stiffness (i.e., higher complex modulus) in comparison to the control bitumen, with more significant modification level reached for the maximum NR concentration (7% NRMB). At lower testing temperatures (i.e., higher loading frequencies) the complex modulus master curves show a decrease in the G^* values for increasing NR dosages as compared to those of the control bitumen. While the change of δ with temperature and frequency is generally consistent with that of the G^* master curves, the differences between the three binders are less evident at lower testing temperatures (higher loading frequencies).

The rheological data shown in the G^* and δ master curves is also plotted in the form of Black Space diagrams (Figure 2c) and used as an indicator of rheological changes induced by NR without the need to shift the data based on the time-temperature-superposition-principle (TTSP) [17]. The Black Space diagrams indicated a shift of the curves towards a more elastic behaviour (i.e., lower δ for both NRMBs) as the NR dosage increased, particularly for the higher 7% NR content. For the 3% NR NRMB the level of modification is restricted to the high temperature (i.e., low frequency) domain with the rheological behaviour at low temperatures (i.e., high frequencies) being similar to that of the control bitumen. The Black diagrams also indicated that increased NR content produced a reduction in G^* that substantiates the increased ductile response induced by NR on the binder rheology (discussed further for the low temperature BBR rheological results). Although not shown in Figure 2 for the sake of brevity, the effects of NR modification for the short- and long-term aged binders were consistent with those discussed for the unaged binders with the effect of ageing simply indicating an overall increase in G^* and a reduction of δ values for all the binders as ageing increases. Overall, the comparison of the rheological parameters presented in Figure 2 substantiated the ability of NR to produce a more elastic response for the NRMBs as compared to that of the control bitumen. These results suggest an improvement of the

permanent deformation resistance for increased NR content NRMBs due to their increased elastic response at high temperatures.

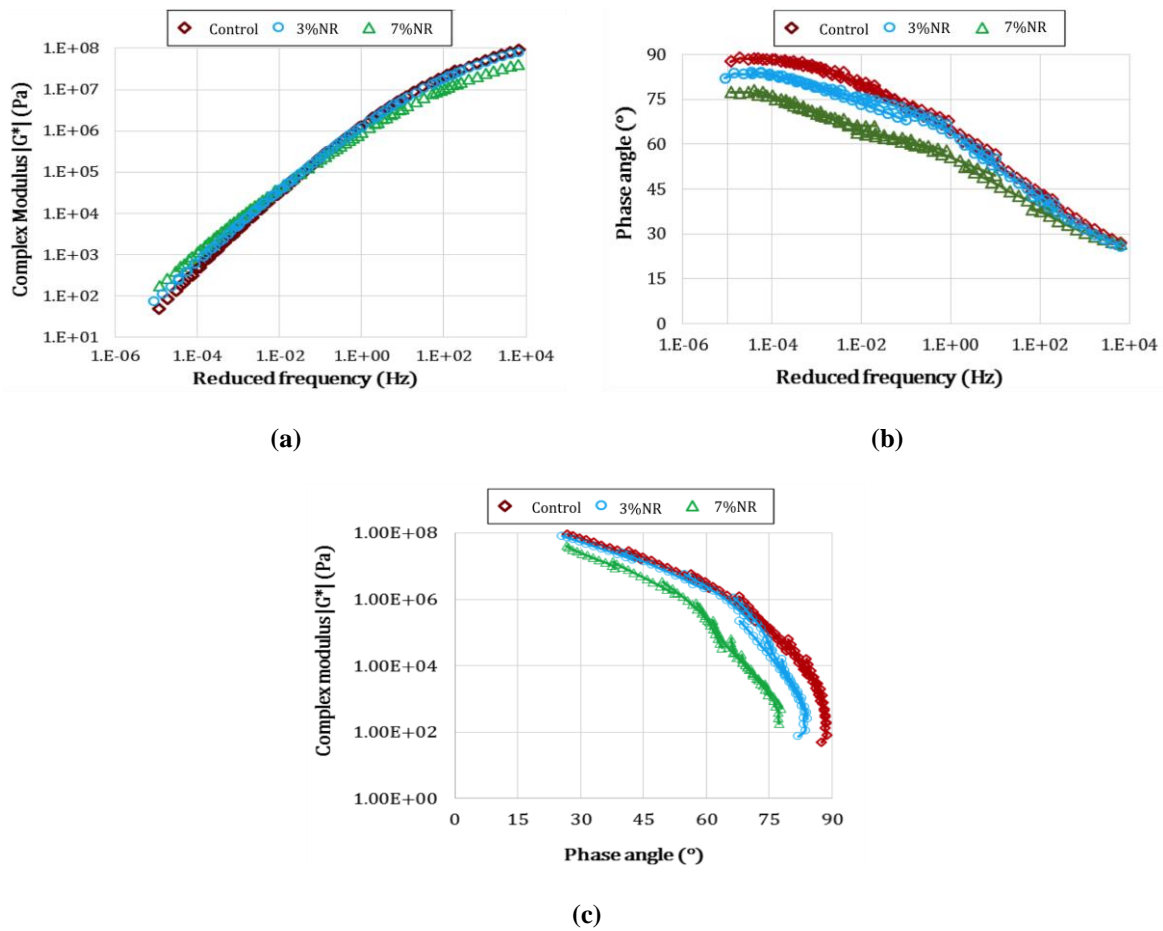
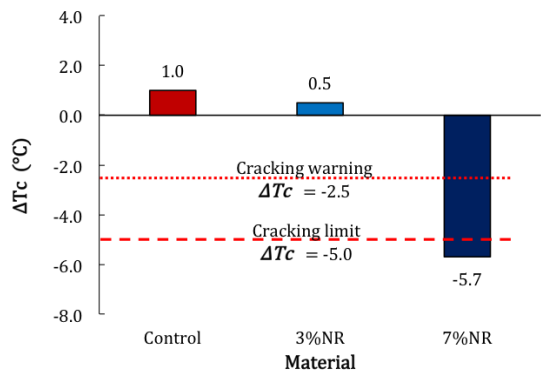


Figure 2. Comparison of the effects of NR on the rheological characteristics in terms of: (a) G^* master curves, (b) δ master curves, and (c) Black-Space diagrams for unaged materials.

3.2. Low-Temperature Effects and Thermal Cracking Susceptibility (BBR)

The S - and m -value comparisons for the two testing temperatures considered in this study are shown in Table 2. From these values, the effect of NR on the low-temperature rheological behaviour showed a reduction of S values (creep stiffness) as NR content increased from the control binder to 3% and 7% NR. However, the effect of NR on the relaxation properties (m -values) suggested a slower relaxation of the thermal stresses of the NRMBs considered in this study in contrast to that of the control bitumen. This means that high NR contents could induce brittle material response as a result of a reduced ability of these binders to stretch (i.e., lower ductility or loss in relaxation). However, the use of m -value does not represent a final indication of poor or good thermal cracking performance, as this parameter is associated with thermal stresses build up and the time required/available by the material to release these, which ultimately depends on the location and climate [34]. In

consequence, the fact that the increase in NR content induced lower m -values does not imply that these NRMBs would have detrimental performance as this will depend on the weather conditions to which the pavement will be subjected to and it also depends on the S values at low temperature, which were found to be lowered by the increase in NR content. As such, if one considers NR application in warmer climates, both S and m -values do not represent a major risk as these locations are not subject to major thermal stresses with lowest temperatures being typically around 15°C at which block cracking or thermal cracking are not distresses of concern. For applications of NR in low-temperature regions, both BBR parameters and ΔT_c need to be considered to find a balanced performance that satisfies the specifications imposed by the location/climate and the intrinsic material low-temperature response.



From

Figure 3 and Table 2 it is observed that the base bitumen and 3% NRMB satisfied both cracking limits for low-temperature for the ΔT_c criterion, whereas the increase of NR to 7% did not comply and it is therefore expected to have high susceptibility to thermal cracking. This is probably a consequence of the effects of high-polymeric modification of bitumen with NR, which could lead to higher susceptibility of NRMBs to thermal cracking at high concentrations, as previously reported by [35, 36]. In brief, from the rheological characterisation at low-temperatures it was found that: (i) NR induced positive effects on the NRMBs in terms of a reduction of the creep stiffness (S) at -18 °C and -24 °C, which is indicative of lower thermal stresses induced by both NR contents considered; (ii) Conversely, the relaxation properties (m -values) were negatively affected by increased NR content, which suggests a lower ability of the NRMBs to relax stresses (i.e., lower ductile response) in contrast to the control bitumen and; (iii) in terms of thermal cracking resistance, it was found that modification of control bitumen with NR content up to 3% would have satisfactory performance on the basis of the ΔT_c failure criteria.

Table 2. Low-temperature properties (S, m-value) and critical temperatures and the corresponding differential temperature ΔT_c .

Material	Stiffness		m-value		Critical Temperature Stiffness, S(c) (°C)	Critical Temperature m-value, m(c) (°C)	ΔT_c (°C)
	(MPa)						
	- 18 °C	- 24 °C	- 18 °C	- 24 °C			
Control	537	854	0.242	0.188	-10.497	-11.500	1.003
Control+3%NR	504	790	0.235	0.174	-11.103	-11.607	0.504
Control+7%NR	462	747	0.206	0.155	-12.608	-6.941	-5.667

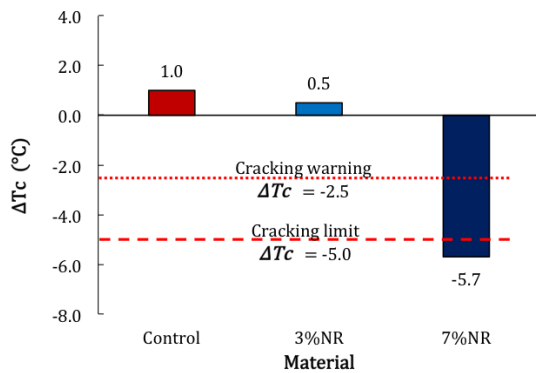


Figure 3. Comparison of the thermal cracking susceptibility for control bitumen and NRMB in terms of ΔT_c values.

4. Performance-Related Effects of NR

4.1. Effects of NR on Fatigue Performance

The characteristic VECD coefficients for the base bitumen (control) and NRMBs are presented in Table 3. The trends observed in the VECD coefficients indicate both the effects of temperature and NR content, which induced differences in the damage response that was captured via the C_0 , C_1 and C_2 coefficients. The results show an increase of C_1 was induced by the increase in NR content, whereas C_2 decreased for increased NR content. These differences substantiate the fact that 7% NRMB presents a higher reduction in initial $/G^*$ at low damage intensity levels, whereas the reduction is less prominent for higher levels of damage. Furthermore, this indicates that both NR contents induced positive effects on the damage resistance (captured by lower decrease in the initial $/G^*$ in comparison to the base bitumen) that turn into higher fatigue cracking resistance as further

discussed in this section. Overall, the assessment of the VECD damage coefficients indicated that the largest C_0 , C_1 and smallest C_2 values relate to the NRMB with 7% NR. The temperature effects of 10°C difference on the fatigue performance were also captured as reflected by the changes in the VECD coefficients, the damage intensity curves and the fatigue life curves. The results show that the higher 25 °C test and analysis produced significantly lower C_0 and C_1 values whereas C_2 remained almost unaltered. The damage intensity curves substantiated significant reduction in the stiffness of the materials as a result of the temperature increase and the fatigue life reflected this effect in higher N_f for the higher testing temperature. Overall, it was found that the addition of NR produced NRMBs with higher damage resistance in contrast to the base bitumen.

Table 3. VECD coefficients for control and NRMB at 15 °C and 25 °C.

Material	15 °C			25 °C		
	C_0 (MPa)	C_1 (MPa)	C_2	C_0 (MPa)	C_1 (MPa)	C_2
Control	44.51	0.092	0.434	13.2	0.084	0.424
Control + 3% NR	44.06	0.117	0.381	12.93	0.104	0.383
Control + 7% NR	46.52	0.149	0.344	16.81	0.179	0.299

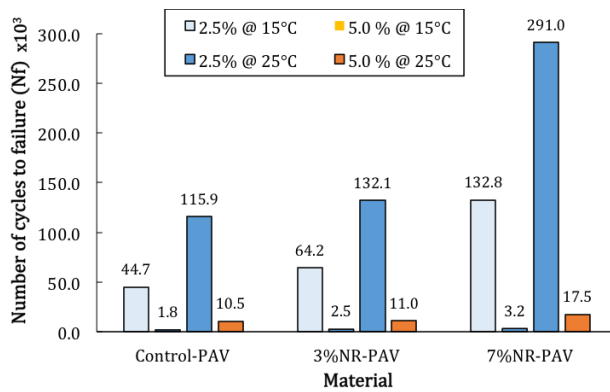


Figure 4. Comparison of fatigue lives for PAV-aged materials at 2.5% and 5% strain at 15 °C and 25 °C.

A comparison of the fatigue lives (number of cycles to reach failure, N_f) are shown in **Figure 4**. As observed previously, temperature and NR content effects were identified. For both temperatures, N_f increased as a function of the NR content, which was consistent for both the low and high strains considered ($\gamma = 2.5\%$ and $\gamma = 5.0\%$). At 15°C testing temperature and 2.5% strain, a 44% increase in fatigue life was found for 3% NR and a 197% increase for 7% NR in contrast to that of control bitumen. At 15 °C and 5% strain, this increase was less prominent (36% for 3%NR and 76% for 7% NR). The effects of increasing the test temperature by 10°C also produced differences in the fatigue lives, which were reflected by higher N_f values for the 25°C testing temperature for all materials and strains considered. From the macroscopic perspective (i.e., visual inspection of

the samples after testing), the fatigue of all the materials exhibited a failure plane that followed a *factory-roof* morphology and a circumferential torsional failure (i.e., growing from the inside to the outside of the bitumen/NRMBs specimens), which is consistent with previous findings by [22] and substantiates an adequate bitumen/plate bonding during testing.

4.2. Effects of NR on Permanent Deformation Resistance

The effects of NR on permanent deformation resistance is presented and discussed in this section in terms of the evolution of accumulated strain over time, the effects of ageing, and the comparison of rutting parameters (non-recoverable creep compliance J_{nr} , percent recovery R -value, stress sensitivity parameter $J_{nr-diff}$).

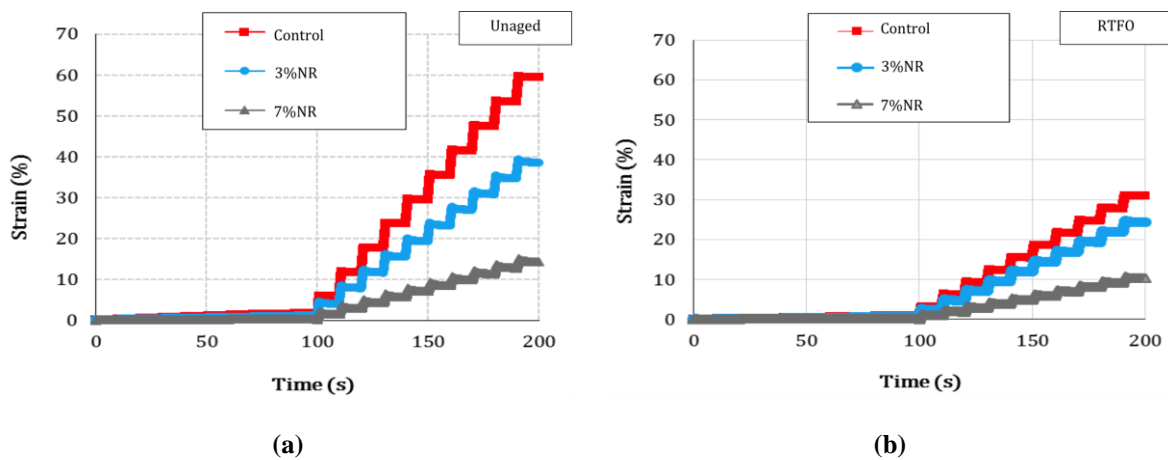


Figure 5. Strain versus time for (a) unaged bitumen, and (b) RTFO aged bitumen, at 60 °C.

The strain and accumulated strain versus time presented in Figure 5 and **Figure 6**, respectively, show a positive effect of NR on increased resistance to permanent deformation in contrast to the base bitumen. In Figure 5, the initial 100 seconds refer to the 0.1 kPa stress condition and the subsequent steps of cumulative strain are due to the impact of a much higher stress regime (3.2 kPa), which is easily observed and captures the effect of NR modification of the base bitumen. The higher NR contents induced a reduction in the accumulated strain for both NRMBs with 3% NR and 7% NR, which was observed for both unaged and STOA conditions and for both stress levels considered (0.1 kPa and 3.2 kPa). This reduction in accumulated strain highlights a positive influence exerted by NR on bitumen modification in terms of reduced permanent deformation susceptibility of the NRMBs in comparison to that of control bitumen (**Figure 6**).

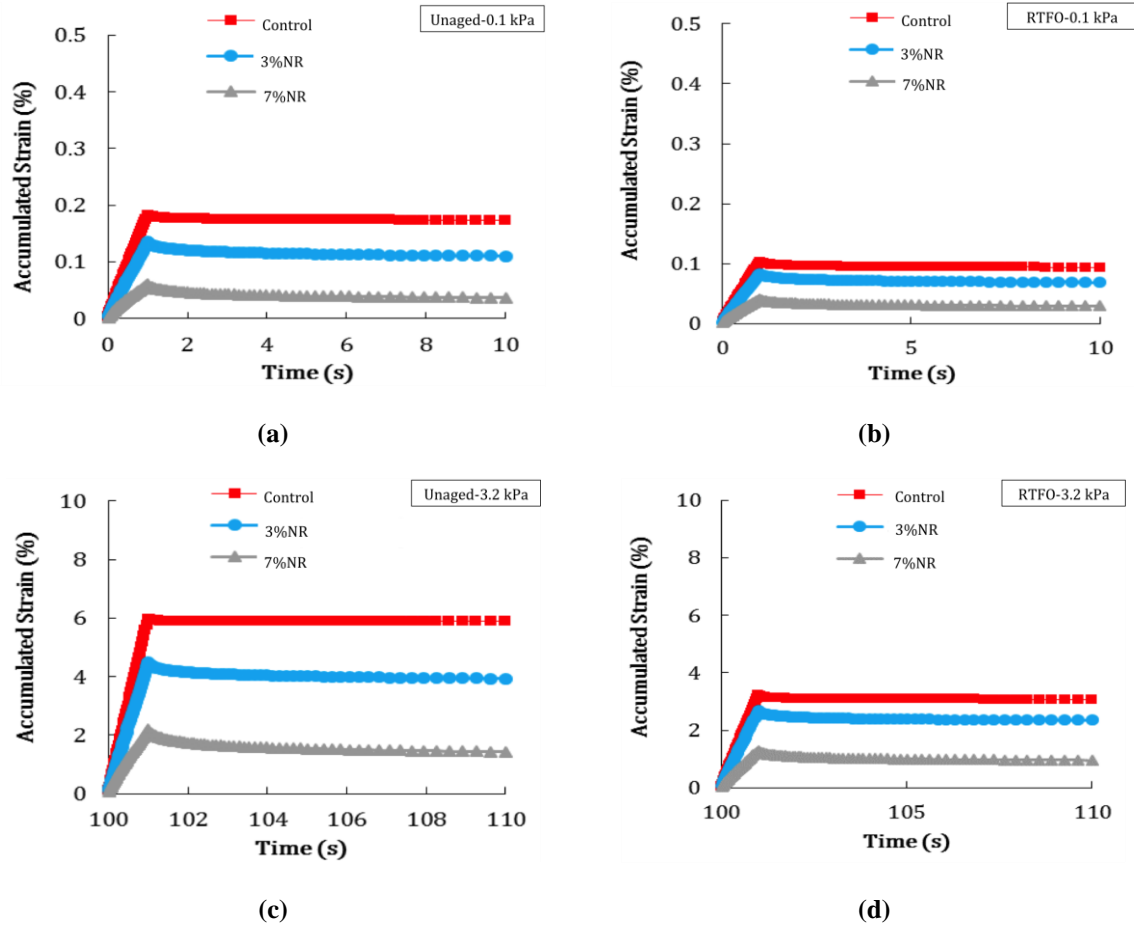


Figure 6. Effects of stress on the accumulated strain for unaged, and RTFO-aged bitumen, at 60 °C. (a) Unaged at 0.1 kPa, (b) RTFO at 0.1 kPa, (c) Unaged at 3.2 kPa, and (d) RTFO at 3.2 kPa.

The effects of NR on the resistance to permanent deformation, assessed in terms of the average percentage recovery (R -value) and non-recoverable creep compliance (J_{nr}), indicated that increasing NR content enhanced this material response as compared to that of the base bitumen. Consistently for both unaged and RTFO-aged materials and at both stress levels, it was observed that J_{nr} decreased and the R -values increased as the NR content increased. Figure 7 show the effects of STOA ageing, NR content and stress level on the non-recoverable creep compliance (J_{nr}). The change from unaged to STOA produced stiffer materials as captured by the lower J_{nr} values for both control bitumen and NRMBs at 3%NR and 7%NR content. The increase in stress level from 0.1 kPa to 3.2 kPa increased the value of J_{nr} by approximately 10%. Furthermore, the effect of increased NR content showed a clear trend of reduction in the J_{nr} values for both stress levels and ageing conditions, which substantiates the positive influence of NR on this material response.

These parameters provide evidence of the positive effects of NR on the permanent deformation resistance of the modified control bitumen. Similarly, the effects of ageing were also demonstrated based on the J_{nr} and R -values shown in Figure 8. Similar to the effect seen for unmodified binders, subjecting the NRMBs to STOA improves

the rutting resistance of the modified binders meaning that positive observations in terms of permanent deformation resistance held true for both NR content and ageing condition.

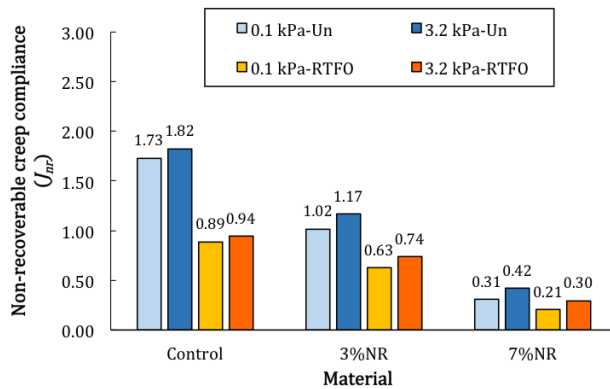


Figure 7. Comparison of the effect of NR and ageing on the permanent deformation, presented in terms of non-recoverable creep compliance (J_{nr}) for unaged materials, and RTFO aged materials.

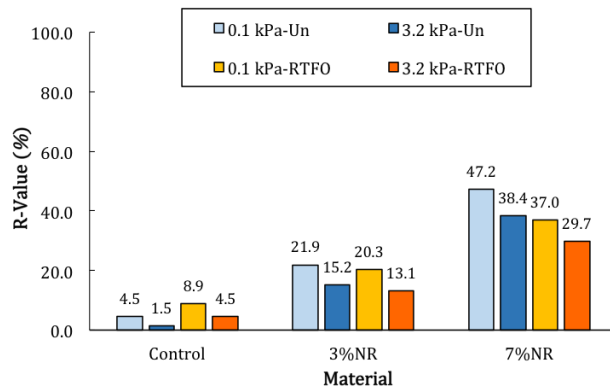


Figure 8. Comparison of the effect of NR and ageing on the permanent deformation, presented in terms of average percentage recovery (R -value) for unaged materials, and RTFO aged materials.

The J_{nr} results are in agreement with those observed for the R-values with higher R-values indicating a higher resistance to permanent deformation. As such, the combined effects of ageing, stress level and NR content are presented in Figure 8, where it becomes clear that ageing and NR content increased the permanent deformation resistance of the binders. Furthermore, as it would be expected for bituminous materials, the increase of stress level induced a substantial decrease of the R-values, indicating the stress sensitivity of the materials with increased NR content. The reduction in the percent recovery (%R) is explained by the possible damaging of the NR crosslink structure (i.e., polymer network stability) induced by this short-term ageing protocol, which inevitably affects the permanent deformation response of the NRMBs at both concentrations studied. Under this oxidative condition, the hardened bitumen phase is responsible for the most part of the rheological response.

Previous research found similar results of dropping recovery values when polymer-modified bitumen blends were subjected to artificial ageing cycles in both RTFO and PAV [35, 36]. Furthermore, the $J_{nr-diff}$ values shown in **Figure 9** substantiate the increase in the stress sensitivity induced by increased NR content for the bitumens considered. However, this is a common effect of high polymer modification of bitumen, which has been reported for the permanent deformation performance of polymer modified asphalt (PMA) mixtures [37, 38]. From these results, however, it is surprising that short-term ageing of the 7% NRMB did have a reduction of the stress sensitivity, which could result from possible errors in rheological data captured by DSR due to the high stiffness gain induced by the high-NR content in contrast to 3% NR or the base bitumen, or it might be related to variability between replicate materials during production of the DSR samples. Therefore, from $J_{nr-diff}$, it might be deduced that higher NR contents led to detrimental effects on the stress sensitivity for both ageing conditions, which needs to be considered for optimum dosage selection in conjunction with other performance-related characterisations to attain a balanced performance for a given NR content. However, based on standard recommendations [39], all the materials fulfilled the criteria as they were all below 75%, which suggests satisfactory stress sensitivity for both NR contents.

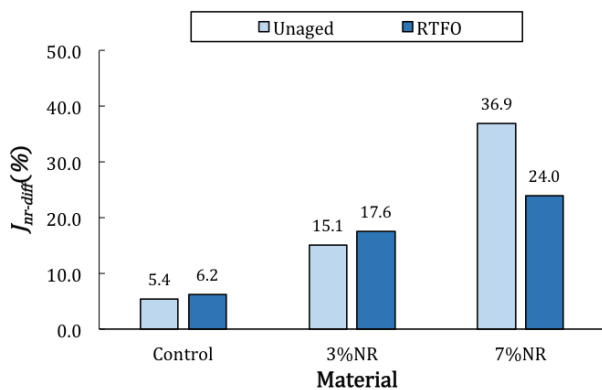


Figure 9. Stress sensitivity for unaged and RTFO-aged materials.

4.3. Influence of NR on the fracture characteristics

The specific total work of fracture (Wt), essential work of failure (We), plastic work of failure (βw_p), and critical crack opening displacement ($CTOD$) are presented in this section to discuss the effects of NR on the fracture properties of the modified binders. Prior to the fracture parameters, the plane-stress/plane-strain mixed mode of testing was verified through the correlation between net section stress and ligament length, which indicated a decrease in the net section stress as the ligament length increased. The Wt values for the base bitumen and NRMB at 3% and 7 % are shown in **Figure 10** where a higher slope can be observed for 7% NR content in

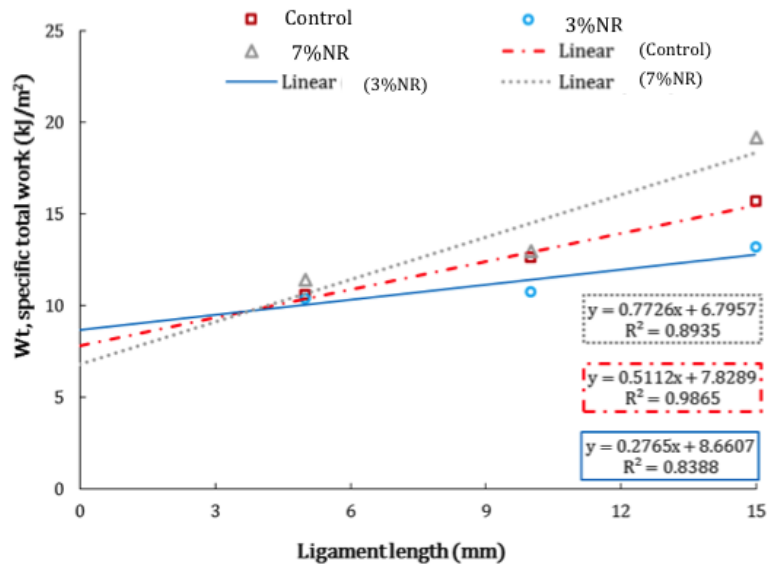
1 contrast to the base bitumen and 3% NR. These results indicate an improved yielding capacity and increase in
2 the fracture energy for NRMB with 7% NR content, whereas the 3% NR content did not produce the same
3 extent of influence on the fracture energy and therefore its yielding capacity is reduced [38]. As observed in
4 **Figure 10**, the essential work of fracture for 3% NR content exceeded that of the base bitumen, whereas 7% NR
5 content provided the lowest W_e values. This indicates that the 3% NR content enhanced the resistance to
6 fracture due to a higher ductile strain tolerance in contrast to those of the base bitumen and 7% NR. This finding
7 is consistent with previous research [39, 40]. The former study showed a positive correlation between higher W_t
8 values and β_{w_p} with enhanced fatigue cracking of real pavement sections due to enhanced ductile strain
9 tolerance. The latter study proved that inclusion of RAP to polymer modified bitumen led to unsatisfactory
10 fracture properties, which included decreased W_t and β_{w_p} values.

11 The plastic work of failure (β_{w_p} values related to the slopes of the curves) is shown in **Figure 10a** and this
12 parameter was used as an indicator of the fatigue cracking resistance of the base bitumen and NRMBs based on
13 recent literature [42]. It can be observed that for the 7% NR (0.7726), the W_t increases at a higher rate in contrast
14 to that of the base bitumen (0.5112) and 3% NR (0.2765), respectively, which would suggest 7% as the most
15 adequate NR concentration. However, other fracture properties are also required for confirming this observation,
16 thus $CTOD$ and W_e were also determined. The $CTOD$ values are in agreement with those of W_e (**Figure 10b**),
17 which substantiates the fact that NR contents above 3% might lead to deleterious effects on the fracture
18 resistance for the material combinations studied in this paper. This could be the result of NR concentrations
19 reaching a maximum level of yielding capacity and strain tolerance that is below the 7% studied in this paper.
20 This might be due to the physicochemical reaction of higher NR content with the base bitumen due to the higher
21 concentration of polymeric chains in the bitumen matrix that are able to alter the ductile response under high
22 strain concentrations at the testing temperatures considered. Similarly, the data presented in Table 4 is consistent
23 with the previous discussion of the fracture parameters in terms of the failure deformation and peak load. These
24 results show that a NR content of 3% leads to improved fracture resistance as a consequence of the lower peak
25 loads and increased deformation. The 3%NR content therefore induced better fracture properties for the
26 materials studied, whereas 7% NR exhibited detrimental effects on the fracture of NRMBs.

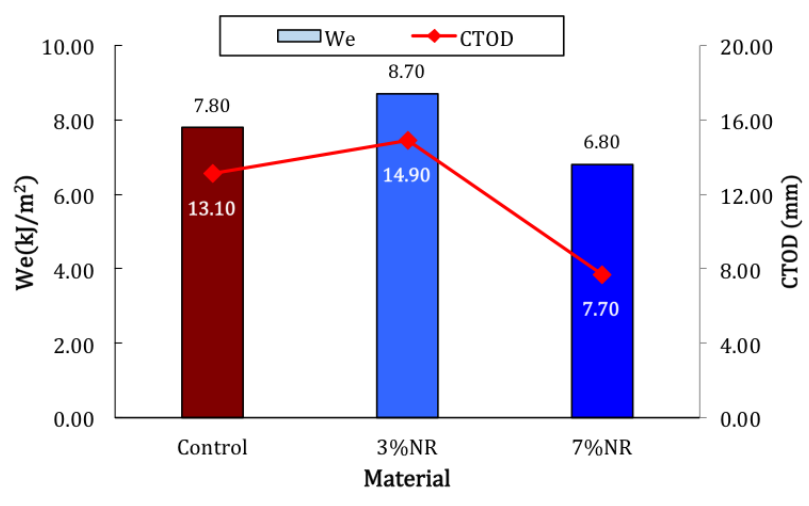
27 Furthermore, the results observed for the maximum concentration of NR in the NRMBs might be associated by
28 the detrimental side effects produced by the artificial ageing regimes (i.e., RTFO and PAV) that are required
29 prior testing for DENT. More precisely, as discussed before, the polymer network or cross-link of the NR
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1 molecules seems to be disintegrated by the short- and long-term ageing, which results in stress concentration in
2 the bitumen matrix. Previous research on fracture response of SBS-modified bitumen through DENT testing
3 found that high polymer concentrations (i.e. higher than 2%) tend to have poorer fracture performance in
4 comparison to reasonably low contents of modifiers. In fact, the use of WMA additives such as Fischer-Tropsch
5 (FT), chemical surfactants (CS) and water-based (WB) have the ability to modify the fracture response, which
6 tends to be concentration-dependent and showed improved fracture response at 2% concentration for CS WMA-
7 additive type, but this response was opposite for the FT and WB types with the only exception of 6% WB
8 concentration [43]. Additional research by [44] substantiated a detrimental ductile response of high-polymer
9 modified bitumens with SBS (i.e., 3-5% concentration by weight of base bitumen). In fact, it was found that
10 both in unaged and PAV-aged conditions, the CTOD values for the SBS-modified samples had lower ductile
11 performance (i.e., lower CTOD values), with one single exception to this trends for a SBS-blend (A5), which
12 had the same CTOD value, measured at deformation rate of 50 mm/min, as the base bitumen after long-term
13 ageing protocol. Additional research on PMB-bitumen modification through various concentrations of RAP
14 binder (i.e., 0%, 15%, 25%, and 40%) indicated that the higher concentrations of this bitumen modified
15 negatively affect the intermediate temperature response in terms of the ductile properties measured through
16 DENT and elastic recovery tests [45]. Conversely, [46] found that for low-SBS polymer concentrations the
17 fracture performance after PAV-ageing seems comparable and at some instances superior to that of base
18 bitumen for concentrations ranging from 1.2% to 1.8% by weight of base bitumen. It was also found that the
19 superior response at RTFO ageing conditions is not equaled by that of PAV-aged materials, indicating a
20 detrimental effect of artificial ageing on the fracture response for these material combinations.
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(a)



(b)

Figure 10. (a) The specific total work of fracture (W_t) and plastic work of failure (βw_p), (b) essential work of failure (W_e), and critical crack opening displacement (CTOD) for neat bitumen and NRMBs.

Table 4. Summary of peak load and deformation to failure for neat binder and NRMBs.

Material	DENT insert length (mm)					
	5		10		15	
	Peak Load (N)	Failure Deformation (mm)	Peak Load (N)	Failure Deformation (mm)	Peak Load (N)	Failure Deformation (mm)
Control	30	34.8	40	67.2	46	118
Control + 3% NR	29	47.2	37	80.6	42	134
Control + 7% NR	44	32	57	56.2	66	94

5. Conclusions

The laboratory investigation described in this paper assessed the effects of NR on the rheological and performance-related characteristics of modified bitumens (NRMB). This was achieved through a comprehensive laboratory programme incorporating the evaluation of NR effects on bitumen rheology and the evaluation of fatigue cracking, thermal cracking, permanent deformation and fracture properties of the NRMB. Two NRMB blends were manufactured with 3% and 7% NR by mass of binder using a 40/60-penetration grade base bitumen (control). The test methods included: (1) Dynamic Shear Rheometer (DSR) master curves and Black Space diagrams together with Bending Beam Rheometer (BBR) tests for rheological assessment over a wide temperature range; (2) Linear Amplitude Sweep (LAS) tests for assessing fatigue at intermediate temperatures; (3) Multiple Stress Creep Recovery (MSCR) tests for permanent deformation at high temperature; and (4) the evaluation of fracture properties at intermediate temperatures using the Double Edged-Notched Test (DENT) test. Based on the experimental results and analysis, the following main conclusions can be drawn:

- 1) The effects of NR on the bitumen rheology produced changes in the rheological parameters (G^* and δ) that were captured via the master curves and Black Diagrams. It was observed that increased NR content produced significant increase of the stiffness of NRMBs (higher G^*) that was particularly pronounced at high testing temperatures. Similarly, the decrease of δ values are consistent with the results of G^* and the Black Diagrams showed a shift of the NRMBs curves towards a zone related to more elastic response in comparison to the base control bitumen. Overall, these results indicated a substantial level of modification of the base bitumen that relates to more elastic NRMBs with higher resistance to permanent deformation as further substantiated with MSCR testing.

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- 2) The rheological characterisation at low-temperatures found that: (i) NR induced positive effects on the NRMBs in terms of a reduction of creep stiffness (S) at -18°C and -24°C , which is indicative of lower thermal stresses being induced in the NRMBs; (ii) The relaxation properties (m -values) were also affected by increased NR content although the m -values showed a lower ability of the NRMBs to relax stresses in contrast to the control bitumen and; (iii) In terms of thermal cracking resistance, it was found that a NR content of 3% would have satisfactory performance on the basis of the ΔT_c failure criterion. From a practical point of view, these results imply some restricted use of NR in cold regions or those with significant day/night temperature gradients and supports its use in tropical regions.
 - 3) The fatigue performance of the NRMB blends assessed by means of the LAS testing indicated that NR increased the fatigue life of the base bitumen by a significant extent at the intermediate temperatures considered (15°C and 25°C). This impact on fatigue life was consistent for both strain levels ($\gamma = 2.5\%$ for thin pavement structures and $\gamma = 5.0\%$ for thick pavement structures) but it was more prominent at the lower strain level. In general, the results showed a higher number of cycles to reach failure (N_f) for 7% NRMB as compared to those of control bitumen and 3% NRMB. These findings were consistent for the temperatures considered, which suggests positive effects of NR on bitumen modification with regard to fatigue cracking.
 - 4) Positive effects of NR on the permanent deformation resistance were observed indicating an increased resistance for both NR contents considered. The maximum resistance was reached for the 7%NR content as it related to the highest R -values and lowest J_{nr} parameter for all combinations studied. These results substantiated the positive influence of NR on the rutting response for the NRMBs when compared to that of base bitumen for both unaged and RTFO-aged materials. However, some detrimental effects on the polymer structure (crosslink of the NR molecules) of the short-term ageing regime did likely occur as observed by slightly reduced R -values for the RTFO-aged materials. The stress effects on the NRMBs resistance to rutting indicated that the improvement of this material response was predominant for higher stresses (3.2 kPa) in contrast to the lower stress considered (0.1 kPa). Nevertheless, the stress sensitivity ($J_{nr-diff}$) increased with NR addition to the base bitumen, reaching a maximum for 3% NR content for both ageing conditions, which is expected as a result of high-content polymeric modification of bitumen, such as that provided by Styrene-Butadiene-Styrene (SBS) in polymer modified mixtures (PMA).
 - 5) The NRMBs exhibited a limited fracture response as observed with the DENT test results in terms of the fracture properties of W_i , W_e , βw_p , and $CTOD$. In practical terms, based on the material combinations

1 considered, these results suggest that NR content should not exceed 3% when attempting to minimize
2 fatigue cracking and improve fracture performance at the micro scale. Similarly, other NR dosages should
3 be explored with different base bitumen types than those included in this investigation for further validation
4 of fracture properties.
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8 Overall, the evidence obtained through the experimental programme and materials tested substantiates some
9 performance-related benefits of NR and its suitability for applications in warmer countries, such as Thailand or
10 those encountered in South East Asia where NR is produced and widely available, for which fatigue and
11 permanent deformation of pavements represents the major challenges for mixture design, performance and
12 durability. Furthermore, a possible increase in the current limits of NR use for road infrastructure is envisioned
13 as a practical application of this research study, when complemented with additional future studies. Similarly,
14 these results encourage use of NR in other countries aiming at promoting sustainable road infrastructure.
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22 Some recommendations for future work include:
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- 24 • Although storage stability of the samples has not been studied by this research work, previous work by
25 [47] substantiated that NR contents of up to 6% exhibit stable performance for use at high
26 temperatures. Furthermore, [48] demonstrated with factual evidence that contents up to 9% can have
27 satisfactory storage stability and indicated that this property is largely dependent on the NR
28 concentration. Similar findings were reported by [49], who reported satisfactory storage stability for
29 blends subjected to conditioning for 72 hours at 163°C. Therefore, future studies might add value to the
30 characterization of NRMB by incorporating the storage stability results.
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- 39 • In terms of the complexity of bitumen modification through NR in comparison to other polymers, our
40 experience and that reported by previous research indicates [50, 51] the following: Overall, it is harder
41 to mix NR latex than other polymers because of its liquid nature in contrast to others being capsules or
42 grinded materials (solid-like). Furthermore, the fact that NRL contains “water” in the latex, and have
43 high ammonia content makes it hard to produce blending due to the physical effects (i.e., frothing and
44 foaming), in addition to the strong smell produced by the ammonia. For these reasons, the liquid NRL
45 is gradually added to the base bitumen at small portions, which allows for reducing the air bubbles and
46 evaporation. This is aided by manual stirring to minimize the foaming of the rubber to reach the top
47 head of the mixing machine. Certainly, this procedure means more time consumption than other
48 polymers but it allows for homogeneous blend if carried out cautiously by future researchers.
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September 8th 2020

Respected
Dr. Dimitrios Aggelis
Editor
Construction and Building Materials, ELSEVIER

Dear Dr. Aggelis:

Hereby I kindly submit the responses to the comments raised by the reviewers to the scientific article entitled: "Performance-Related and Rheological Characterisation of Natural Rubber Modified Bitumen" by Jarurat Wititanapanit, Juan S. Carvajal and Gordon Airey. Through this communication I would like to inform you that this article is original in nature and that it has not been and will not be presented to any other journal or academic event. In the same way I allow myself to confirm that the authors of this article assume collective responsibility for the information contained in the manuscript.

All the comments raised by the reviewers have been addressed accordingly and the responses provided on a point-by-point basis in this letter (answered in blue color). Similarly, the revised paper with track changes has been included as part of the documents in this submission.

Reviewer #1

Good paper on NR modified asphalt. Below are my personal comments for your reference.

1. The basic information on NR is missing. NR latex or NR powder? And the molecular weight?
Detailed information included in the updated manuscript with track changes on page 2 and 3.
2. The specific DSR equipment should be given in experimental sections. TA xx? Malvern xx? Anton Paar MCR
Dynamic Shear Rheometer (DSR) devices included a Bohlin Gemini® used for master curves, and LAS; a Bohlin CVO® used for master curves and MSCR testing. This is now included in the revised manuscript, pages 4 and 8.
3. P6 Line 15. As recommended by AASHTO T315, for low temperature DSR tests, 8 mm parallel plates should be firstly heated to 64 °C or upper temperature before the installation of asphalt sample. In this case, the adhesion between asphalt and plates can be guaranteed.
These guidelines were carefully followed for the experimental procedure to ensure proper bitumen-plate adhesion and restrict delamination.
4. P8 Line 47. Please pay attention to the latest AASHTO specification, 20 cycles are used for 0.1 kPa creep stress.
Corrected in test method description. Page 8.
5. As presented in your master curves, NR molecules does not seem to greatly crosslink to each other since one could not see the phase angle plateau region, which can be easily found in well dispersed SBS modified asphalt. Besides, the reviewer also suspects that the neat bitumen 40/60 could also be the reason. If softer bitumen was used, the NR molecular chains might have a better elastic behavior.
This is a very logical reasoning and we agree with the idea of softer bitumen providing improved crosslink that lead to better elastic behavior. However, the fact we used a harder bitumen inevitably produced the viscoelastic response evinced in the discussed results.
6. If possible, it would be better to include -6 and -12 °C BBR results to make the calculation of critical temperatures more precisely. As one can see, your S and m-value results at -18 and -24°C are over the critical values 300 MPa and 0.3.
Unfortunately, the only BBR data collected included those temperatures presented in the paper. However, these data allows for understanding the effects of NR at the low-testing temperatures and aids distinguishing whether the material is suitable for colder or warmer regions. We agree it would be quite interesting to study additional temperature for a more comprehensive comparison, but current circumstances of laboratory closures and the fact the research project finished already hinder the possibility of including additional testing data, for which we offer sincere apologies to the reviewer.
7. After RTFO aging, R% reduced for both NR samples. Please explain.
Explained in text, page 17:

"The reduction in the percent recovery (%R) is explained by the possible damaging of the NR crosslink structure (i.e., polymer network stability) induced by this short-terms ageing protocol, which inevitably

affects the permanent deformation response of the NRMBs at both concentrations studied. Under this oxidative condition, the hardened bitumen phase is responsible for the most part of the rheological response. Previous research found similar results of dropping recovery values when polymer-modified bitumen blends were subjected to artificial ageing cycles in both RTFO and PAV (Cuciniello et al. 2018, Cuciniello et al. 2019)”.

Cuciniello, G., Leandri, P., Filippi, S., Presti, D. L., Losa, M., & Airey, G. (2018). Effect of ageing on the morphology and creep and recovery of polymer-modified bitumens. *Materials and Structures*, 51(5), 136.

Cuciniello, G., Leandri, P., Presti, D. L., Losa, M., & Airey, G. (2019). Investigating the Effect of Artificial Ageing on the Creep and Recovery of SBS-Modified Bitumen. In *MATEC Web of Conferences* (Vol. 271, p. 03009). EDP Sciences.

8. The possible reason that 7% NRMB presents poor fracture performance could be the of discrete NR molecules, which are not bounded together, which caused stress concentration inside the asphalt samples. The authors are encouraged to cite some references on DENT results for SBS modified asphalt. Through the comparison, one could understand easier for NRMBs.

This reasoning proposed by the reviewer is agreed and has been added to the manuscript, along with some references of DENT results for SBS-modified bitumen.

Included in Page 19 and Page 20:

“The results observed for the maximum concentration of NR in the NRMBs might be associated by the detrimental side effects produced by the artificial ageing regimes (i.e., RTFO and PAV) that are required prior testing for DENT. More precisely, as discussed before, the polymer network or cross-link of the NR molecules seems to be disintegrated by the short- and long-term ageing, which results in stress concentration in the bitumen matrix. Previous research on fracture response of SBS-modified bitumen through DENT testing found that high polymer concentrations (i.e. higher than 2%) tend to have poorer fracture performance in comparison to reasonably low contents of modifiers. In fact, the use of WMA additives such as Fischer-Tropsch (FT), chemical surfactants (CS) and water-based (WB) have the ability to modify the fracture response, which tends to be concentration-dependent and showed improved fracture response at 2% concentration for CS WMA-additive type, but this response was opposite for the FT and WB types with the only exception of 6% WB concentration (Kataware & Singh, 2018). Additional research by Paliukaite et al. (2015) substantiated a detrimental ductile response of high-polymer modified bitumens with SBS (i.e., 3-5% concentration by weight of base bitumen). In fact, it was found that both in unaged and PAV-aged conditions, the CTOD values for the SBS-modified samples had lower ductile performance (i.e. lower CTOD values), with one single exception to this trends for a SBS-blend (A5), which had the same CTOD value, measured at deformation rate of 50 mm/min, as the base bitumen after long-term ageing protocol. Additional research on PMB-bitumen modification through various concentrations of RAP binder (i.e., 0%, 15%, 25%, and 40%) indicated that the higher concentrations of this bitumen modified negatively affect the intermediate temperature response in terms of the ductile properties measured through DENT and elastic recovery tests (Singh et al., 2019). Conversely, Kanabar (2010) found that for low-SBS polymer concentrations the fracture performance after PAV-ageing seems comparable and at some instances superior to that of base bitumen for concentrations ranging from 1.2% to 1.8% by weight of base bitumen. It was also found that the superior response at RTFO ageing conditions is not equaled by that of PAV-aged materials, indicating a detrimental effect of artificial ageing on the fracture response for these material combinations”.

Kataware, A. V., & Singh, D. (2018). Evaluation of intermediate temperature cracking performance of warm mix additive modified asphalt binders. *Construction and Building Materials*, 184, 165-176.

Paliukaite, M., Verigin, M., & Hesp, S. (2015). Double-edge-notched tension testing of asphalt cement for the control of cracking in flexible asphalt pavements. *Bituminous mixtures and pavements VI*, 13.

Singh, D., Girimath, S., & Ashish, P. K. (2019). Effect of recycled asphalt binder on high and intermediate temperature performance of polymer modified asphalt binder. *International Journal of Pavement Research and Technology*, 12(5), 486-496.

Kanabar, N. (2010). *Comparison of ethylene terpolymer, styrene butadiene, and polyphosphoric acid type modifiers for asphalt cement* (Doctoral dissertation). Queens University, Canada.

Reviewer #3

The paper relates well to the topics of this Journal, and I did not find plagiarism or republication problems. The paper is not exciting because it does not seem to bring sufficiently new ideas, even though the authors explained that few papers have worked in the modifications of bitumen with natural rubber (I found 78 documents on that topic in a quick search). Nevertheless, the article accomplishes all other formal objectives of a research paper (i.e., a well-designed methodology, correct analysis and discussion of results, conclusions are based on the results of the work). **Overall, this paper can be accepted for publication after minor changes.**

Some questions to be analyzed or explained in more detail:

1. The number of binders studied was short (only the base binder and two NR modified binders). If authors intend to use NR for bitumen modification, they should have compared the results obtained with those of a commercial modified PMB. Another alternative is to modify the same base bitumen with SBS or recycled rubber since those are the materials used for asphalt binder modification. If authors can present one of those PMB materials, the quality of the paper would be significantly improved.

This comment is reasonable and valid, however, the main objective was investigating the effects of NR on bitumen modification from a comprehensive set of experimental methods helping to understand a wide range of properties at high-intermediate-low testing conditions, and for this reason the experimental programme included the set of materials already presented. On the other hand, at this moment it is not possible for the authors to conduct further manufacturing and testing on bitumen samples and NRMBs as laboratories are currently closed due to current COVID circumstances and the fact the experimental work associated to this research project has been completed, for which the authors present sincere apologies.

2. More information on the natural rubber should be given (Is it liquid or solid? What are its origin and global properties?). If this is the novelty of the paper, the authors should clearly demonstrate its importance. Physicochemical properties and a more detailed description of the natural rubber have been included in the materials section. Page 2 and 3.

3. The selection of a hard bitumen for bitumen modification is not usual. Moreover, the amounts of NR used were arbitrary (i.e., polymer modification is usually made with up to 5% polymer, while modification with recycled rubber can use up to 22% rubber). Please give additional details on these points.

One of the most commonly used penetration grade bitumen in South East Asia is AC60/70, for that reason and to evaluate more critical conditions of the materials, an AC50/70 was used for the investigation of the NR effects in the sense this bitumen type does not massively differ from the 60/70 type and due to material availability in the UK at the time the research was conducted. Regarding concentrations of NR, the values were not arbitrary as these considered the typical amounts normally used in Southeast Asia and these were selected in accordance with previously published academic literature, including, for instance, the following references:

- Abdulrahman, S., Hainin, M. R., Idham, M. K., Hassan, N. A., Warid, M. N. M., Yaacob, H., ... & Puan, O. C. (2019, May). Physical properties of warm cup lump modified bitumen. In *IOP Conference Series: Materials Science and Engineering* (Vol. 527, No. 1, p. 012048). IOP Publishing.
 - a. These authors incorporated concentrations of 2.5%, 5.0%, 7.5%, and 10% cup lump rubber to 60/70 base bitumen.
- Azahar, N. M., Hassan, N. A., Putrajaya, R., Hainin, M. R., Puan, O. C., Shukry, N. A. M., & Hezmi, M. A. (2019, January). Engineering properties of asphalt binder modified with cup lump rubber. In *IOP Conference Series: Earth and Environmental Science* (Vol. 220, No. 1, p. 012014).
 - a. Using a 60/70 as base bitumen, the authors incorporated cup lump rubber at 5%, 10%, and 15% concentrations by weight of base bitumen.
- Bakar, S. K. A., Abdulah, M. E., Kamal, M. M., Abd Rahman, R., Hadithon, K. A., Buhari, R., & Tajudin, S. A. A. (2018). Evaluating the rheological properties of waste natural rubber latex modified binder. In *E3S Web of Conferences* (Vol. 34, p. 01037). EDP Sciences.
 - a. This investigation studied concentrations of waste NR latex ranging from 5% to 15% by weight of base bitumen (penetration grade 80/100)

4. The industrial production of these "new" binders with NR is an issue to be addressed for some possible readers of the paper. Would this NR modified binder have enough storage stability? Is it easier or harder to modify binders with NR in comparison with other polymers?

This has been added as a recommendation for future work.

Although storage stability of the samples has not been studied in our research work, previous work by Al-Mansob et al. (2014a) substantiated that NR contents of up to 6% exhibit stable performance for use at high

temperatures. Furthermore, Al-Mansob et al. (2014b) demonstrated with factual evidence that contents up to 9% can have satisfactory storage stability and indicated that this property is largely dependent on the NR concentration. Similar findings were reported by Usman et al. (2019), who reported satisfactory storage stability for blends subjected to conditioning for 72 hours at 163°C. Therefore, future studies might add value to the characterization of NRMB by incorporating the storage stability results.

Al-Mansob, R. A., Ismail, A., Alduri, A. N., Azhari, C. H., Karim, M. R., & Yusoff, N. I. M. (2014). Physical and rheological properties of epoxidized natural rubber modified bitumens. *Construction and Building Materials*, 63, 242-248.

Al-Mansob, R. A., Ismail, A., Md Yusoff, N. I., Azhari, C. H., Karim, M. R., Alduri, A., & Baghini, M. S. (2014). Rheological characteristics of epoxidized natural rubber modified bitumen. In *Applied Mechanics and Materials* (Vol. 505, pp. 174-179). Trans Tech Publications Ltd.

Usman, A., Izzaty, N., Ibrahim, R., Sutanto, M. H., & Sunarjono, S. (2019). Storage Stability and Morphology of Latex Modified Bitumen. *International Journal of Innovative and Exploring Engineering (IJITEE)*, ISSN, 2278-3075.

In terms of the complexity of bitumen modification through NR in comparison to other polymers, our experience and that reported by previous research indicates (Biro & Fazekas, 2012; Porto et al., 2019) the following:

Overall, it is harder to mix NR latex than other polymers because of its liquid nature in contrast to others being capsules or grinded materials (solid-like). Furthermore, the fact that NRL contains “water” in the latex, and has high ammonia content makes it hard to produce blending due to the physical effects (i.e., frothing and foaming), in addition to the strong smell produced by the ammonia. For these reasons, the liquid NRL is gradually added to the base bitumen at small portions, which allows for reducing the air bubbles and evaporation. This is aided by manual stirring to minimize the foaming of the rubber to reach the top head of the mixing machine. Certainly, this procedure means more time consumption than other polymers but it allows for homogeneous blend if carried out cautiously.

Biro, S., & Fazekas, B. (2012). Asphalt rubber versus other modified bitumens. In *The Asphalt Rubber 2012 Conference Proceedings, Munich* (pp. 351-366).

Porto, M., Caputo, P., Loise, V., Eskandarsefat, S., Teltayev, B., & Oliviero Rossi, C. (2019). Bitumen and bitumen modification: A review on latest advances. *Applied Sciences*, 9(4), 742.

5. The low performance at low temperatures, the stress sensitivity in MSCR and the results of NR modified binders in the fractures test are unexpected. Authors mentioned that this is not a problem because these binders would be applied in southern Asia, but this is a limitation of this material. The authors are encouraged to compare this low-performance problem with that of other polymer modified binders, to demonstrate that this is a typical problem of PMBs (probably it is not).
This reasoning proposed by the reviewer is agreed and has been added to the manuscript, along with some references of DENT results for SBS-modified bitumen. Included in Page 19 and Page 20
6. Authors can evaluate the elastic recovery of the NR modified binders in MSCR, and present those results. That would be interesting to present an additional property of these binders.
A comprehensive comparison of the permanent deformation results for the materials had already been included and discussed in the paper, in the section of MSCR results. This analysis included the non-recoverable creep compliance (J_{nr}), the percent recovery (*R-value*) and the stress sensitivity parameter ($J_{nr-diff}$). Through these parameters, a clear understanding of the effects of NR on the base bitumen is appreciated, which the authors consider as valid and sufficient to support the rutting performance of the manufactured blends, as it is consistent with many published work studying the permanent deformation response. Therefore, the authors appreciate the comment raised by the reviewer and hope the assessment presented is valid enough to support the aim and objectives of the research paper.
7. Taking into account my previous comments, the approach presented in this paper does not seem to be genuinely innovative, but that is compensated with a well-designed methodology for the study. The organization of the article is right. The description of some materials and methods can be improved, especially the NR description and the fracture test. The discussion of results is good, and further scientific additions to justify the performance at low-temperatures can be added. The conclusions are correct and based on the results of this work. Written English quality is excellent. The objectives are clear, and the paper globally answers those objectives (although the comparison with a commercial PMB is missing).
The authors truly value these final comments about the submitted paper and hope that the changes addressed on the document will satisfy the suggestions provided by the peer reviewers.

I appreciate your kind attention to this letter and the revised manuscript.

Sincerely,

A handwritten signature in black ink, reading "Juan S. Carvajal Muñoz". The signature is written in a cursive style and is positioned above a horizontal line.

Juan S. Carvajal-Muñoz

PhD Student, Nottingham Transportation Engineering Centre (NTEC). The University of Nottingham

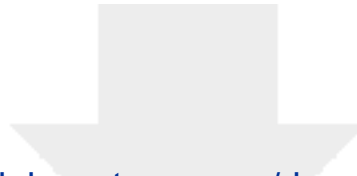
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Research highlights

- The effects of Natural Rubber (NR) were captured through rheological master curves and black diagrams.
- Positive effects on permanent deformation resistance of NR were observed.
- Fatigue cracking resistance was improved by NR addition.
- Thermal cracking resistance (ΔT_c) was satisfactory for 3% NR content.
- Fracture analysis via DENT test substantiated benefits on cracking performance of 3% NR content.



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Declaration of interests

The authors of the manuscript titled "**Performance-Related and Rheological Characterisation of Natural Rubber Modified Bitumen**" declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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

Jarurat Wititanapanit: Conceptualization, Methodology, Investigation, Formal-Analysis, Writing- original draft, Project Administration. **Juan S. Carvajal-Munoz:** Writing- original draft, Writing- review and editing, Visualization, Co-Supervision. **Gordon Airey:** Conceptualization, Methodology, Visualization, Writing-review and editing, Supervision.

Performance-Related and Rheological Characterisation of Natural Rubber Modified Bitumen

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Performance-Related and Rheological Characterisation of Natural Rubber Modified Bitumen

Abstract

This paper ~~has been~~ motivated by the limited ~~previous~~ research ~~available~~ evaluating the ~~comprehensive~~ effects of natural rubber ~~bitumen modification~~ (NR) for pavement applications. ~~The study has comprised a detailed comprehensive~~ laboratory programme ~~was carried out to evaluate the influence of NR on the high-intermediate-low temperature performance, including rheological characterisations and performance-related tests (fatigue cracking, thermal cracking, fracture and permanent deformation resistance).~~ The main ~~results~~ findings ~~indicated~~ indicate some beneficial effects of ~~that using~~ NR on the performance-related response for fatigue and permanent deformation. NR (3 and 7%) induced ~~positive effects on the fatigue cracking and rutting performance.~~ ~~Performance.~~ Additional benefits were ~~also found in terms of~~ observed regarding the ~~fracture strength and~~ the thermal cracking resistance (AT_c). The ~~viscoelastic~~ effects of NR ~~on the rheological modification of base bitumen~~ substantiated ~~produced~~ significant changes in the ~~viscoelastic characteristics~~ rheological properties of the ~~control base~~ bitumen. Overall, ~~the results of this research study encourage the use of NR for pavement applications~~ the use of NR at a concentration of up to 7% by weight of bitumen proved to be a reasonable option for use in pavement applications, having more substantial benefits in tropical regions in agreement with its predominant effects on the bitumen modification at intermediate-to-high temperature conditions.

Keywords: Natural Rubber; Bitumen Modification; Fatigue; Rutting; Fracture; Performance; Rheology.

1. Introduction

Natural rubber (NR) is a material produced from native trees (*Hevea brasiliensis*) in ~~South Asia, which is~~ commonly used as a manufactured product either solely or combined with other materials in ~~various industries (e.g., the automobile, textile, insulating membranes, adhesives, footwear and construction) industries.~~ NR is widely used ~~due as a consequence of~~ its high stretch ratio (i.e., ~~high ductile response or~~ elasticity), ~~its~~ ability to withstand external loading (i.e., ~~permanent deformation resistance~~) and ~~the high~~ water resistance. ~~Although in spite of being extensively~~ produced ~~extensively~~ and commercialised from the ~~early~~ 70's, the ~~current~~ bulging stocks of NR ~~currently available~~ in Southeast Asian countries and the trends of decreasing trade price ~~have encouraged~~ ~~are gradually encouraging~~ governments and environmental agencies to ~~find~~ ~~sought for~~ alternative applications in road infrastructure projects [1, 2].

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7 ~~At the same time~~ Similarly, the pressing environmental needs for recycling ~~and demands the~~ selection of
8 ~~environmentally friendly~~ environmentally friendly alternatives ~~as surrogates of~~ finite natural resources, ~~which~~
9 ~~has encouraged the~~ ~~supposes the~~ incorporation of waste materials ~~such as recycled tyre rubber [3-6] for~~ road
10 infrastructure applications, ~~such as the use of recycled tyre rubber [3-6]. At this respect, From from~~ the late 80's
11 to the early 90's, research studies have assessed the effects of waste rubber (typically reclaimed from tyres) on
12 bituminous mixtures incorporating multiple approaches and techniques [7-9]. For instance, [10] provided
13 practical guidelines for the design, construction and maintenance of tyre-rubber modified mixtures. More
14 recently, [5] presented a comprehensive literature review of the various benefits associated with the use of
15 recycled tyre-rubber (RTR) with a critical analysis of the existing technologies and specifications that apply to
16 the production process (i.e., production, handling and storage).

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23 ~~As previously mentioned~~ Nevertheless, most investigations of ~~using~~ rubber ~~applications to~~ for road infrastructure
24 ~~applications have dealt with~~ relate to RTR effects on bituminous mixtures [11-14] but only limited information is
25 available ~~on the virgin rubber (-for NR-)~~ in terms of its ~~multi-scale~~ effects ~~on the mixture components and~~
26 ~~performance of~~ on bituminous mixtures. For instance, [15] studied the ~~consistency characteristics~~ effects
27 of Natural Rubber ~~on~~ Modified Bitumen Blends (NRMB) based on penetration, softening point and toughness,
28 ~~which substantiated significant~~ The main results validated the improvement of ~~the~~ permanent deformation
29 resistance ~~induced by NR~~. Similarly, [16] concluded that ~~bitumen modification through NR can~~
30 ~~improve~~ improves the durability of polymer modified asphalt (PMA) mixtures, ~~and this effect is more prominent~~
31 ~~when combined through the combination of NR~~ with peroxy formic acid to produce epoxidised natural rubber
32 ~~that induced a high level of modification of the base bitumen.~~

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39 ~~There is therefore a~~ The need to attain further experimental evidence ~~to provide a deeper~~ that aids understanding
40 ~~of~~ the effects of NR on bitumen modification and performance of bituminous mixtures ~~supports this research~~
41 ~~study~~. Subsequently, this paper aims to investigate the influence exerted by NR on the rheological properties
42 and performance-related characteristics of NRMB ~~(i.e., fatigue cracking, thermal cracking, fracture, and~~
43 ~~permanent deformation)~~ in terms of fatigue, rutting and fracture strength. The paper incorporates a range of
44 ~~rheological~~ tests and analytical theories ~~to to provide data that help determining~~ study the performance of NR
45 modified bitumen ~~for road infrastructure applications in warmer climates such as those specified in Thailand or~~
46 ~~similar countries~~. The paper ~~structure includes a detailed description of the~~ describes the details and materials
47 ~~and~~ experimental techniques ~~of five binder test methods as well as material details focused for assessing the~~
48 NRMBs ~~using different NR contents~~. The ~~rheological and fracture~~ laboratory data ~~is then~~ is further analysed

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7 following in agreement with standardised relevant analytical approaches and current scientific literature. The
8 paper is completed with a list associated with the test methods before compiling a list of the major findings,
9 relevant conclusions and recommendations for further research.

10 11 12 2. Materials and Testing Methods

13 14 2.1. Experimental Programme

15
16 A comprehensive laboratory programme was carried out as shown in Figure 1, with NR contents of 3 and 7%
17 by total weight of modified bitumen. The natural rubber latex used for bitumen modification is in liquid form
18 and containing high ammonia (HA), with a chemical structure of cis-1, 4-polyisoprene (also called isoprene)
19 with a chemical formula of C_5H_8 with a molecular weight distribution of $10^5 - 10^7$ g/mol. The main properties of
20 the NR used include those presented in Table 1. A 40/60-penetration grade bitumen was used as the base
21 bitumen (referred to as "control" in the manuscript). with NR contents of 0%, 3% and 7% by total weight of
22 modified bitumen were used. Five bitumen The laboratory tests with specific objectives were included in the
23 experimental programme as shown are listed in Figure 1, with each one of the Each tests result correspond to the
24 average of being conducted on three test replicates, with the exception of the DENT test that was (conducted on
25 2 replicates as per standard guidelines).

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32 **Table 1.** Natural rubber properties.

Parameters	Unit	Specification	Results
Total solids content (TSC)	% by mass	61.50 min.	61.80
Dry rubber content	% by mass	60.00 min.	60.21
pH value at 25.4°C	--	11 max.	10.48
Pottassium Hidroside Number (KOH)	% by mass	0.70 max.	0.69
Alkalinity (as ammonia on total weight)	% by mass	0.65-0.75	0.68
Non rubber content	% by mass	1.70 max.	1.59
Volatile fatty acid number	--	0.05 max.	0.0229
Mechanical stability time at 55% TSC	Sec.	1000 - 1200	1080
Specific gravity at 25°C	--	0.94	0.9448
Magnesium content	ppm	40 max.	36.75

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Viscosity at 60% TSC, spindle no.1 at 60 rpm	cps	--	59.50
Coagulum content, 80 mesh	ppm	--	20

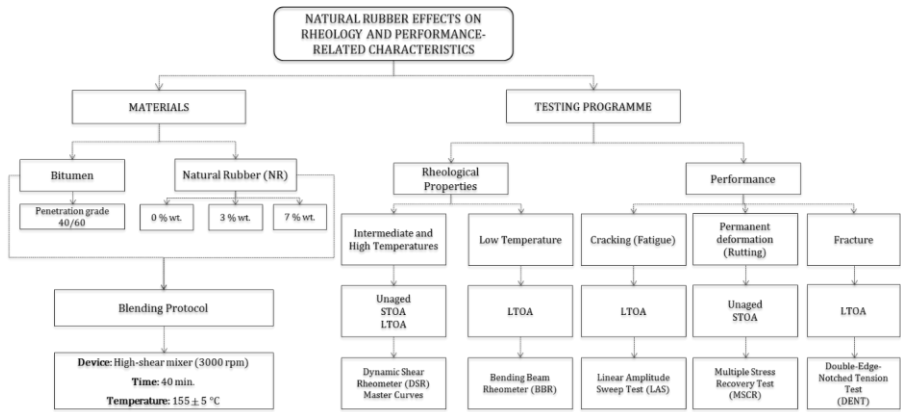


Figure 1. Testing programme for NRMB characterisation. **Note:** STOA refers to samples subjected to RTFO, LTOA refers to samples subjected to RTFO + PAV.

2.2. Blending Protocol

A high-shear mixing approach was followed to guarantee sufficient material dispersion and interaction time between the base bitumen and NR. For this purpose, a high-shear mixer with double-head was used. The procedure for NRMB manufacture included the following steps: (1) The base bitumen (1500g) was preheated in an oven at 150°C for 120 minutes. Simultaneously, NR was weighed for mixing at the desired concentration. (2) The base bitumen was extracted from the oven and transferred to a hotplate with the high-shear mixer setup and the ability to keep the temperature at 150°C. (3) The required amount of NR (45g and 105g for 3% and 7% blend, respectively) were gradually added to the neat bitumen in order to minimize formation of lumps and air bubbles (due to the presence of water-base liquid) and thereby maximize blending efficiency. (4) The bitumen was manually stirred with a glass rod during NR addition until a fairly homogeneous blend was obtained with minimal formation of air bubbles. (5) Once the blend reached the target temperature on the hotplate (150 °C), checked periodically with a hand thermometer, mechanical blending commenced from low speed (800 rpm) up to a maximum of 2,400 rpm (for 3% NR content) and 4,200 rpm (for 7 % NR content).

2.3. Linear Viscoelastic Rheological Characterisation

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7 Strain-controlled frequency sweep tests were conducted using a [Bohlin Gemini®](#) Dynamic Shear Rheometer
8 (DSR) to determine the viscoelastic properties of the base bitumen and to assess the level of modification
9 induced by NR. For this, parallel-plate geometry was used (8 mm and 25 mm) with 2 mm and 1 mm gap,
10 respectively. The bitumen was heated up in the oven at 150 °C and then poured into the 8 mm and 25 mm
11 moulds and samples were mounted in the DSR after the samples had cooled down and could be removed from
12 the moulds. Low to intermediate temperatures (5 °C to 35 °C) were assessed with the 8 mm geometry, whereas
13 high temperatures (35 °C to 75 °C) used the 25 mm geometry. Frequencies from 0.1 Hz to 10 Hz were used with
14 a strain level of 0.5% (within the Linear Viscoelastic Region (LVE), based on amplitude sweep tests). The time-
15 temperature superposition principle (TTSP) was applied for shifting of the rheological data (i.e., $|G^*|$ and δ) to
16 produce smooth master curves. The shifting of the data was done using a 25 °C reference temperature with the
17 application of the Williams-Landel-Ferry (WLF) model. In addition to the [master curves of \$|G^*|\$ and \$\delta\$](#)
18 [curves, a comparison of these parameters was made through Black Space Diagrams](#) and due to their ~~its~~
19 simplicity ~~(as it does not require shifting of rheological data. These rheological parameters were used), a~~
20 [comparison of these parameters was also made using Black Space Diagrams in order](#) to study the relationship
21 between stiffness and viscoelastic properties of the base bitumen and NRMBs [17].
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30 **2.4. Performance-Related Tests**

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32 A DSR was used to characterise the rheological and performance properties of the base bitumen and NRMB
33 blends. In addition to standard oscillatory linear viscoelastic (LVE) frequency sweep tests, the DSR was also
34 used to study the fatigue and permanent deformation performance-related characteristics of the materials.
35 Fatigue [cracking](#) was assessed through the Linear Amplitude Sweep (LAS) test and [permanent deformation](#)
36 [\(rutting\)](#) by means of the Multiple Stress Creep Recovery (MSCR) test. The fracture characteristics of the
37 [binders-materials](#) were ~~also~~ determined using the Double-Edged Notched Tension (DENT) test. These ~~tests~~
38 are considered performance-related tests due to their ability to correlate material response to those of real pavement
39 structures subjected to various loading regimes [18]. The detailed description of these tests is presented in
40 subsequent sections.
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46 **2.4.1. Linear Amplitude Sweep (LAS)**

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48 The LAS test (AASHTO TP101) [19] was used to study the fatigue resistance of the base bitumen and NRMBs
49 as a function of ageing and NR content. The test method is intended for accelerated damage and involves cyclic
50 loading of bitumen samples that are subjected to increasing strain amplitudes. This leads to damage
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7 accumulation, which is used to assess fatigue performance in terms of the number of cycles required to failure
8 (i.e., a decrease of the initial $|G^*|$ value at the peak shear stress, as per [19]). Aged bitumen samples of 8 mm
9 diameter and 2 mm thickness are tested in a strain-controlled mode using the DSR parallel plate geometry. Two
10 test stages are ~~carried out~~ conducted as follows: (1) The initial stage is non-destructive and involves assessing the
11 undamaged rheological properties of the bitumen samples in terms of the complex modulus (G^*) and phase
12 angle (δ). This stage is done via a frequency sweep test (i.e., oscillatory loading from 0.2 Hz to 30 Hz at 0.1%
13 strain amplitude). (2) Subsequently, the bitumen sample is subjected to a rest period of 10s before the onset of
14 the secondary stage. This secondary stage is destructive in nature through applying increasing strain amplitudes
15 (from 1% to 30% strain) at a constant frequency of 10 Hz and is referred to as the continuous oscillatory strain
16 sweep test. In brief, the test parameters include: (1) 100 loading cycles at 0.1% strain to obtain undamaged
17 material response, and (2) 30 subsequent rounds of 100 cycles from 1%-30% strain, increasing linearly by 1%,
18 for a total of 3,100 cycles of loading [20].

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21 Considering previous literature recommendations for minimizing the risk of delamination (i.e., poor
22 bitumen/plate bonding), caution was taken to select the testing temperature [21]. To control ~~this~~
23 issuedelamination issues, the failure plane of the sample was checked at the end of the test to identify the typical
24 torsional-fatigue failure of each specimen (i.e., circumferential crack) [22]. The temperature for LAS test was
25 ~~set as~~ 15 °C and 25 °C for all materials, based on previous literature studying fatigue at intermediate
26 temperatures [23]. The selection of two test temperatures was considered to assess the effects of this factor on
27 the materials' fatigue response. In this study, two strain levels were considered to take into account both
28 "strong" and "weak" pavement structures. For a "strong" layer with thickness higher than 100 mm, a low strain
29 ($\gamma = 2.5\%$) was selected. Similarly, for a "weak" layer with thickness lower than 100 mm, a higher strain ($\gamma =$
30 5.0%) was considered [24].

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33 Viscoelastic continuum damage (VECD) mechanics, adopting Schapery's theory, was used to simulate damage
34 growth and thus predict fatigue life as a function of strain (Equation 3). It is based on the simplified classic
35 fatigue law (Equation 1) with A and B being obtained from the mathematical relationship between strain (γ) and
36 cycles to failure (N_f) as shown in Equation 2,

$$N_f = A(\gamma_{max})^B \quad \text{Equation 1}$$

$$A = \frac{f(D_f)^k}{k \left(\pi \frac{I_D}{|G^*|} C_1 C_2 \right)^\alpha} |G^*|^{-\alpha} \quad \text{Equation 2}$$

$$B = -2\alpha$$

$$N_f = \frac{f(D_f)^k}{k \left(\pi \frac{I_D}{|G^*|} C_1 C_2 \right)^\alpha} |G^*|^{-\alpha} (\gamma_{max})^{-2\alpha}$$

Equation 3

Where,

$$k = 1 + (1 - C_2)\alpha,$$

f is the loading frequency, Hz,

$|G^*|$ is the undamaged complex shear modulus,

γ_{max} is the maximum expected binder strain for a given pavement structure in %,

D_f is the damage accumulation at failure, defined as the $D(t)$ corresponding to the reduction in initial $|G^*|$ at the peak shear stress.

2.4.2. Double-Edged Notched Tension (DENT)

The fracture characteristics in the ductile state of bitumen/NRMBs were studied following the Canadian Standard (Test Method LS-299) [25]. The essential work of fracture required to generate new surfaces was studied and determined through the DENT test, following the Canadian Standard (Test Method LS-299) [25]. DENT test, which was used to determine the ductile failure resistance under horizontal tensile load application, at a specified temperature (15°C). Initially, as per standard guidelines, a 15 °C test temperature was considered based on the standard, but at this condition this induced the samples exhibited a premature brittle-failure (i.e., sample breakage). For this reason, the selection of and a higher testing temperature of 25 °C was therefore selected required in order to facilitate a more ductile material response. The fracture properties studied included: (1) The essential work of fracture (W_e), (2) plastic work of fracture (W_p), and (3) the approximated critical crack-tip opening displacement ($CTOD$). These properties were calculated to study the resistance of the bitumen/NRMBs to ductile fracture. The DENT test was conducted after conditioning of the samples had been conditioned in a water bath (i.e., the specimens were submerged 25 mm below water surface) at the target test temperature of 25 °C for 180 minutes. The deformation load was applied at a controlled rate of 50 mm/min deformation and it stopped when rate until the samples reached the ductile failure. During testing, data of the elongation length (in mm), and force (in Newtons) was recorded and subsequently analysed to determine the fracture parameters (i.e., W_e , W_p and $CTOD$).

The significance of the measured Calculation of fracture properties for the comparison of the materials' fracture performance include relate to: (1) the total work of fracture (W_t), which refers to the area under the load versus

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7 load-line displacement curve, kJ (Equation 4); (2) the specific total work of fracture (w_t), related to each
8 replicate sample tested, kJ/m² (Equation 5); (3) the specific essential work of fracture (w_e), the energy required
9 to fracture or break the sample without plastic deformation away from the fracture zone, kJ/m²; (4) the specific
10 plastic work of fracture (w_p), the non-essential work dissipated during the deformation of a volume of bitumen
11 around the fracture zone, MJ/m³; (5) the geometric constant of the plastic zone (β), and (6) the critical tip
12 opening displacement (CTOD), mm (Equation 6 and Equation 7).

$$13 \quad W_t = \int_0^{t_f} P \times d \quad \text{Equation 4}$$

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19 Where,

20 P is the load in Newtons,

21 d is the displacement in the test in m,

22 t_f is the time when the maximum stroke is reached or ductile failure, whichever is attained first.

$$23 \quad w_t = \frac{(\text{average } W_t)}{(B \times l)} \quad \text{Equation 5}$$

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28 Where,

29 B is the sample thickness in m, and

30 l is the ligament length (the space between the notches) in m.

$$31 \quad COTD = \frac{w_e}{\sigma_n} \quad \text{Equation 6}$$

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35 Where,

36 σ_n is the net section stress of specimen in N/m², calculated using

$$37 \quad \sigma_n = \frac{P_{peak}}{(B \times l)} \quad \text{Equation 7}$$

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Where, P_{peak} is the average peak load of the specimen tested with the smallest ligament length (i.e., the average maximum load for the 5 mm ligament specimens).

Determination of w_e and βw_p was done either graphically by plotting w_t for the three ligament lengths or mathematically using the least squares fitting method. w_e refers to the specific essential work of fracture (i.e., w_t value when $l = 0.0$) and βw_p is the slope of the best fit straight line (for $w_t = w_e + \beta w_p l$).

2.4.3. Multiple Stress Creep Recovery (MSCR)

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7 The MSCR test was used to determine the permanent deformation susceptibility of the base bitumen and
8 NRMBs. This test was conducted on unaged and RTFO-aged materials to study their elastic recovery properties.
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10 A DSR parallel-plate geometry with spindle diameter of 25 mm and 1 mm testing gap was used to undertake the
11 test, [using a Bohlin CVO® equipment](#). Test temperature of 60°C was selected based upon typical ranges
12 reported in literature for pavement surface temperature in Thailand (65-68 °C [26]; and 53-59 °C [27]). The test
13 followed the AASHTO TP 70 Standard method [28]. In brief, the sample was loaded for ~~240~~ cycles of creep-
14 recovery at ~~two~~ [0.1 kPa](#) stress levels [and 10 cycles for the \(0.1 kPa and 3.2 kPa stress condition\)](#). First, a 1
15 second creep load is applied and subsequently released for 9 seconds. Data was recorded by the DSR equipment
16 and subsequently analysed in terms of the % recovery (*R-Value*), the non-recoverable creep compliance (J_{nr}),
17 and the stress sensitivity parameter ($J_{nr-diff}$). Equations as per standard were used to calculate the parameters for
18 both stress conditions (0.1 kPa and 3.2 kPa). The *R-value* refers to the ability of bitumen to recover (*i.e.*,
19 dissipate stresses) after repeated loading and higher values reflect better material response in terms of permanent
20 deformation. The J_{nr} is an indicator of permanent deformation resistance that for higher values indicates a higher
21 susceptibility to rutting distress of a given bitumen. The $J_{nr-diff}$ is used to distinguish the stress sensitivity of the
22 bitumen due to the change of low stress (0.1 kPa) to high stress (3.2 kPa) conditions. Higher values ~~of~~ [for](#) this
23 parameter indicate higher sensitivity to the change in the loading stress to which the bitumen is subjected to
24 [under specific temperature conditions](#).
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34 **2.5. Low-temperature Rheological Characterisation**

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7 The low-temperature rheological properties for the bitumen/NRMBs were studied by means of the Bending
8 Beam Rheometer (BBR) in accordance with the AASHTO T313-12 Standard [29]. In brief, the test method
9 involved the application of a 980 mN load to the beam specimen during 60 seconds of loading using the three-
10 point-bending approach. During the loading period, the low-temperature creep stiffness (S) and relaxation
11 properties (m-values) were recorded and later analysed for assessing the temperature/~~dosage-concentration~~
12 effects and determination of critical low-temperature values ($T_c(S)$ and $T_c(m)$) and the differential of these
13 values, ~~Delta Tc~~ $-(\Delta T_c)$. The measured stiffness was calculated using the loading, deflection values and
14 geometrical features of the beams manufactured by using the beam theory. The estimated stiffness was
15 calculated ~~as indicated~~ from a mathematical fitting of the data. The m-value refers to the slope of the
16 relationship between the logarithmic of measured stiffness and is obtained at the logarithm of time for the total
17 of 240 seconds of loading. ~~Test temperatures included for the low-temperature characterisation included -18 °C~~
18 ~~and -24 °C.~~

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26 ~~Test temperatures were -18 °C and -24 °C and~~ ΔT_c values were calculated using Equation 8. This parameter is a
27 measure of the closeness of the critical low temperatures measured from the Stiffness and m-value with respect
28 to their influence on the low temperature. Previous research has suggested this parameter is valid for more
29 accurate prediction of field performance of bitumen (fatigue cracking response) as a result of its relation to
30 bitumen ductility [30]. This research proposed two limits for ΔT_c that refer to a cracking warning level and a
31 limiting value of -2.5 °C and -5.0 °C. These criteria were incorporated ~~in this research infor~~ the discussion of
32 results. The critical low-temperature values ($T_c(S)$ and $T_c(m)$) were determined as per standard. From these
33 values, the higher temperature is selected as the critical one for a given material [31].

$$\Delta T_c = T_c(\text{Stiffness}) - T_c(m - \text{value}) \quad \text{Equation 8}$$

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39 Where: T_c is the critical temperature for each of the BBR parameters (S , m -value).

40 41 42 **2.6. Ageing Protocols**

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44 To simulate ageing occurring during manufacturing and laying of bituminous mixtures, Short-Term Oven
45 Ageing (STOA) and Long-Term Oven Ageing (LTOA) protocols were conducted on the base bitumen and
46 NRMBs manufactured through the blending approach previously described. The STOA was carried out with the
47 Rolling Thin Film Oven (RTFO) test [32] and LTOA with the Pressure Ageing Vessel (PAV) [33].

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49 The effects of NR on NRMBs are analysed in terms of: (i) rheological characteristics at low-intermediate-high
50 temperatures, and (ii) performance-related responses (fatigue performance, permanent deformation resistance

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7 and fracture properties). The effects of short- and long-term oven ageing on the rheology and performance-
8 related characteristics of NRMBs are also quantified.
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10 11 **3. Rheological Changes Induced by NR**

12 **3.1. Intermediate-High Temperature Effects (Master Curves)**

13
14 The effects of NR in terms of the rheological changes induced to the base bitumen at the ~~two-three~~ NR ~~contents~~
15 ~~concentrations~~ were assessed in terms of complex modulus (G^*) ~~and~~ phase angle (δ) master curves ~~as well~~
16 ~~as and the Black Space diagrams~~ ~~Diagrams~~. ~~For the sake of brevity, o~~ Only the results of the unaged materials are
17 shown ~~taking into consideration that for the sake of brevity although the same similar~~ observations ~~were attained~~
18 ~~for the short-term (RTFO) and long-term (RTFO + PAV) ageing conditions~~ in terms of G^* and δ master curves
19 and Black Space diagrams ~~were found for the short-term (RTFO) and long-term (RTFO + PAV) ageing~~
20 ~~conditions~~. The rheological results in Figure 2 show that the effects of NR on the rheological parameters G^* and
21 δ are more pronounced at higher temperatures and lower loading frequencies as demonstrated in Figure 2a, and
22 Figure 2b. These results indicate that NR produces NRMBs with an increased elastic response (*i.e.*, lower phase
23 angles) and increased stiffness (*i.e.*, higher complex modulus) in comparison to the control bitumen, with ~~this~~
24 ~~modification being~~ more significant ~~modification level reached for the for the maximum NR concentration (-7%~~
25 NRMB). At lower testing temperatures (*i.e.*, higher loading frequencies) the complex modulus master curves
26 show a decrease in the G^* values for increasing NR dosages as compared to those of the control bitumen. While
27 the change of δ with temperature and frequency is generally consistent with that of the G^* master curves, the
28 differences between the three binders are less evident at lower testing temperatures (higher loading frequencies).
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32 The rheological data shown in the G^* and δ master curves is also plotted in the form of Black Space diagrams
33 (Figure 2c) and used as an indicator of rheological changes induced by NR without the need to shift the data
34 based on the time-temperature-superposition-principle (TTSP) [17]. The Black Space diagrams indicated a shift
35 of the curves towards a more elastic behaviour (*i.e.*, lower δ for both NRMBs) as the NR dosage increased,
36 particularly for the higher 7% NR content. For the 3% NR NRMB the level of modification is restricted to the
37 high temperature (*i.e.*, low frequency) domain with the rheological behaviour at low temperatures (*i.e.*, high
38 frequencies) being similar to that of the control bitumen. The Black diagrams also indicated that increased NR
39 content produced a reduction in G^* that substantiates the increased ductile response induced by NR on the
40 binder rheology (discussed further for the low temperature BBR rheological results). Although not shown in
41 Figure 2 for the sake of brevity, the effects of NR modification for the short- and long-term aged binders were
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 7 consistent with those discussed for the unaged binders with the effect of ageing simply indicating an overall
 8 increase in G^* and a reduction of δ values for all the binders as ageing increases. Overall, the comparison of the
 9 rheological parameters presented in Figure 2 substantiated the ability of NR to produce a more elastic response
 10 for the NRMBs as compared to that of the control bitumen. These results suggest an improvement of the
 11 permanent deformation resistance for increased NR content NRMBs due to their increased elastic response at
 12 high temperatures.
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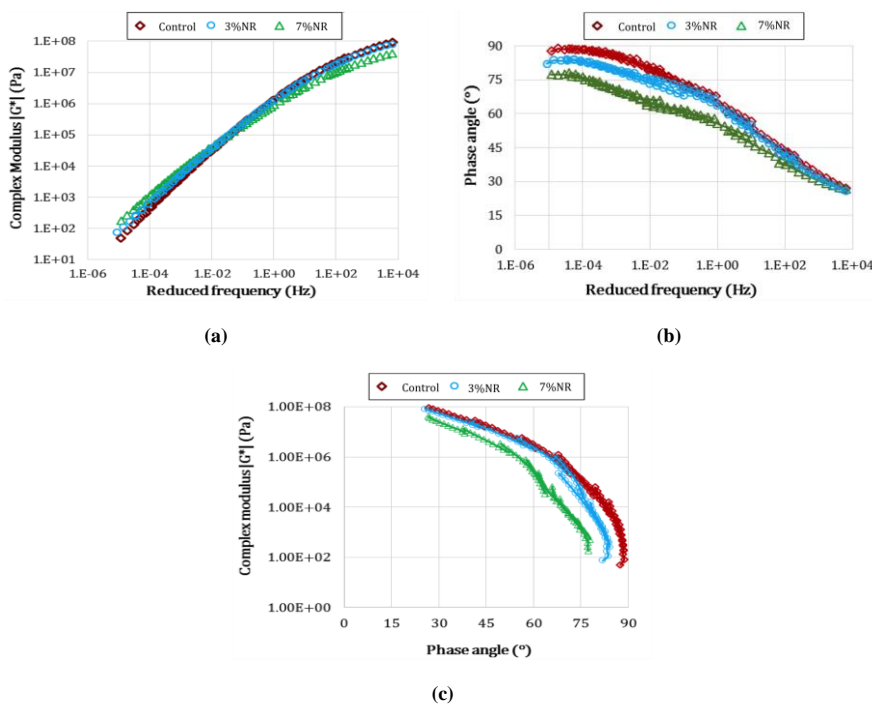
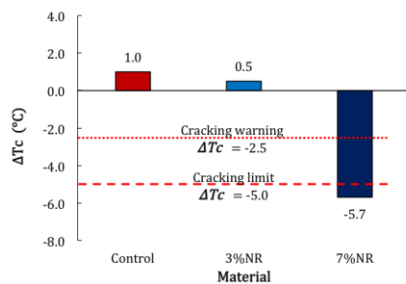


Figure 2. Comparison of the effects of NR on the rheological characteristics in terms of: (a) G^* master curves, (b) δ master curves, and (c) Black-Space diagrams for unaged materials.

3.2. Low-Temperature Effects and Thermal Cracking Susceptibility (BBR)

The S - and m -value comparisons for the two testing temperatures considered in this study are shown in Table 2. From these values, the effect of NR on the low-temperature rheological behaviour showed a reduction of S values (creep stiffness) as NR content increased from the control binder to 3% and 7% NR. However, the effect of NR on the relaxation properties (m -values) suggested a slower relaxation of the thermal stresses of the NRMBs considered in this study in contrast to that of the control bitumen. This means that high NR contents

could induce brittle material response as a result of a reduced ability of these binders to stretch (i.e., lower ductility or loss in relaxation). However, the use of *m-value* does not represent a final indication of poor or good thermal cracking performance, as this parameter is associated with thermal stresses build up and the time required/available by the material to release these, which ultimately depends on the location and climate [34]. In consequence, the fact that the increase in NR content induced lower *m-values* does not imply that these NRMBs would have detrimental performance as this will depend on the weather conditions to which the pavement will be subjected to and it also depends on the *S* values at low temperature, which were found to be lowered by the increase in NR content. As such, if one considers NR application in warmer climates, both *S* and *m-values* do not represent a major risk as these locations are not subject to major thermal stresses with lowest temperatures being typically around 15°C at which block cracking or thermal cracking are not distresses of concern. For applications of NR in low-temperature regions, both BBR parameters and ΔT_c need to be considered to find a balanced performance that satisfies the specifications imposed by the location/climate and the intrinsic material low-temperature response.



From

Figure 3 and Table 2 it is observed that the base bitumen and 3% NRMB satisfied both cracking limits for low-temperature for the ΔT_c criterion, whereas the increase of NR to 7% did not comply and it is therefore expected to have high susceptibility to thermal cracking. This is probably a consequence of the effects of high-polymeric modification of bitumen with NR, which could lead to higher susceptibility of NRMBs to thermal cracking at high concentrations, as previously reported by [35, 36]. In brief, from the rheological characterisation at low-temperatures it was found that: (i) NR induced positive effects on the NRMBs in terms of a reduction of the creep stiffness (*S*) at -18 °C and -24 °C, which is indicative of lower thermal stresses induced by both NR contents considered; (ii) Conversely, the relaxation properties (*m-values*) were negatively affected by increased NR content, which suggests a lower ability of the NRMBs to relax stresses (i.e., lower ductile response) in contrast to the control bitumen and; (iii) in terms of thermal cracking resistance, it was found that modification

of control bitumen with NR content up to 3% would have satisfactory performance on the basis of the ΔT_c failure criteria.

Table 2. Low-temperature properties (S, m-value) and critical temperatures and the corresponding differential temperature ΔT_c .

Material	Stiffness		m-value		Critical Temperature Stiffness, S(c) (°C)	Critical Temperature m-value, m(c) (°C)	ΔT_c (°C)
	(MPa)		-18 °C	-24 °C			
	-18 °C	-24 °C					
Control	537	854	0.242	0.188	-10.497	-11.500	1.003
Control+3%NR	504	790	0.235	0.174	-11.103	-11.607	0.504
Control+7%NR	462	747	0.206	0.155	-12.608	-6.941	-5.667

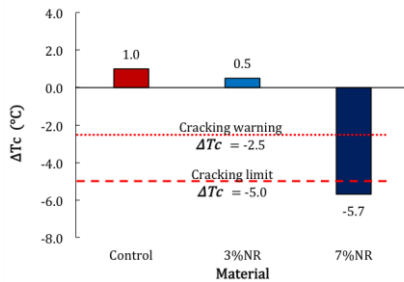


Figure 3. Comparison of the thermal cracking susceptibility for control bitumen and NRMB in terms of ΔT_c values.

4. Performance-Related Effects of NR

4.1. Effects of NR on Fatigue Performance

The characteristic VECD coefficients for the base bitumen (control) and NRMBs are presented in Table 3. The trends observed in the VECD coefficients indicate both the effects of temperature and NR content, which induced differences in the damage response that was captured via the C_0 , C_1 and C_2 coefficients. The results show an increase of C_1 was induced by the increase in NR content, whereas C_2 decreased for increased NR content. These differences substantiate the fact that 7% NRMB presents a higher reduction in initial $|G^*|$ at low damage intensity levels, whereas the reduction is less prominent for higher levels of damage. Furthermore, this

indicates that both NR contents induced positive effects on the damage resistance (captured by lower decrease in the initial $|G^*|$ in comparison to the base bitumen) that turn into higher fatigue cracking resistance as further discussed in this section. Overall, the assessment of the VECD damage coefficients indicated that the largest C_0 , C_1 and smallest C_2 values relate to the NRMB with 7% NR. The temperature effects of 10°C difference on the fatigue performance were also captured as reflected by the changes in the VECD coefficients, the damage intensity curves and the fatigue life curves. The results show that the higher 25 °C test and analysis produced significantly lower C_0 and C_1 values whereas C_2 remained almost unaltered. The damage intensity curves substantiated significant reduction in the stiffness of the materials as a result of the temperature increase and the fatigue life reflected this effect in higher N_f for the higher testing temperature. Overall, it was found that the addition of NR produced NRMBs with higher damage resistance in contrast to the base bitumen.

Table 3. VECD coefficients for control and NRMB at 15 °C and 25 °C.

Material	15 °C			25 °C		
	C_0 (MPa)	C_1 (MPa)	C_2	C_0 (MPa)	C_1 (MPa)	C_2
Control	44.51	0.092	0.434	13.2	0.084	0.424
Control + 3% NR	44.06	0.117	0.381	12.93	0.104	0.383
Control + 7% NR	46.52	0.149	0.344	16.81	0.179	0.299

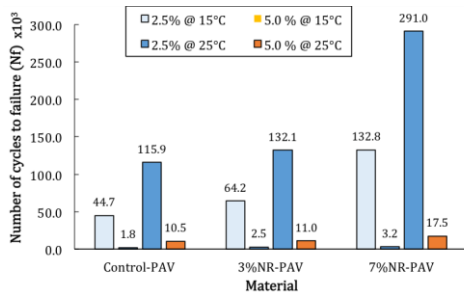


Figure 4. Comparison of fatigue lives for PAV-aged materials at 2.5% and 5% strain at 15 °C and 25 °C.

A comparison of the fatigue lives (number of cycles to reach failure, N_f) are shown in **Figure 4**. As observed previously, temperature and NR content effects were identified. For both temperatures, N_f increased as a function of the NR content, which was consistent for both the low and high strains considered ($\gamma = 2.5\%$ and $\gamma = 5.0\%$). At 15°C testing temperature and 2.5% strain, a 44% increase in fatigue life was found for 3% NR and a 197% increase for 7% NR in contrast to that of control bitumen. At 15 °C and 5% strain, this increase was less prominent (36% for 3%NR and 76% for 7% NR). The effects of increasing the test temperature by 10°C also

produced differences in the fatigue lives, which were reflected by higher N_f values for the 25°C testing temperature for all materials and strains considered. From the macroscopic perspective (i.e., visual inspection of the samples after testing), the fatigue of all the materials exhibited a failure plane that followed a *factory-roof* morphology and a circumferential torsional failure (i.e., growing from the inside to the outside of the bitumen/NRMBs specimens), which is consistent with previous findings by [22] and substantiates an adequate bitumen/plate bonding during testing.

4.2. Effects of NR on Permanent Deformation Resistance

The effects of NR on permanent deformation resistance is presented and discussed in this section in terms of the evolution of accumulated strain over time, the effects of ageing, and the comparison of rutting parameters (non-recoverable creep compliance J_{nr} , and percent recovery R -value, stress sensitivity parameter $J_{nr-diff}$).

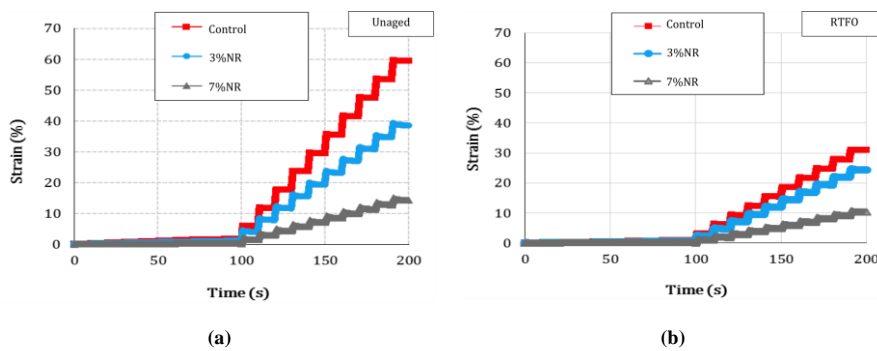


Figure 5. Strain versus time for (a) unaged bitumen, and (b) RTFO aged bitumen, at 60 °C.

The strain and accumulated strain versus time presented in Figure 5 and Figure 6, respectively, show a positive effect of NR on increased resistance to permanent deformation in contrast to the base bitumen. In Figure 5, the initial 100 seconds refer to the 0.1 kPa stress condition and the subsequent steps of cumulative strain are due to the impact of a much higher stress regime (3.2 kPa), which is easily observed and captures the effect of NR modification of the base bitumen. The higher NR contents induced a reduction in the accumulated strain for both NRMBs with 3% NR and 7% NR, which was observed for both unaged and STOA conditions and for both stress levels considered (0.1 kPa and 3.2 kPa). This reduction in accumulated strain highlights a positive influence exerted by NR on bitumen modification in terms of a ~~reduced~~ permanent deformation susceptibility of the NRMBs in comparison to that of control bitumen (Figure 6).

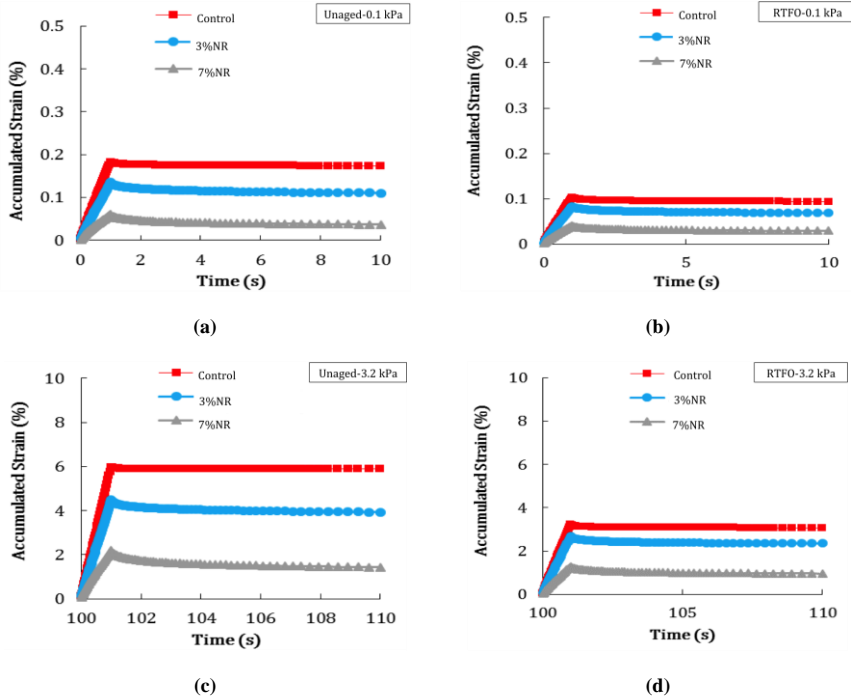


Figure 6. Effects of stress on the accumulated strain for unaged, and RTFO-aged bitumen, at 60 °C. (a) Unaged at 0.1 kPa, (b) RTFO at 0.1 kPa, (c) Unaged at 3.2 kPa, and (d) RTFO at 3.2 kPa.

The effects of NR on the resistance to permanent deformation, assessed in terms of the average percentage recovery (*R-value*) and non-recoverable creep compliance (J_{nr}), indicated that increasing NR content enhanced this material response as compared to that of the base bitumen. Consistently for both unaged and RTFO-aged materials and at both stress levels, it was observed that J_{nr} decreased and the *R-values* increased as the NR content increased. Figure 7 show the effects of STOA ageing, NR content and stress level on the non-recoverable creep compliance (J_{nr}). The change from unaged to STOA produced stiffer materials as captured by the lower J_{nr} values for both control bitumen and NRMBS at 3%NR and 7%NR content. The increase in stress level from 0.1 kPa to 3.2 kPa increased the value of J_{nr} by approximately 10%. Furthermore, the effect of increased NR content showed a clear trend of reduction in the J_{nr} values for both stress levels and ageing conditions, which substantiates the positive influence of NR on this material response.

These parameters provide evidence of the positive effects of NR on the permanent deformation resistance of the modified control bitumen. Similarly, the effects of ageing were also demonstrated based on the J_{nr} and *R-values* shown in Figure 8. Similar to the effect seen for unmodified binders, subjecting the NRMBS to STOA improves

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the rutting resistance of the modified binders meaning that positive observations in terms of permanent deformation resistance held true for both NR content and ageing condition.

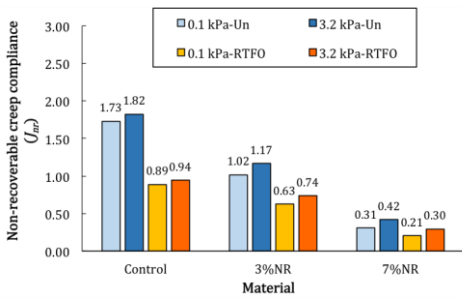


Figure 7. Comparison of the effect of NR and ageing on the permanent deformation, presented in terms of non-recoverable creep compliance (J_{nr}) for unaged materials, and RTFO aged materials.

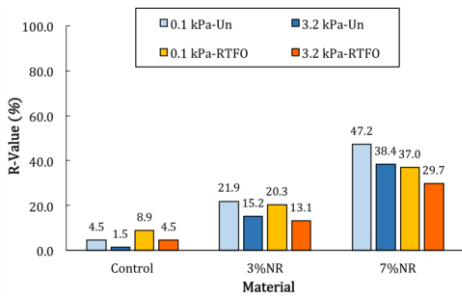


Figure 8. Comparison of the effect of NR and ageing on the permanent deformation, presented in terms of average percentage recovery (R -value) for unaged materials, and RTFO aged materials.

The J_{nr} results are in agreement with those observed for the R -values with higher R -values indicating a higher resistance to permanent deformation. As such, the combined effects of ageing, stress level and NR content are presented in Figure 8, where it becomes clear that ageing and NR content increased the permanent deformation resistance of the binders. Furthermore, as it would be expected for bituminous materials, the increase of stress level induced a substantial decrease of the R -values, indicating the stress sensitivity of the materials with increased NR content. [The reduction in the percent recovery \(%R\) is explained by the possible damaging of the NR crosslink structure \(i.e., polymer network stability\) induced by this short-term ageing protocol, which inevitably affects the permanent deformation response of the NRMBs at both concentrations studied. Under this oxidative condition, the hardened bitumen phase is responsible for the most part of the rheological response.](#)

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Previous research found similar results of dropping recovery values when polymer-modified bitumen blends were subjected to artificial ageing cycles in both RTFO and PAV [35, 36]. Furthermore, the $J_{nr-diff}$ values shown in Figure 9 substantiate the increase in the stress sensitivity induced by increased NR content for the bitumen s considered. However, this is a common effect of high polymer modification of bitumen, which has been reported for the permanent deformation performance of polymer modified asphalt (PMA) mixtures [375, 386]. From these results, however, it is surprising that short-term ageing of the 7% NRMB did have a reduction of the stress sensitivity, which could result from possible errors in rheological data captured by DSR due to the high stiffness gain induced by the high-NR content in contrast to 3% NR or the base bitumen, or it might be related to variability between replicate materials during production of the DSR samples. Therefore, from $J_{nr-diff}$, it might be deduced that higher NR contents led to detrimental effects on the stress sensitivity for both ageing conditions, which needs to be considered for optimum dosage selection in conjunction with other performance-related characterisations to attain a balanced performance for a given NR content. However, based on standard recommendations [397], all the materials fulfilled the criteria as they were all below 75%, which suggests satisfactory stress sensitivity for both NR contents.

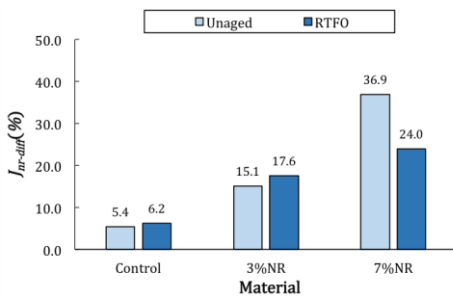


Figure 9. Stress sensitivity for unaged and RTFO-aged materials.

4.3. Influence of NR on the fracture characteristics

The specific total work of fracture (W_t), essential work of failure (W_e), plastic work of failure (βw_p), and critical crack opening displacement ($CTOD$) are presented in this section to discuss the effects of NR on the fracture properties of the modified binders. Prior to the fracture parameters, the plane-stress/plane-strain mixed mode of testing was verified through the correlation between net section stress and ligament length, which indicated a decrease in the net section stress as the ligament length increased. The W_t values for the base bitumen and NRMB at 3% and 7 % are shown in Figure 10 where a higher slope can be observed for 7% NR content in

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7 contrast to the base bitumen and 3% NR. These results indicate an improved yielding capacity and increase in
8 the fracture energy for NRMB with 7% NR content, whereas the 3% NR content did not produce the same
9 extent of influence on the fracture energy and therefore its yielding capacity is reduced [38]. As observed in
10 **Figure 10**, the essential work of fracture for 3% NR content exceeded that of the base bitumen, whereas 7% NR
11 content provided the lowest W_e values. This indicates that the 3% NR content enhanced the resistance to
12 fracture due to a higher ductile strain tolerance in contrast to those of the base bitumen and 7% NR. This finding
13 is consistent with previous research [39, 40]. The former study showed a positive correlation between higher W_t
14 values and β_{w_p} with enhanced fatigue cracking of real pavement sections due to enhanced ductile strain
15 tolerance. The latter study proved that inclusion of RAP to polymer modified bitumen led to unsatisfactory
16 fracture properties, which included decreased W_t and β_{w_p} values.

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18 The plastic work of failure (β_{w_p} values related to the slopes of the curves) is shown in **Figure 10a** and this
19 parameter was used as an indicator of the fatigue cracking resistance of the base bitumen and NRMBs based on
20 recent literature [4942]. It can be observed that for the 7% NR (0.7726), the W_t increases at a higher rate in
21 contrast to that of the base bitumen (0.5112) and 3% NR (0.2765), respectively, which would suggest 7% as the
22 most adequate NR concentration. However, other fracture properties are also required for confirming this
23 observation, thus $CTOD$ and W_e were also determined. The $CTOD$ values are in agreement with those of W_e
24 (**Figure 10b**), which substantiates the fact that NR contents above 3% might lead to deleterious effects on the
25 fracture resistance for the material combinations studied in this paper. This could be the result of NR
26 concentrations reaching a maximum level of yielding capacity and strain tolerance that is below the 7% studied
27 in this paper. This might be due to the physicochemical reaction of higher NR content with the base bitumen due
28 to the higher concentration of polymeric chains in the bitumen matrix that are able to alter the ductile response
29 under high strain concentrations at the testing temperatures considered. [Similarly](#) ~~Furthermore~~, the data
30 presented in Table 4 is consistent with the previous discussion of the fracture parameters in terms of the failure
31 deformation and peak load. These results show that a NR content of 3% leads to improved fracture resistance as
32 a consequence of the lower peak loads and increased deformation. The 3%NR content therefore induced better
33 fracture properties for the materials studied, whereas 7% NR exhibited detrimental effects on the fracture of
34 NRMBs.

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36 [Furthermore, the results observed for the maximum concentration of NR in the NRMBs might be associated by
37 the detrimental side effects produced by the artificial ageing regimes \(i.e., RTFO and PAV\) that are required](#)

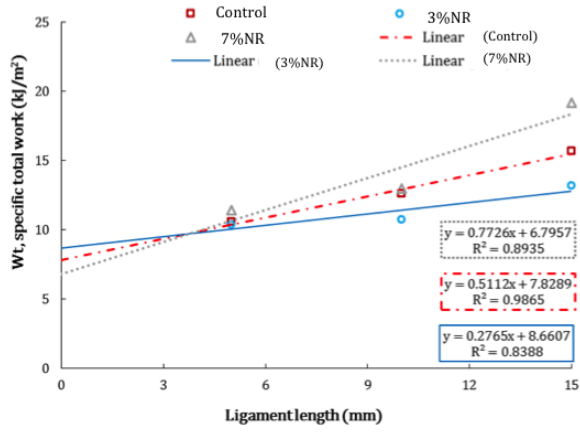
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prior testing for DENT. More precisely, as discussed before, the polymer network or cross-link of the NR molecules seems to be disintegrated by the short- and long-term ageing, which results in stress concentration in the bitumen matrix. Previous research on fracture response of SBS-modified bitumen through DENT testing found that high polymer concentrations (i.e. higher than 2%) tend to have poorer fracture performance in comparison to reasonably low contents of modifiers. In fact, the use of WMA additives such as Fischer-Tropsch (FT), chemical surfactants (CS) and water-based (WB) have the ability to modify the fracture response, which tends to be concentration-dependent and showed improved fracture response at 2% concentration for CS WMA-additive type, but this response was opposite for the FT and WB types with the only exception of 6% WB concentration [43]. Additional research by [44] substantiated a detrimental ductile response of high-polymer modified bitumens with SBS (i.e., 3-5% concentration by weight of base bitumen). In fact, it was found that both in unaged and PAV-aged conditions, the CTOD values for the SBS-modified samples had lower ductile performance (i.e. lower CTOD values), with one single exception to this trends for a SBS-blend (A5), which had the same CTOD value, measured at deformation rate of 50 mm/min, as the base bitumen after long-term ageing protocol. Additional research on PMB-bitumen modification through various concentrations of RAP binder (i.e., 0%, 15%, 25%, and 40%) indicated that the higher concentrations of this bitumen modified negatively affect the intermediate temperature response in terms of the ductile properties measured through DENT and elastic recovery tests [45]. Conversely, [46] found that for low-SBS polymer concentrations the fracture performance after PAV-ageing seems comparable and at some instances superior to that of base bitumen for concentrations ranging from 1.2% to 1.8% by weight of base bitumen. It was also found that the superior response at RTFO ageing conditions is not equaled by that of PAV-aged materials, indicating a detrimental effect of artificial ageing on the fracture response for these material combinations.

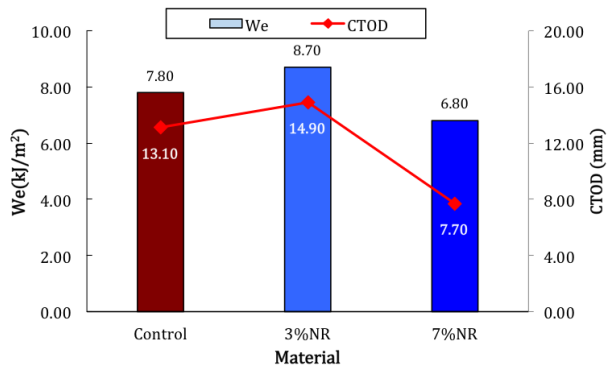
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(a)



(b)

Figure 10. (a) The specific total work of fracture (W_t) and plastic work of failure (βw_p), (b) essential work of failure (W_e), and critical crack opening displacement (CTOD) for neat bitumen and NRMBs.

Table 4. Summary of peak load and deformation to failure for neat binder and NRMBs.

Material	DENT insert length (mm)					
	5		10		15	
	Peak Load (N)	Failure Deformation (mm)	Peak Load (N)	Failure Deformation (mm)	Peak Load (N)	Failure Deformation (mm)
Control	30	34.8	40	67.2	46	118
Control + 3% NR	29	47.2	37	80.6	42	134
Control + 7% NR	44	32	57	56.2	66	94

5. Conclusions

The laboratory investigation described in this paper assessed the effects of NR on the rheological and performance-related characteristics of modified bitumens (NRMB). This was achieved through a comprehensive laboratory programme that incorporated the evaluation of NR effects on bitumen rheology and the evaluation of fatigue cracking, thermal cracking, permanent deformation and fracture properties of the NR-modified binders NRMB. Two NRMB blends were manufactured with 3% and 7% NR by mass of the final modified binder using a 40/60-penetration grade base bitumen (control). The test methods included: (1) Dynamic Shear Rheometer (DSR) master curves and Black Space diagrams together with Bending Beam Rheometer (BBR) tests for rheological assessment over a wide temperature range; (2) Linear Amplitude Sweep (LAS) tests for assessing fatigue at intermediate temperatures; (3) Multiple Stress Creep Recovery (MSCR) tests for permanent deformation at high temperature; and (4) the evaluation of fracture properties at intermediate temperatures using the Double Edged-Notched Test (DENT) test. Based on the experimental results and analysis, the following main conclusions can be drawn:

- 1) The effects of NR on the bitumen rheology produced changes in the rheological parameters (G^* and δ) that were captured via the master curves and Black Diagrams. It was observed that increased NR content produced significant increase of the stiffness of NRMBs (higher G^*) that was particularly pronounced at high testing temperatures. Similarly, the decrease of δ values are consistent with the results of G^* and the Black Diagrams showed a shift of the NRMBs curves towards a zone related to more elastic response in

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7 comparison to the base control bitumen. Overall, these results indicated a substantial level of modification
8 of the base bitumen that relates to more elastic NRMBs with higher resistance to permanent deformation as
9 further substantiated with MSCR testing.

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12 2) The rheological characterisation at low-temperatures found that: (i) NR induced positive effects on the
13 NRMBs in terms of a reduction of creep stiffness (S) at -18°C and -24°C , which is indicative of lower
14 thermal stresses being induced in the NRMBs; (ii) The relaxation properties (m -values) were also affected
15 by increased NR content although the m -values showed a lower ability of the NRMBs to relax stresses in
16 contrast to the control bitumen and; (iii) In terms of thermal cracking resistance, it was found that a NR
17 content of 3% would have satisfactory performance on the basis of the ΔT_c failure criterion. From a
18 practical point of view, these results imply some restricted use of NR in cold regions or those with
19 significant day/night temperature gradients and supports its use in tropical regions.

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22 3) The fatigue performance of the NRMB blends assessed by means of the LAS testing indicated that NR
23 increased the fatigue life of the base bitumen by a significant extent at the intermediate temperatures
24 considered (15°C and 25°C). This impact on fatigue life was consistent for both strain levels ($\gamma = 2.5\%$ for
25 thin pavement structures and $\gamma = 5.0\%$ for thick pavement structures) but it was more prominent at the
26 lower strain level. In general, the results showed a higher number of cycles to reach failure (N_f) for 7%
27 NRMB as compared to those of control bitumen and 3% NRMB. These findings were consistent for the
28 temperatures considered, which suggests positive effects of NR on bitumen modification with regard to
29 fatigue cracking.

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32 4) Positive effects of NR on the permanent deformation resistance were observed indicating an increased
33 resistance for both NR contents considered. The maximum resistance was reached for the 7%NR content as
34 it related to the highest R -values and lowest J_{nr} parameter for all combinations studied. These results
35 substantiated the positive influence of NR on the rutting response for the NRMBs when compared to that of
36 base bitumen for both unaged and RTFO-aged materials. [However, some detrimental effects on the polymer
37 structure \(crosslink of the NR molecules\) of the short-term ageing regime did likely occur as observed by
38 slightly reduced R-values for the RTFO-aged materials.](#) The stress effects on the NRMBs resistance to
39 rutting indicated that the improvement of this material response was predominant for higher stresses (3.2
40 kPa) in contrast to the lower stress considered (0.1 kPa). Nevertheless, the stress sensitivity ($J_{nr-diff}$)
41 increased with NR addition to the base bitumen, reaching a maximum for 3% NR content for both ageing
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7 conditions, which is expected as a result of high-content polymeric modification of bitumen, such as that
8 provided by Styrene-Butadiene-Styrene (SBS) in polymer modified mixtures (PMA).

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- 10 5) The NRMBs exhibited a limited fracture response as observed with the DENT test results in terms of the
11 fracture properties of W_i , W_e , βW_p , and $CTOD$. In practical terms, based on the material combinations
12 considered, these results suggest that NR content should not exceed 3% when attempting to minimize
13 fatigue cracking and improve fracture performance at the micro scale. Similarly, other NR dosages should
14 be explored with different base bitumen types than those included in this investigation for further validation
15 of fracture properties.
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20 Overall, the evidence obtained through the experimental programme and materials tested substantiates some
21 performance-related benefits of NR and its suitability for applications in warmer countries, such as Thailand or
22 those encountered in South East Asia where NR is produced and widely available, for which where the rutting
23 distress and fatigue of fatigue and permanent deformation of pavements represents the major challenges for
24 mixture design, performance and durability. Furthermore, a possible increase in the current limits of NR use for
25 road infrastructure is envisioned as a practical application of this research study, when complemented with
26 additional future studies. Similarly, these results encourage use of NR in other countries aiming at promoting
27 sustainable road infrastructure.
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31 Some recommendations for future work include:

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- 33 • Although storage stability of the samples has not been studied by this research work, previous work by
34 [47] substantiated that NR contents of up to 6% exhibit stable performance for use at high
35 temperatures. Furthermore, [48] demonstrated with factual evidence that contents up to 9% can have
36 satisfactory storage stability and indicated that this property is largely dependent on the NR
37 concentration. Similar findings were reported by [49], who reported satisfactory storage stability for
38 blends subjected to conditioning for 72 hours at 163°C. Therefore, future studies might add value to the
39 characterization of NRMB by incorporating the storage stability results.
 - 40 • In terms of the complexity of bitumen modification through NR in comparison to other polymers, our
41 experience and that reported by previous research indicates [50, 51] the following: Overall, it is harder
42 to mix NR latex than other polymers because of its liquid nature in contrast to others being capsules or
43 grinded materials (solid-like). Furthermore, the fact that NRL contains “water” in the latex, and have
44 high ammonia content makes it hard to produce blending due to the physical effects (i.e., frothing and
45 foaming), in addition to the strong smell produced by the ammonia. For these reasons, the liquid NRL
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7 is gradually added to the base bitumen at small portions, which allows for reducing the air bubbles and
8 evaporation. This is aided by manual stirring to minimize the foaming of the rubber to reach the top
9 head of the mixing machine. Certainly, this procedure means more time consumption than other
10 polymers but it allows for homogeneous blend if carried out cautiously by future researchers. ▲

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11 12 13 14 15 16 17 **Acknowledgements**

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