



Critical assessment of new polymer-modified bitumen for porous asphalt mixtures

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ABSTRACT

New experimental polymer-modified bitumen with a high-vinyl content polymer was fabricated for porous asphalt (PA) mixtures. The bitumen with maximum stability was achieved using storage stability, gelation criteria and physical bitumen tests. A dynamic shear rheometer was used to compare the complex modulus, number of fatigue cycles, yield stress, and non-recoverable creep compliance of the experimental bitumen with a reference virgin bitumen 50/70 and a PMB 45/80-65 binder. PA mixtures were also designed to analyze the abrasion resistance and binder drainage characteristics. It was concluded that the experimental bitumen with 4.5% polymer content showed higher elastic response, better fatigue resistance, and improved rutting behavior than the reference PMB. PA mixtures with the new experimental bitumen exhibited higher abrasion resistance, but underwent higher binder drainage, which was addressed by the incorporation of aramid pulp and glass-hybrid fibers.

1. Introduction

Increasing traffic load has led to requirements of more durable asphalt mixtures. In addition to traffic, adverse weather conditions reduce the serviceability of the pavement. These problems worsen in porous asphalt (PA) mixtures, due to their exposed structure that facilitates excessive oxidation of bitumen [1,2]. Due to combined effects of oxidation, climatic conditions, and vehicle loading, raveling constitutes the predominant distress in PA pavements [3,4,5]. To improve raveling resistance, polymer-modified bitumen (PMB) can be used to enhance the interaction between binder and aggregates without causing segregation of material [6,7,8]. Therefore, PA mixtures with PMB are less susceptible to abrasion due to their greater adhesion and having fewer fissures in binder [9,10]. PMB has very high temperature stability so there is noticeable reduction in the rut depth [11,12]. Additionally, PA mixtures with PMB binder have more elasticity, which improves the low temperature crack resistance [10,13].

According to Shell bitumen handbook (sixth edition) [14], plasticity interval is “the temperature range between the measure of high temperature performance (e.g. the softening point or criteria based on the

complex modulus in the SHRP PG grading approach) and the low temperature measure of performance (e.g. a brittleness point or limiting stiffness value determined by the BBR)”. The polymer modification enhances the plasticity interval based on the polymer properties, polymer content, nature of base bitumen and degree of modification [14]. According to studies, if the polymer content is less than 2.5% then the properties of base bitumen dominates the resultant binder properties with higher viscous components. If polymer content is 5%, then the resultant bitumen exhibits an equal contribution from polymer and bitumen. At polymer content higher than 7.5%, polymer phase is more dominant and the bitumen acts like rubber [15,16].

Bitumen is most commonly modified with elastomers [17]. Elastomers resist deformation by stretching and then recover the original shape (shell). Styrene-Butadiene-Styrene copolymers (SBS) structure shown in Fig. 1, are among the elastomers frequently used to modify bitumen [7,17–23]. They form a three-dimensional network by physical crosslinking of molecules [14]. The butadiene block is highly elastic while the styrene block is stiff and shows good compatibility with the aromatic fraction of bitumen [24]. SBS polymers provide high elasticity, low temperature cracking [25], better rutting behavior at high

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temperatures [26], high viscosity [15], high softening point and a low penetration value [13,27]. However, use of this polymer is critical, because of some drawbacks such as lack of storage stability because of low thermodynamic compatibility with bitumen, and high rate of polymer ageing due to ruptures in polymer chains caused by UV radiation (CH bonds) [15,21]. Styrene is easily dissolved in bitumen due to the presence of aromatic rings whereas butadiene has simple carbon hydrogen bonds (CH bonds). The presence of aromatic rings in bitumen enables swelling to up to nine times its initial volume [24,28]. High-vinyl content SBS (>35%) improves the viscosity and improves the bond between polymer and bitumen [16] and has a lower rate of ageing. According to Singh et al., 2018 [21], it was found from gel permeation chromatography (GPC) the high-vinyl SBS polymer reduces the rate of erosion compared to that with lower vinyl content. High-vinyl SBS polymer may reduce the issues relating to storage stability, high viscosity, and degradation of SBS polymers [16,21].

To characterize a new bitumen and analyze the rheological parameters of modified bitumen, the temperature and frequency sweep test using dynamic shear rheometer (DSR) is most commonly used [12,29–33]. The time sweep test is used to analyze fatigue characteristics in asphalt material [34]. However, this test is time consuming and so shear stress tests and amplitude sweep tests, such as the Linear Amplitude Sweep test (LAS), can be used. The LAS test has gained popularity as many studies state that its results can estimate the fatigue characteristics of the binder in the field [35,36]. The LAS test involves complex mathematical modeling, and it is difficult to account for non-linear viscoelastic properties [31,35]. The BYET (Binder Yield Energy Test) with monotonic loading has shown good correlations with LAS test results and fatigue of the asphalt mixtures [31]. Nevertheless, many researchers have questioned the validity of BYET because of multi-peak behavior [36,37]. For the analysis of rheological performance of bitumen at high temperatures, Multi-Stress Creep and Recovery Test (MSCRT) has been widely performed by many researchers [31,38–42]. Vischer et al. [43] found in their study that this test accurately simulates the realistic rutting in the asphalt mixtures and the non-recoverable creep compliance (J_{nr}) parameter indicates the sensitivity to rutting. Therefore in this study, temperature and frequency sweep test, LAS, BYET and MSCRT were performed.

Objectives and methodology

The main objective of this research is to develop a new bitumen using high-vinyl content SBS polymer, catering to the requirements of PA mixtures. The bitumen developed was analyzed using conventional physical tests such as penetration, softening, viscosity, storage stability, gelation; and rheological tests such as temperature and frequency sweep test, LAS, BYET, and MSCRT. The second objective is to investigate the laboratory performance of PA mixtures manufactured with the new experimental bitumen, comparing it with conventional virgin bitumen and a commercial polymer-modified bitumen. Finally, the suitability of the new bitumen combined with aramid and glass fibers was also assessed.

This study was conducted in three phases:

Phase 1. *Preparation of the new experimental bitumen*: Tests were carried out and the experimental procedure was adopted for the fabrication of new experimental bitumen.

Phase 2. *Rheological testing*: Tests were carried out to assess the rheological characteristics of the new experimental bitumen performance compared to virgin bitumen 50/70 and a commercial PMB 45/60–85.

Phase 3. *PA mixture testing*: Comparison of PA mixtures prepared with the new experimental bitumen with the commercial PMB 45/60–85 and virgin bitumen 50/70, including the study of the effect of fibers in PA mixtures combined with the new experimental binder.

2. Materials and methods

2.1. Preparation of the new experimental bitumen

New experimental bitumen was fabricated using the raw materials – virgin binder (70/100), high-vinyl content styrene-butadiene copolymer (Table 1) and Sulphur as a cross linker.

The fabrication of experimental bitumen is done based on: conventional tests i.e. penetration (EN 1426:2015), softening point (EN 1427:2015) and viscosity tests (EN 13,302 at temperatures 135 °C, 150 °C and 175 °C); and tests specifically used with PMB i.e. storage stability (ASTM D7173 or EN 13399) and gelation tests (laboratory procedure).

2.1.1. Storage stability and gelation tests

The thermodynamic compatibility between polymer and bitumen is limited, due to their chemical structure. When polymer-modified bitumen is prepared by physical mixing, the polymer tends to separate during storage at high temperatures, giving rise to coarse phase dispersion on cooling, which results in the formation of a polymer-rich phase swollen by the aromatic compounds of the bitumen at the top, and asphaltene-rich phase with no polymer at the bottom. These phenomena can be controlled by using chemical cross-linkers, like Sulphur or other reactive components [14,21,44]. In storage stability test, the bitumen fabricated is poured carefully to avoid incorporation of air bubbles in the tubes of height 100 mm to 120 mm. The tubes are kept in the oven at a temperature 180 °C for 3 days, and then cut into three equal parts. Later, the samples from the top and the bottom of the tube are tested for their penetration and softening point [45]. The difference between the penetration and softening point of the samples from top and bottom should not be more than 9 (0.1mm) and 5 (°C) respectively for PMB 45/80-65 according to EN 14023.

Chemical crosslinking as a strategy to attain storage stability must be carefully controlled, since overdosing of Sulphur may lead to gelation of the modified bitumen which may result in serious problems in the bitumen plants. During laboratory small scale PMB manufacture, the Weissenberg effect (phenomenon occurring when an elastic liquid climbs up during stirring and wraps itself completely around the rotating rod) [24] has been interpreted as the first indicator of gelation risk. In gelation test, the modified bitumen is kept in an oven at 180 °C for 7 days and visual inspection is done every 24 h for any signs of gelation.

2.1.2. New experimental bitumen fabrication procedure

New experimental bitumen was fabricated using the raw materials – virgin binder (70/100), high-vinyl content styrene-butadiene copolymer (Table 1) and Sulphur as a cross linker. The objective was to attain a stable new experimental bitumen with the highest polymer content, and an iterative process divided into trials involving different procedures and polymer dilutions. The process consisted of manufacturing a high concentration masterbatch (7.5% SBS) and then creating dilutions to reach different polymer concentrations in the range from 3% to 7%. The experimental procedure adopted for the fabrication of bitumen is given in Fig. 2. Dispersion of the polymer was carried out using the Silverson high shear device and further dilution was done at low shear using a Palas device (Fig. 3). The dilutions were prepared to find out the phase

Table 1
Typical values of polymer used in the study (Kraton D0243).

Test	Standard	Typical Value
Tensile strength (MPa)	ISO37	2
Specific gravity	ISO 2781	0.94
Vinyl content (%)	KM03	> 35
Hardness (shore A, 10sec)	ISO 868	70
Volatile matter (%m)	KM 04	≤ 0.3
Ash (%m)	ISO 247	0.2–0.5

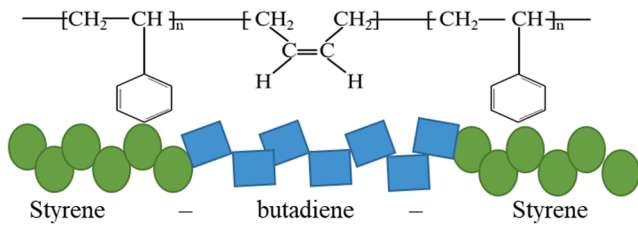


Fig. 1. Structure of SBS polymer.

inversion point from being bitumen dominant to being polymer dominant. Dispersion, chemical reaction, and dilutions of the masterbatch were carried out at the high temperature of 180 °C. After fabrication, the storage stability & gelation tests were performed both on the masterbatches and dilutions and the results are shown in Table 2. As can be seen, with higher polymer contents (higher than 4.5%), the resulting bitumen failed the gelation and storage stability tests and so were not stable. However, the dilutions passed the two tests, which suggests the bitumen was stable at lower polymer contents. Stability was achieved due to an adequate ratio of polymer/chemical crosslinker. Another explanation may be the phase conversion: at low polymer contents, the binder is bitumen dominated, however on increasing the polymer content, the bitumen becomes polymer dominated. In addition to this phenomenon, polymer content and source of bitumen can also affect the stability of the bitumen.

The optimum properties were obtained for 4.5% polymer concentration, hereafter named experimental bitumen (abbreviated as EXPBIT). Table 3 presents the results of the test performed on the experimental bitumen according to the EN: 14023 guidelines (for polymer modified bitumen) and it passed the requirements for PMB 45/80–65 bitumen.

2.2. Rheological characteristics of experimental bitumen

In the second phase of the study, the rheological properties of the new experimental bitumen were analyzed in comparison to conventionally used binders in PA mixtures: virgin bitumen 50/70 (abbreviated as VIRBIT) and commercial polymer-modified bitumen PMB 45/80–65 (abbreviated as PMB). Table 3 shows the properties of the three binders used in the study.

2.2.1. Temperature and frequency sweep test

Dynamic shear rheometer can be used to determine the stiffness and the phase angle of the bitumen. The rheological parameters such as complex modulus, phase angle, and storage modulus were computed by using temperature frequency sweep tests. The specimen is subjected to a strain of 1% to maintain the specimen within the linear visco-elastic range. In this study, the stiffness and phase angle were determined at every temperature in increments of 10 °C from 10 °C to 80 °C, to compute the stiffness at a wide range of frequencies. The distance between the two plates was maintained at 2 mm with the 8 mm spindle, which was used for temperatures 10 °C to 40 °C. For higher temperatures of 40 °C to 80 °C, a 1 mm gap and spindle of diameter 25 mm was used. The tests were performed at eight frequencies from 0.1 Hz to 10 Hz. The mastercurves of complex modulus and phase angles can be plotted using the time–temperature superposition principle. Some researchers have questioned the applicability of the time–temperature superposition principle (TTP) on polymer-modified bitumen [17,46,47,48]. While others have applied TTP for the analysis of PMBs [49–52]. In a study [17], it was found that for unaged PMB, at low polymer contents and low temperatures the TTP is valid as the bitumen phase dominates. However, at high temperatures, the polymer phase dominates and due to loss of its structure, the applicability of TTP can be

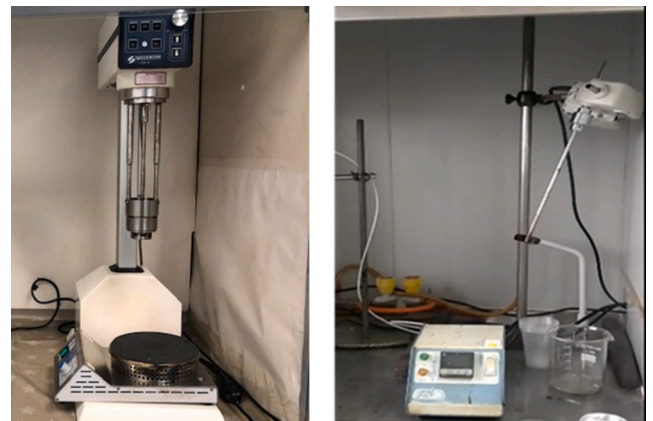


Fig. 3. a. Silverson machine for high shear mixing; b. Palas device for low shear mixing.

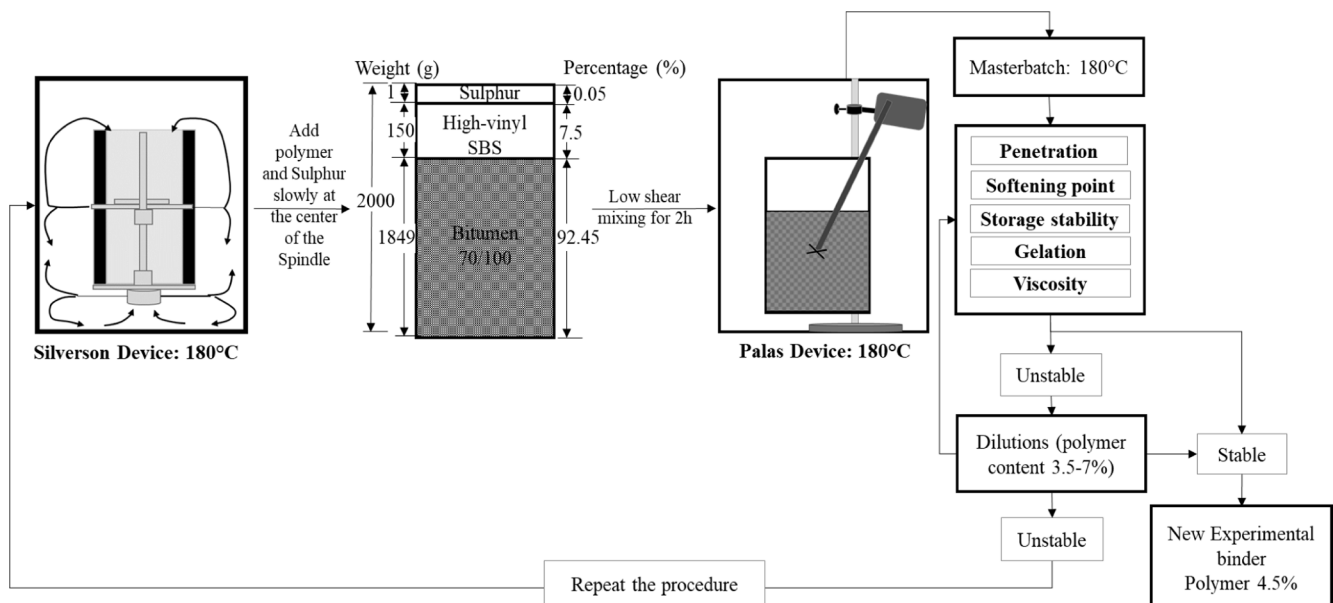


Fig. 2. Preparation of experimental binder.

Table 2
Properties of bitumen obtained in Trials 1, 2 and 3.

Trials	Polymer Content (%)	Penetration point (0.1 mm)	SofteningPoint (°C)	Penetration index	Storage Stability	Gelation
Masterbatch 1	7.5	47	83.1	4.57	Failed	Failed
Masterbatch 2	7.5	48	83.4	4.66	Failed	Passed
Masterbatch 3	7.5	45	84.2	4.6	Failed	Failed
Dilution	7	50	87.6	5.3	Failed	Failed
Dilution	6	48	76.8	3.77	Failed	Failed
Dilution	4.5	54	68.4	2.77	Passed	Passed
Dilution	4.5	49	69.3	2.66	Passed	Passed
Dilution	4.3	55	68.3	2.81	Passed	Passed
Dilution	4.2	51	64.6	1.95	Passed	Passed
Dilution	4	50	62.8	1.57	Passed	Passed

Table 3
Properties of the reference binders for PA mixtures: VIRBIT and PMB.

Properties	Standards	Virgin bitumen 50/70 (VIRBIT)	PMB 45/80-65 (PMB)	Experimental bitumen (EXPBIT)
Polymer content (%)		–	–	4.5
Penetration, (0.1 mm)	EN 1426	57	55	52
Softening Point (°C)	EN 1427	51.6	74.1	67.6
Penetration Index	EN 12591-Annex	–0.50	3.74	3.63
Cohesion (J/cm ²) 5 °C	EN 13,588			6.27
Density (g/cm ³)		1.035	1,028	
Elastic Recovery at 25 °C (%)	EN 13,398	–	92	73
Frass Point (°C)		–13	–	–
Viscosity at 100 °C (MPa)		–	23,099	–
Viscosity at 135 °C (MPa)		–	1951	1471
Viscosity at 150 °C (MPa)	EN 13,302	–	924	737
Viscosity at 175 °C (MPa)		–	–	247
Storage stability	EN 13,399	–	–	Passed
Gelation	–	–	–	Passed

questioned. Therefore, in this study, considering that polymer content is low to medium (lower than 5%), it is assumed that TTP is valid. The mastercurves were prepared by shifting the data to a reference temperature of 20 °C. Equation (1) shows the sigmoidal function used to fit the data.

$$\text{Log}G' = \delta + \frac{\alpha}{1 + e^{\beta + \gamma(\log a_T + \log g)}} \quad (1)$$

Where, a_T is a shifting factor, and $\delta, \alpha, \beta, \gamma$ are fitting parameters determined using the least square method.

2.2.2. Linear Amplitude Sweep (LAS) test

The Linear Amplitude Sweep (LAS) test, proposed by Hintz et al. [35], is an accelerated damage test performed at 25 °C to assess the resistance to deformation of binder. This test method employs cyclic loading, increasing the load amplitudes linearly. The gap was kept 2 mm and spindle of 8 mm diameter is used. This test was performed according to AASHTO TP-101. The temperature of 25 °C eliminates the flow behavior [38]. The two main steps of this test are:

Frequency sweep: The frequency is increased from 0.2 to 30 Hz at 0.1% shear strain amplitude, this is done to obtain rheological properties within the linear visco-elastic range. A graph is plotted of storage modulus versus frequency. Then, the parameter B is computed from the slope of the graph (Equation (2))

$$\alpha = \frac{1}{m} \quad (2)$$

Amplitude sweep: The amplitude of shear strain is increased from 0.1 to 30%. This is done to assess the damage to the specimen. The parameter A (Equation (8)) is computed by visco-elastic continuum damage (VECD) theory. The damage accumulation and integrity parameter are computed as given in Equation (3) and (4) respectively.

$$D(t) \approx \sum_{i=1}^N [\hat{\alpha} \gamma_o^2 (C_{i-1} - C_i)]^{\frac{1}{1-\alpha}} (t_i - t_{i-1})^{\frac{1}{1-\alpha}} \quad (3)$$

Where, γ_o = applied strain for a given data point, percent
 $|G^*|$ = Complex shear modulus, MPa
 $\alpha = \frac{1}{m}$ (m is the slope of the graph of storage modulus versus frequency)
 t = testing time, second

where $C(t)$ is integrity parameter, $= \frac{|G^*|_{(t)}}{|G^*|_{\text{initial}}}$ (4)

Where, $|G^*|_{(t)}$ is the initial value of $|G^*|$ at time (t) and $|G^*|_{\text{initial}}$ is the initial value of $|G^*|$.

The relationship between C(t) and D(t) is calculated using Equation (5).

$$C(t) = C_0 - C_1 D^{C_2} \quad (5)$$

With $C_0 = 1$, the initial value of C and C_1 and C_2 = curve-fit coefficients calculated through the power law adapted from Hintz, et al., in the form shown in Equation (6):

$$\text{Log}(C_0 - C(t)) = \log(C_1) + C_2 \log(D(t)) \quad (6)$$

A, B parameters are used to compute the total number of the cycles to failure (N_f) using Equation (7).

$$N_f = A.(\gamma_{max})^{-B} \quad (7)$$

Where, $A = \frac{f(D_f)}{(1+(1-C_2)/\alpha)(\pi C_1 C_2)^\alpha}$
 $D_f = \frac{(C_0 - C_{atpeakstress})^{\frac{1}{C_2}}}{C_1}$
 f = loading frequency,
 $k = 1 + (1 - C_2)\alpha$,
 $B = 2\alpha$
 γ_{max} = the maximum expected binder strain for a given pavement structure.

2.2.3. Binder Yield Energy Test (BYET)

In the Binder Yield Energy Test (BYET), a constant monotonic shear loading of the specimen at constant shear rate is used to compute the fracture cracking. The main objective is to test the sample using a constant shear rate until the sample yields and peak shear strength is obtained. This test is conducted according to AASHTO TP 123 using the dynamic shear rheometer device with a 2 mm gap and 8 mm spindle at 25 °C temperature. The constant strain rate of 2.315% s⁻¹ is applied for

30 min until 4167% strain is achieved by the specimen. A stress–strain graph is plotted where the peak indicates the maximum yield stress and the area under the curve up to the strain corresponding to maximum yield point indicates the binder yield energy.

2.2.4. Multi Stress Creep and Recovery Test (MSCRT)

The Multi Stress Creep and Recovery Test (MSCRT) is performed to compute the stress sensitivity of a bitumen at the selected temperature. This test was performed according to EN: 16,659 with a 1 mm gap using a 25 mm spindle at 60 °C. In this test, specimens were subjected to a constant stress of 0.1 kPa for 1 s followed by 9 s rest, the process was terminated after 10 cycles. The percent recovery and non-recoverable creep compliance of the bitumen were computed and were averaged for the ten cycles to obtain the non-recoverable creep compliance (J_{nr}) and the percentage of recovery (%R) at the stress levels 0.1 kPa and 3.2 kPa. The parameter J_{nr} indicates the sensitivity of bitumen towards permanent deformation under repetition of applied stress (Equation (8)). The parameter %R (Equation (9)) indicates the ability of bitumen to recover from deformation. It also estimates the degree of polymer modification in a binder. Equation (10) enables the calculation of the stress sensitivity of the binder.

$$\text{Non-recoverable creep compliance } (J_{nr}) = \frac{\text{non-recovered strain}}{\text{applied stress}} \quad (8)$$

$$\%R = \frac{\text{recovered strain}}{\text{Total strain}} \quad (9)$$

$$J_{nr-diff} (\%) = \frac{J_{nr3.2} - J_{nr0.1}}{J_{nr0.1}} * 100 \quad (10)$$

Where, %R is the percentage recovery, $J_{nr-diff}$ is the percentage of variation of the non-recoverable creep compliance value corresponding to stress level 3.2 ($J_{nr3.2}$) compared with the non-recoverable creep compliance value corresponding to stress of 0.1 kPa ($J_{nr0.1}$).

2.3. Porous asphalt mixtures

2.3.1. Sample preparation

The mixtures were prepared according to the PG-3 Spanish technical guidelines [53], with the gradation given in Table 4. The samples prepared in this study are described in Table 5. The density of fiber and its tensile strength are included in Table 5, the rest of the properties of fibers being detailed in previous studies on aramid pulp and glass hybrid fibers [54]. The aggregates were pre-heated at 180 °C for 6 h and bitumen is heated for 2 h (virgin bitumen at 155 °C and polymer-modified bitumen and experimental bitumen at 165 °C). The fibers are commonly used as additives in the asphalt mixtures [55–58]. The fibers were added as received from the manufacturers in aggregates, mixed thoroughly for approximately 30 s, and then the bitumen was mixed thoroughly with the fiber-aggregate mixture. The specimens were compacted in Marshall mould at 50 blows on each face (EN 12697–34) and conditioned at room temperature for one day prior to testing.

2.3.2. Porous asphalt mixture tests and statistical analysis

Air voids refer the total air void content, however there is a portion of voids filled with bitumen that do not participate in the transmissibility of the water. The PA mixtures allow water through their structure due to the presence of high number of interconnected air voids. The total air void content was computed based on EN 12697–8 and the interconnected air void content were estimated from permeability of the

Table 4
Aggregate gradation of PA mixtures.

Sieve size (mm)	22	16	8	4	2	1	0.5	0.25	0.063
Passing (%)	100	100	54.5	19.1	14.1	10.3	7.8	6.3	5.0

Table 5
Properties of mixtures prepared in the study.

Mixture types	Composition	Density of fiber (g/cc)	Tensile strength (GPa)	Fiber content (% by wt. of mix)
VB4.5	Virgin bitumen 50/70	–	–	–
PMB4.5	PMB45/80–65	–	–	–
EXP4.5	Experimental binder	–	–	–
EXPST0.3	Experimental binder and glass-cellulose hybrid fiber	0.35–0.55	>1	0.30
EXPST0.5	Experimental binder and glass-cellulose hybrid fiber	0.35–0.55	>1	0.50
EXPPULP0.03	Experimental binder and aramid fiber	1.44	2.7–3.6	0.03
EXPPULP0.05	Experimental binder and aramid pulp fiber	1.44	2.7–3.6	0.05

mixtures.

The Cantabro test measures the abrasion resistance of the PA mixtures. In this study, the test was conducted under dry conditions (according to EN 12697–17) as well as wet conditions (according to NLT 362/92). In this second scenario, the samples are kept in water for 24 h at 60 °C, and after that, they are kept for 24 h at 25 °C before performing the test. The permissible limits for abrasion resistance for the highest traffic category according to PG-3, article 543 are 20% and 35% for dry and wet conditions respectively. The particle loss is calculated according to Equation (11).

$$\text{Particle loss} (\%) = \frac{\text{initial mass}(\text{g}) - \text{final mass}(\text{g})}{\text{initial mass}(\text{g})} * 100 \quad (11)$$

Pertaining to high air voids, the binder in the PA mix may drain downwards due to the action of gravity. Therefore, draindown tests were performed. According to EN 12697–18:2017, the binder drainage was computed by keeping uncompacted PA mixtures at 180 °C for three hours in a wire mesh basket and calculating the change in weight. The recommended binder draindown in PA mixtures should be less than 0.3% [59,60].

The statistical analysis is important to assess the significance of the difference among the various samples. The results of porous asphalt mixture tests were first checked for normality by performing the Anderson Normality test. Afterwards, the results following normal distribution were compared based on two-sample student’s parametric tests. While the non-parametric Mann-Whitney tests were performed on test results that did not follow normal distribution. A level of significance (α) of 0.05 was used which indicates a 5% risk of concluding that a difference exists when there is no actual difference. If the mixtures share a common group letter, this indicates that their means are not statistically different from each other.

3. Results and discussion

3.1. Rheological testing

3.1.1. Dynamic Shear Rheometer

The data obtained from temperature and frequency sweep tests was used for the calculating the complex modulus and phase angle of the three binders (experimental, virgin, and reference PMB 45/80–65). Mastercurves of complex modulus and phase angle are provided in Fig. 4a and a black diagram in Fig. 4b, respectively. At low frequencies, a clear difference between the complex modulus and phase angle of three binders can be observed. The PMB showed the highest complex modulus that means that it is stiffer at lower frequencies as compared to the other two bitumen. EXPBIT showed higher complex modulus than VIRBIT indicating greater stiffness, however at intermediate frequency range both mastercurves indicated similar stiffness of bitumen. Eventually, at higher frequencies, the complex modulus of PMB also coincided with the other two bitumens which signifies that at higher frequencies there was no significant difference in the stiffness among the three bitumens. However, concerning phase angle, the mastercurves of VIRBIT exhibited higher phase angle that indicated lower elasticity and high deformation ability. On the other hand, the phase angle mastercurves of both the modified bitumens (PMB and EXPBIT) were overlapping, which suggests a similar elastic response. In both mastercurves, PMB exhibited a plateau at intermediate loading frequency, which is an indication of an elastic polymer network, as observed in past studies [3,15].

As shown in Fig. 4b, PMB and EXPBIT exhibited similar behavior, which was significantly different to the black diagram of the VIRBIT. At higher complex modulus ($>10^5$ Pa), insignificant differences were observed among the three bitumens while at lower complex modulus ($<10^5$ Pa), EXPBIT appears to show the lowest phase angle and VIRBIT exhibited the highest phase angle, indicating a greater viscous response than elastic response for the virgin binder.

Fig. 5 shows the values of complex modulus and phase angle at temperatures 10 °C, 40 °C, 80 °C with frequency of 1.59 Hz (10 rad/s) which is usually considered as a reference frequency corresponding to the shearing action at traffic speed of 90 kmph (50 mph) according to Superpave specifications.

VIRBIT exhibited the highest phase angle, which indicates its lower elastic response. At lower temperatures, VIRBIT exhibited high complex modulus whereas the stiffness was reduced on increasing the temperature suggesting an increment in proportion of viscous component when increasing the temperature. Among modified bitumens (PMB and EXPBIT), it is clear that the EXPBIT adapted the best to variation in temperature, because when the temperature was low, which can induce cracking problems, the EXPBIT exhibited a lower modulus. However,

when the temperature was high, which leads to deformation problems, it displayed higher modulus than PMB. The high-vinyl SBS content may have a softening effect due to which the stiffness was lower, however the elasticity was higher as compared to PMB. Since phase angle is an indicator of recovery and non-recoverable deformation, due to lower value, EXPBIT exhibited greater elasticity than PMB over the entire range of temperature. This indicates that the viscous component is lesser in EXPBIT than in PMB.

3.1.2. LAS results

The cycles until fatigue failure from the LAS test performed at 25 °C are shown in Fig. 6a and the plot of integrity parameter vs damage intensity is given in Fig. 6b. The fatigue behavior was analyzed using the VECD theory, next parameters A and B were computed and then the number of fatigue cycles (N_f) at various strain levels were computed. For each applied strain level, the pattern was similar: the highest number of cycles to failure is exhibited by the experimental bitumen, followed by PMB and then virgin bitumen. The virgin bitumen and experimental bitumen showed similar integrity parameters, however, VIRBIT has the lowest fatigue life. PMB showed a higher value of the integrity parameter than EXPBIT, however, the number of cycles to fatigue failure for PMB was lower than EXPBIT. This indicates that the presence of high-vinyl SBS in EXPBIT had a positive influence on the elasticity that may enable mixtures with EXPBIT to undergo more cycles before fatigue failure. The increased elasticity may be attributed to the addition of high-vinyl SBS. The fatigue life of EXPBIT was significantly greater than PMB (36.5%) and VIRBIT (63.9%) at low strain level (2.5%) whereas this increment was lower at high strain level (10%) compared to PMB (23.5%) and VIRBIT (48.24%). For thick pavements or low-traffic roads, the strain levels are lower and thus it can be concluded that EXPBIT may exhibit better fatigue performance than PMB and VIRBIT binder in this case compared to thin pavements or high traffic roads.

3.1.3. BYET results

The BYET test was performed at 25 °C temperature. The stress–strain plot for the asphalt binders are shown in Fig. 7a and the yield stress and the area under the curve are shown in Fig. 7b. The yield stress was highest for EXPBIT, therefore the addition of high-vinyl SBS may improve the fatigue life and may reduce fatigue cracking. The virgin binder displayed the lowest yield stress as well as the area under the curve. Greater area under the curve means higher yield energy, therefore, the modified binders can withstand a higher load before yielding compared to virgin binder.

Multi-peak phenomena were observed in the case of modified binders PMB and EXPBIT, this phenomenon was also previously noted by other researchers [34,61]. This may be due to the high elasticity and

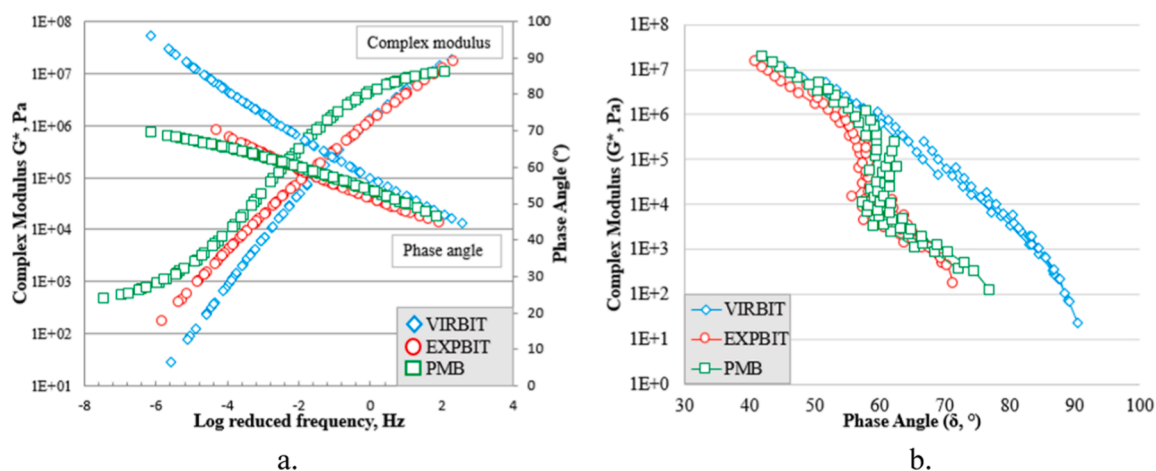


Fig. 4. DSR results of the binders: a. Master curves ($T_{ref} = 20$ °C); b. Black diagram (Temperatures 10–80 °C).

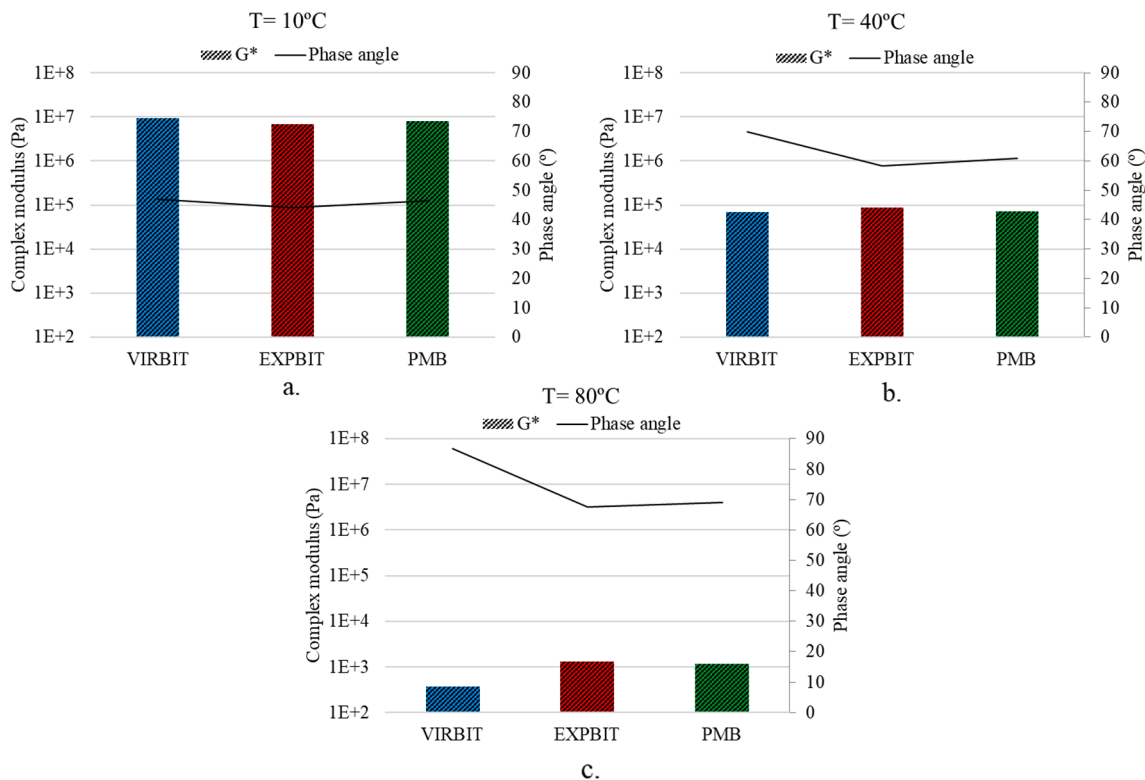


Fig. 5. Stiffness and phase angle parameters for each binder type at temperature a. 10 °C; b. 40 °C; c. 80 °C.

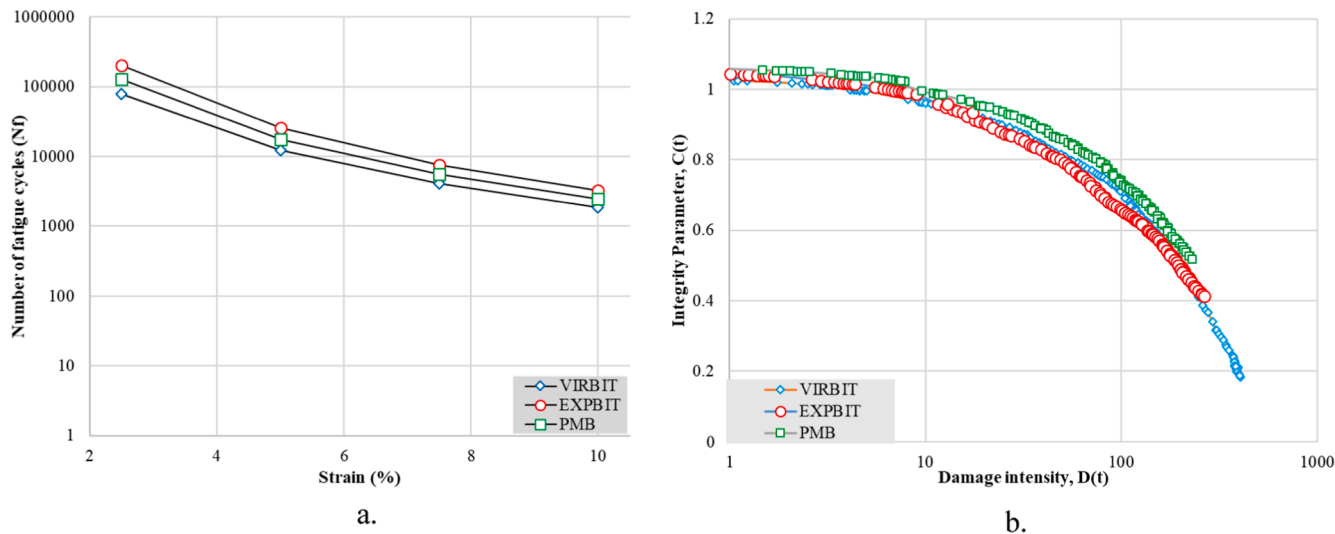


Fig. 6. Results from N_f from LAS tests.

good recovery because of the presence of polymer. According to Johnson [34], the first peak indicates the asphaltene-maltene relationship whereas the secondary peak is an indicator of the strength of polymer cross-linking. The results in this study were computed from the first peak of the graph. It is very interesting to note that the results were in agreement with LAS results that indicated that the experimental bitumen exhibits the largest number of cycles before failure.

3.1.4. MSCRT results

MSCRT shows the bitumen performance at high temperature that is an indicator of rutting characteristics of the asphalt pavement. As observed from Fig. 8a, VIRBIT showed higher strain compared to other binders. This means that under the same loading and time, the strain

produced in the virgin bitumen was much higher, which is due to the low stiffness of this bitumen at high temperatures. During the loading and unloading of the curve, the virgin bitumen exhibited less visco-elastic behavior as negligible reshaping of sample was observed. Meanwhile, for modified binders (PMB and EXPBIT), high recovery was observed. The EXPBIT and the PMB exhibited very similar properties, with a slightly higher strain observed in the case of experimental binder due to the lower stiffness of EXPBIT.

The plots for recovery (%R) vs creep compliance (J_{nr}) along with polymer modification curve are shown in Fig. 8b. The polymer modification curve is an indicator of elasticity according to AASHTO M 332. If the recovery (%R) of bitumen lies above this curve, this means that the bitumen has good elastomeric behavior and vice-versa [62]. The %R of

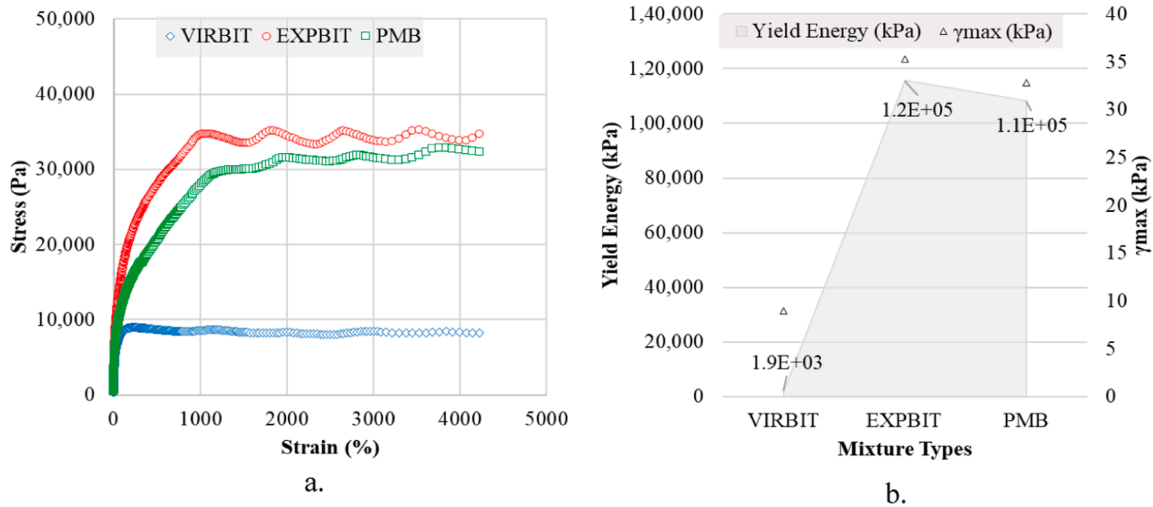


Fig. 7. BYET results, a. yield stress and yield energy plots; b. stress and strain plots.

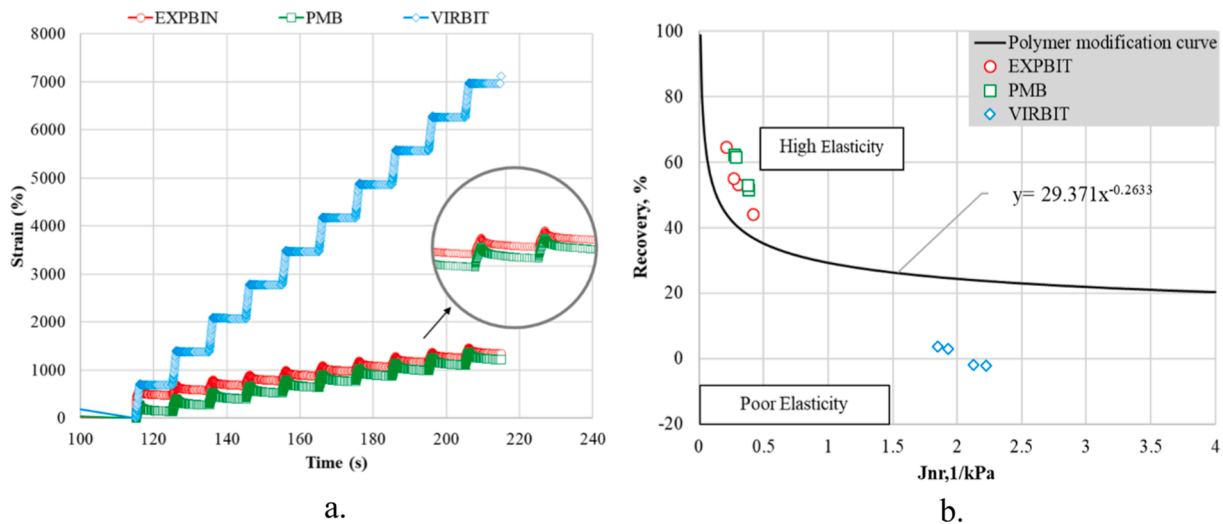


Fig. 8. MSCRT results: a. strain vs time plot, b. Percentage recovery and J_{nr} values.

the two modified bitumens (EXPBIT and PMB) was located above this curve, therefore they showed high elasticity while %R for VIRBIT was located below the modification curve and thus exhibits poor elasticity. The virgin bitumen showed negative percent recovery at 3.2 kPa stress level. Negative recovery indicates that the strain increases even in the unloading phase; which is common in soft bitumen [63].

At a stress level of 3.2 kPa, J_{nr} indicates the rutting performance of the binder. For extreme, very heavy, heavy and standard traffic, J_{nr} should be less than 0.5, 1, 2 and 4 kPa^{-1} respectively [64]. The two modified bitumens were within the limit for extreme traffic, whereas the value of J_{nr} for virgin bitumen was higher than 2 kPa^{-1} , which suggests that this bitumen was suitable for standard traffic level according to Superpave. $J_{nr-diff}$ should not be higher than 75% for 0.1 and 3.2 kPa stress levels. All three bitumen samples were well within this limit. Moreover, it should be noted that the J_{nr} value for experimental binder was less than PMB, which suggests that the rutting performance of EXPBIT is better than PMB.

3.2. Porous asphalt mixtures testing

Rheological tests have shown that the EXPBIT performs better than both PMB and VIRBIT in fatigue and rutting criteria. However, the PA mixtures suffer the problem of raveling that poses a great concern.

Therefore, PA mixtures were prepared to analyze the performance of the EXPBIT towards air void content, raveling, binder drainage. In addition to this, additional mixtures were prepared to assess the suitability of using the new binder with aramid and glass fibers, which have been proved to improve the mechanical resistance of PA mixtures [54].

3.2.1. Air voids content

Fig. 9 shows the air void content and the interconnected air void content of the different mixture types. For the three binders the air void content was very similar. However, on addition of fibers, the air void content was slightly reduced. The presence of fibers may block part of the air voids and compromise the permeability of the mixtures if the amount of fibers is not correctly designed. As can be observed all mixture types displayed air void contents higher than 20%. Significant differences were observed only in the case of glass-hybrid fibers (EXPST0.3 and EXPST0.5). Considering the interconnected air voids, indicated by the permeability of the mixture types, the highest reduction was observed in the EXPST0.3 case, which may be due to development of a stronger matrix.

3.2.2. Draindown test

Binder drainage is an important concern in PA mixtures due to the low percentage of fine aggregates. When the asphalt mix is subjected to

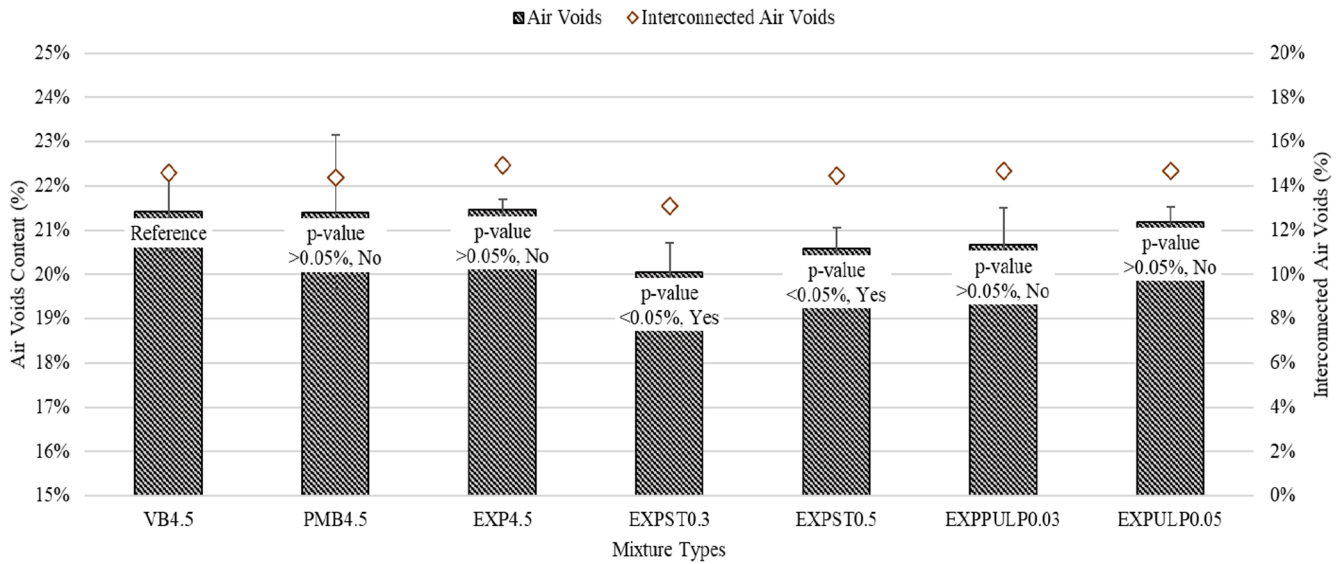


Fig. 9. Air void content for mixture types, error bars represent the standard deviation about the mean and the labels represent the p-value and significant difference from VB4.5 using the two-sample student's test.

high temperatures, the binder tends to drain down the structure of the air voids. As observed in Fig. 10, for the same bitumen content of 4.5%, the experimental bitumen displayed higher draindown than virgin binder and PMB. The reason for this result may be the different manufacturing temperature of the three bitumens. In this study the manufacturing temperature of PA mixtures with EXPBIT is 165 °C. For lower binder draindown, it is recommended to reduce the manufacturing temperature in the future. The binder drainage of EXP4.5 was close to the maximum recommended limit of 0.3%, however, it did not surpass the permissible limit considerably. When the fibers were added at 4.5% binder content, it was found that the binder drainage was reduced considerably, as both glass fibers and aramid pulp (EXPST0.3, EXPST0.5, EXPULP0.03 and EXPULP0.05) improved the binder stability. The possible explanation for this phenomenon may be the high bitumen absorption by the fibers or the development of a strong 3-D network on addition of fibers which is in agreement with studies that suggest that fibers improve the binder stability of PA mixtures [54,56–58].

3.2.3. Cantabro test

Fig. 11 represents the abrasion loss of all mixture types. The highest abrasion loss both in dry and wet conditions corresponded to the mixtures with VIRBIT. According to statistical analysis, significant differences were observed among the different mixture types as shown in Table 6. Comparing with VB4.5, experimental binder with and without the fibers has shown significant differences as the p-value is less than

0.05 both under dry and wet conditions. However, the experimental binder showed very similar abrasion loss to PMB. This can be attributed to high viscosity due to presence of polymers in the binder, which improves adhesion and raveling resistance. In dry conditions, the EXPBIT performs better than the PMB binder, but, in wet conditions, the particle loss is higher. However, the error bar suggests negligible difference in particle loss among the two modified bitumens.

On addition of fibers, the particle loss was further reduced as the mixtures with fibers EXPST0.3, EXPST0.5, EXPPULP0.03, EXPULP0.05 have lower particle loss compared to the EXP4.5 mixture in dry conditions (see Table 6). The difference is more prominent under wet conditions. The combination of glass fibers at 0.3% with experimental binder has the lowest abrasion loss both in dry and wet conditions (particle loss 5% and 5.9% in dry and wet conditions respectively). EXPST0.5 has higher abrasion loss than the EXP4.5 mixture, which may be due to the high amount of fiber that may have resulted in cluster formation. However, the difference between the mixtures with the two fiber contents is not significant. The addition of aramid pulp fibers has also reduced the particle loss of mixtures with fibers and the change in their fiber content had negligible influence.

Fig. 12 shows the plot of particle loss vs. air voids. A general trend under dry and wet conditions can be observed that on increasing in air voids, the particle loss increases as well. EXP4.5 mixtures have shown good abrasion resistance as well as high air void content followed by PMB4.5, which exhibits same particle loss in dry and wet conditions indicating less moisture damage. EXPST0.3 has the highest abrasion

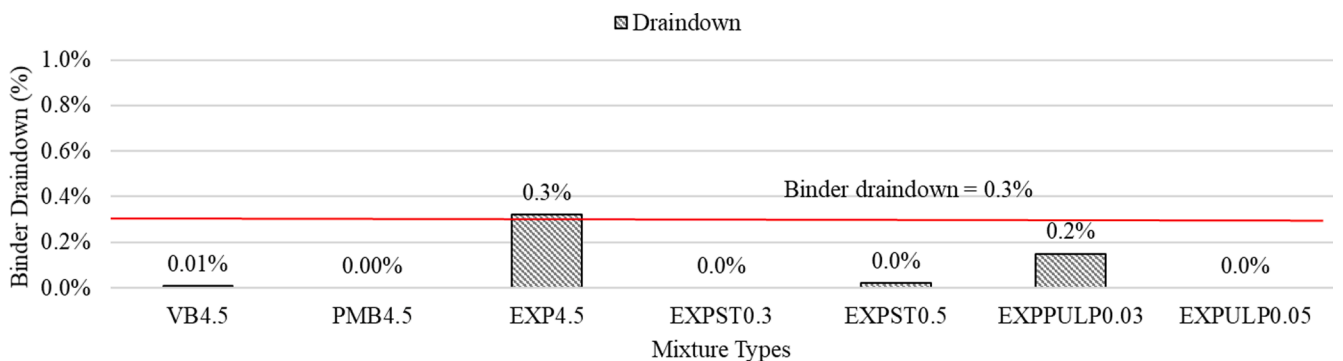


Fig. 10. Draindown of the mixtures.

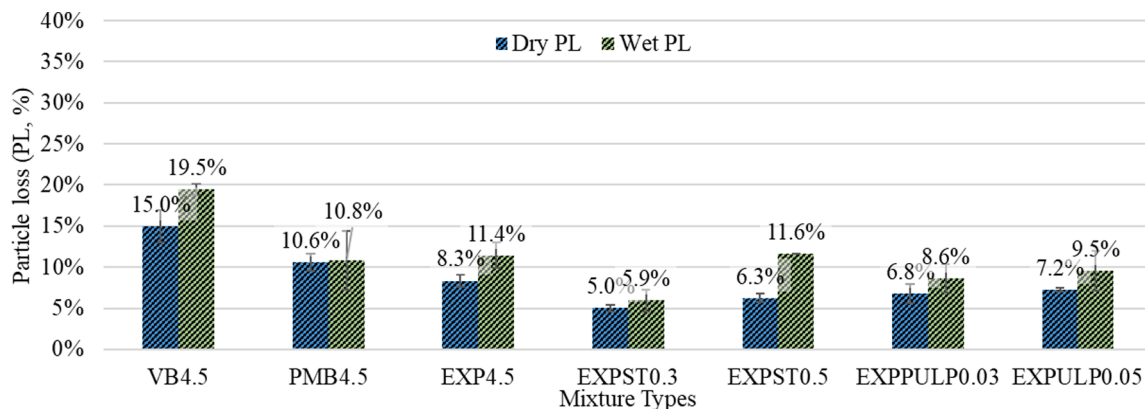


Fig. 11. Particle loss (PL) under dry and wet conditions, error bars represent the standard deviation about the mean.

Table 6
Statistical analysis of abrasion resistance under dry and wet conditions (two-sample *t* test).

	VB4.5	PMB4.5	EXP4.5	EXPST0.3	EXPST0.5	EXPPULP0.03	EXPULP0.05
Dry Conditions							
Normal	yes	yes	yes	yes	yes	yes	yes
p-value		0.152	0.001	0	0	0.003	0.001
Significance		no	yes	yes	yes	yes	yes
Wet conditions							
Normal	yes	yes	yes	yes	yes	yes	yes
p-value		0.043	0.002	0	0.002	0.001	0.007
Significance		yes	yes	yes	yes	yes	yes

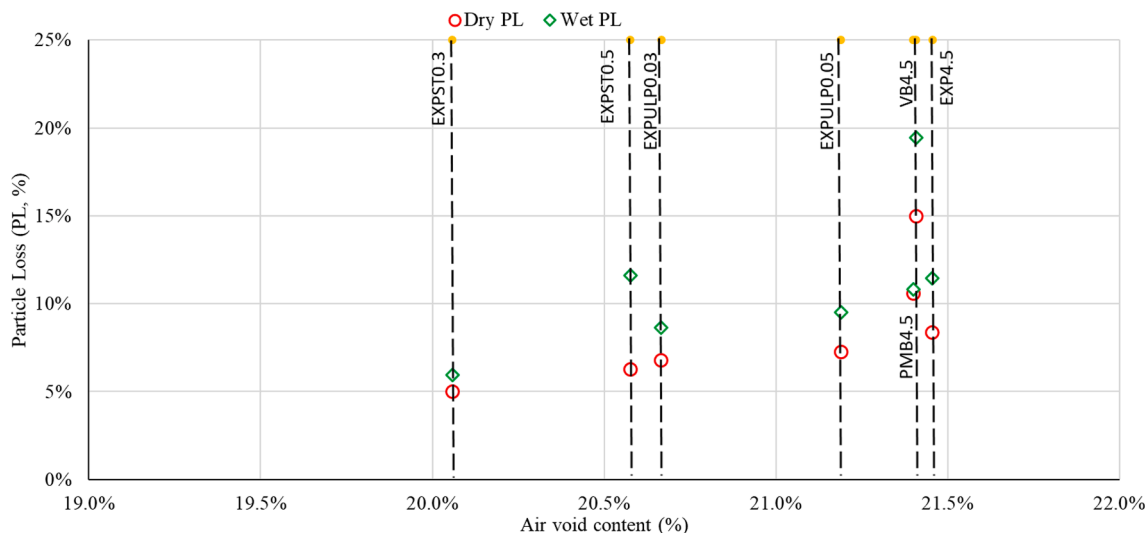


Fig. 12. Particle loss Vs. Air void content.

resistance, both in dry and wet conditions, but its air void content is lowest (20.1%), although, still higher than the minimum permissible limit of 20%.

4. Conclusions

In this study, a new modified bitumen using SBS polymer with high-vinyl content was developed to improve the mechanical resistance of porous asphalt (PA) mixtures. Experimental bitumen was compared with commercially available PMB45/80–65 and virgin bitumen 50/70 as these binders have a very similar penetration value. The rheological tests include temperature and frequency sweep tests, linear amplitude sweep test, binder yield energy test, and multiple stress creep recovery

test. Moreover, PA mixtures were prepared with these three asphalt binders to assess the behavior of the experimental bitumen in comparison to others. The synergistic benefits of fibers with the experimental bitumen were also investigated using two different fibers: glass-hybrid fibers and aramid pulp fibers. The following conclusions were drawn:

- After several trials, a polymer content of 4.5% was found to be stable as it conformed to the requirements of existing PMB based on conventional tests. An increase in the polymer content resulted in storage stability failure and gelation during fabrication of binder.
- PMB showed the highest stiffness while virgin bitumen exhibited the highest phase angle over a wide range of frequencies. At 1.59 Hz frequency, experimental bitumen showed best adaptation to

variation in temperature. At low temperature, the experimental bitumen exhibited lower complex modulus that indicated higher cracking resistance. At high temperatures, experimental bitumen exhibited higher complex modulus, which indicates high stiffness under deformation. The high-vinyl SBS content may have softened the bitumen leading to lower complex modulus but higher elasticity.

- Linear amplitude sweep tests highlighted that the number of fatigue cycles to failure is greater for the experimental bitumen in comparison with commercial PMB and virgin bitumen at any given strain level. Therefore, addition of SBS in experimental bitumen may have a positive influence on the fatigue performance.
- Binder yield energy test results showed the multi-peak phenomena for PMB and experimental bitumen. The results were in agreement with the linear amplitude sweep tests, the highest yield stress and yield energy were observed for the new experimental binder.
- At high temperatures, according to multi-stress creep and recovery test results, the experimental bitumen showed very similar high temperature characteristics to PMB. Experimental bitumen exhibited lowest creep compliance and highest percentage of recovery, indicating better elastomeric behavior and superior rutting performance compared to PMB and virgin bitumen.
- Porous asphalt mixtures prepared with experimental bitumen showed similar air voids compared to the reference mixtures with virgin bitumen and PMB. However, the draindown results indicated lower stability of experimental binder. The incorporation of fibers which remarkably improved the binder stability.
- Concerning the influence of experimental bitumen on the abrasion resistance of PA mixtures, the particle loss was found to be similar to PMB mixtures and lower than virgin bitumen under both dry and wet conditions.
- On addition of fibers, synergistic benefits can be observed as the abrasion resistance is greatest for the experimental bitumen with a fiber content of 0.3% glass hybrid fibers mixtures, followed by fiber content of 0.03% aramid pulp fibers.

Consequently, it can be concluded that the new experimental bitumen combined with fibers showed a better performance in this study than PMB for PA mixtures. For future research, an important aspect will be to investigate the effect of aging on the physical, chemical, and rheological properties of the experimental bitumen and evaluate the performance of PA mixtures by simulating the field aging in the laboratory.

CRedit authorship contribution statement

Anik Gupta: Conceptualization, Investigation, Methodology, Writing - original draft. **Pedro Lastra-Gonzalez:** Writing - review & editing. **Jorge Rodriguez-Hernandez:** Visualization, Supervision. **María González González:** Visualization, Writing - review & editing, Investigation. **Daniel Castro-Fresno:** Supervision.

Declaration of Competing Interest

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

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