INVESTIGATING THE CHARACTERISTICS OF MODIFIED BITUMEN WITH POLYPHOSPHORIC ACID POLYMERS AND RECYCLED RUBBER

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Resume

The impact of polyphosphoric acid (PPA) polymers and crumb rubber (CR) on bitumen's properties to enhance its performance was examined in this study. Laboratory samples were created with varying percentages of PPA (0.5% to 2%) and CR (6% to 18%) and subjected to extensive testing. Findings indicated that the combination of PPA and CR led to notable improvements in bitumen performance, including increased viscosity, softening point, elasticity, and recovery rates at elevated temperatures. Specifically, samples with 18% CR and 1.0% or 2.0% PPA demonstrated exceptional rheological properties and strength. The optimal combination (18% CR and 1.0% PPA) resulted in a uniform structure, while the maximum CR content did not compromise bitumen integrity. Both additives effectively minimized temperature sensitivity and aging, enhancing the bitumen's performance class significantly.

Article info

Received 4 April 2024
Accepted 24 July 2024
Online 23 August 2024

Keywords:

rutting resistance
modified bitumen
performance grades (PG)
polyphosphoric acid polymer (PPA)
waste rubber (CR) powder

Available online: https://doi.org/10.26552/com.C.2024.050

ISSN 1335-4205 (print version)
ISSN 2585-7878 (online version)

1 Introduction

Bitumen, a solid waterproof and thermoplastic adhesive, serves as a crucial component in bonding aggregates together. The incorporation of additives, particularly polymers, stands out as an effective strategy to enhance the performance of asphalt pavement [1]. Over the course of history, numerous research endeavors have been devoted to the bitumen additives and the enhancement of bitumen properties [2-3]. For instance, Padhan and Gupta conducted a study on bitumen modified with Polyethylene Terephthalate (PET), derived polyurethane polymer, employing a battery of standard bitumen tests and Superpave bitumen tests to assess its impact [4]. The inclusion of this additive yielded significant improvements in the performance and properties of bitumen. A study analyzed the post-hardening changes in epoxy-modified bitumen, finding that aging levels correlate with epoxy modification and sulfoxide compounds indicate oxidative hardening [5]. Moreover, the findings underscored that epoxy augmentation bolstered the resistance of bitumen against aging. Furthermore, in 2019, researchers explored the rheological and microstructural characteristics of carbon nanomaterials-modified bitumen [6]. Their investigations revealed that Carbon Nanotube (CNT (and Graphene(Gr)-modified bitumen outperformed unmodified bitumen in terms of rheological and microstructural properties, with the optimal blend identified as modified bitumen containing 1.5% CNT and 1% Gr.

Lin et al. highlighted the challenge posed by the increasing production of Waste Express Bags (WEB) in the context of population growth and the logistics industry [7]. Their study aimed to explore the potential of using WEBs to enhance bitumen properties. Through a series of tests on varying doses of WEB-modified bitumen, they observed significant improvements in key bitumen characteristics such as penetration, ductility, softening point, and rotational viscosity. Building on this, asphalt experts delved into emulsion bitumen modification using nanocomposites, noting changes in penetration and softening point alongside enhanced rheological behavior [8-9]. Subsequent research in 2020 investigated the impact of natural rubber (NR) on bitumen performance, revealing enhancements in rutting, fatigue resistance, thermal cracking, and moisture sensitivity. Further studies in the same year examined the use of Nano-silica composite and Waste Denim Fibers (WDF) to enhance bitumen rutting performance, leading to improved performance grades [10]. In one study, the PPA value obtained from ATR-FTIR may serve as an indicator for the modification of PPA/CR. The mechanism of PPA/CR modification is elucidated. Additionally, the rheological properties and storage stability of asphalt are influenced by the composition of Asphalt. Moreover, the addition of PPA enhances the stability of the CR-modified asphalt during the thermal storage process. The rheological properties of the CR-modified asphalt are improved through the incorporation of PPA [11]. Another study investigates the impact of poly phosphoric acid (PPA) on the rheological properties of styrene butadiene styrene (SBS) and crumb rubber (CR) modified asphalt binders at elevated temperatures, with varying PPA concentrations and comparison to a reference binder with 5% SBS. Various laboratory tests were conducted to evaluate the asphalt binders' rheological behavior, indicating that the addition of PPA improved resistance to permanent deformation and elastic recovery, with the most significant effects observed in PPA-SBS mixtures followed by PPA-CR mixtures, showcasing enhanced anti-aging characteristics and loading resistance in CR-modified asphalt binders, particularly at a minimum PPA content of 0.8% [12]. Moreover, the utilization of polymers, particularly polypropylene, was noted to enhance the thermal-mechanical performance of bitumen [13]. Notably, the study on waste polystyrene (PS) as a bitumen modifier demonstrated compliance with regulations and superior performance compared to conventional binders, offering a sustainable solution to the environmental challenges posed by the PS production. Experts in the asphalt industry also point out that improving the self-healing properties of bituminous mixtures to increases the life of pavement, and road safety, and they investigated the effect of bitumen modification with different percentages of polyurethane polymer toluene diisocyanate (TDI-Co) on the self-healing properties of asphalt, and found that the use of polyurethane polymer increases the self-healing performance of bitumen [14]. In another study in 2022, palm oil clinker (POC) was studied as an additive to asphalt, and it was found that it substantially improves the properties of bitumen, including reducing penetration, and increasing the softening point [15]. In another study, the intermolecular behaviors of PPA/SBR modified asphalt were investigated through molecular dynamics (MD) simulation. The high temperature performance of SBR modified asphalt was found to improve with the addition of PPA. The incorporation of PPA was observed to hinder the diffusion and mobility of SBR asphalt molecules, ultimately enhancing the stability and orderliness of the molecular structure. Those findings highlight the potential benefits of using PPA in SBR modified asphalt for improved performance [16]. The researchers discussed the possibility of mixing the polymer, and hydrocarbon molecules with fossil origin as well as industrial polymerization, but did not mention the functional difference between these materials [17-18].

Several research studies have been undertaken to examine the impact of incorporating CR and PPA on the characteristics of bitumen. Nevertheless, only a limited number of these studies have specifically explored the consequences of simultaneously introducing PPA and waste CR into bitumen [19-20]. Behavioral changes due to adding numerous additives usually occur within the range of linear viscoelastic performance. However, it is necessary to investigate the behavior at higher temperatures, higher stresses and higher loading rates. Although there are many articles on the use of recycled rubber powder, and much research has been done, this study intends to study the simultaneous use of rubber powder, and polyphosphoric acid polymer. Therefore, the main purpose of this study is to investigate the effect of PPA, and waste rubber powder on properties and performance of bitumen. For this purpose, samples of bitumen with different percentages of PPA, and CR were prepared in the laboratory, and subjected to various tests.

2 Research method and experiments

In this study, the focus was on examining the impact of PPA and CR on the characteristics and behavior of bitumen. Through the addition of these materials in varying proportions as additives to bitumen, the modified bitumen underwent testing, based on the performance grade (PG) and Sharp supplementary grade (PG^+). Statistical analyses were employed to scrutinize the results and determine the influence of different parameters on the performance of the asphalt mixture. The materials used in the research included PG 58-22 bitumen, CR with specific grading, and polyphosphoric acid polymer at varying concentrations. Tables 1, 2 and 3 present some of their specifications. The selection of bitumen with high penetration was based on the modification of lower-quality bitumen, with additives chosen based on prior research and experiences. Notably, the addition of CR ranged from 6% to 24%, with concentrations exceeding 20% leading to mixing challenges and the formation of agglomerates. The study excluded the 24% concentration and focused on 6%, 12%, and 18% levels. The CR percentage, or crumb rubber percentage, represents the ratio of rubber crumb weight to the weight of bitumen, excluding any additional additives. Figure 1 illustrates the gradation graph depicting the CR particle distribution, while Figure 2 provides a visual representation of the materials involved in the preparation process, including waste rubber powder, polyphosphoric acid polymer, and the mixing apparatus.

***Table 1** Physical characteristics of bitumen used in this research*

Bitumen tests	Measurement unit	Test Results	Standards ASTM
Specific gravity	-	1.0179	D75
Needle penetration	0.1 mm	90	D5
Softening point	°C	45.8	D36
High-temperature viscosity of bitumen with rotary viscometer (RV)	Pa.s -135 °C	0.245	D4402
Flash point	° C	322	D92
Solubility in trichloroethylene	%	99.7	D2042

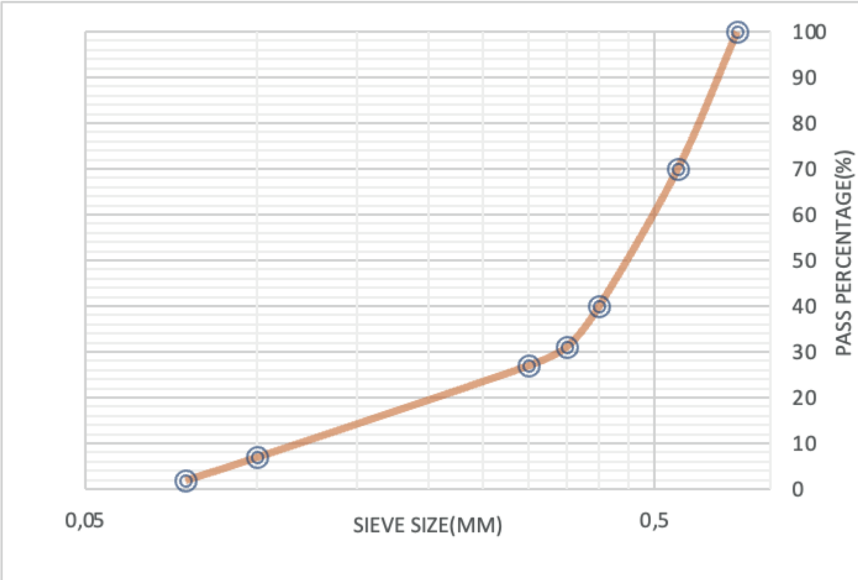

***Figure 1** Gradation graph representing the CR particle distribution*

***Table 2** Physical characteristics of CR used in this research*

Specifications	Unit	Result
Humidity	Percent	0.15
Weight density	Astm D7263 (g/cm^3)	0.33
Contaminants	Percent	0.07
Abrasion resistance	ASTM D1630-06	Appropriate level, below 20%
Compression capability	ASTM D395-14	Excellent

The use of PPA as an additive to enhance bitumen properties has shown promise in recent investigations, with its reactive nature playing a significant role in improving the physical and rheological properties of bitumen. Understanding the structure of this compound and its impact on asphalt mixtures can provide valuable insights into enhancing bitumen properties through the utilization of PPA.

In the process of preparing a specimen containing waste rubber powder, a meticulous approach was followed to ensure optimal blending. The bitumen was heated to 150 °C, and CR was added at 2500 rpm. Subsequently, the temperature was raised to 180 °C, and a thorough mixing operation was carried out for 120 minutes at 3500 rpm. Specific temperature ranges and mixing durations were adhered to for specimens containing PPA to ensure proper blending. For specimens incorporating both PPA and CR, a detailed procedure involving sequential heating, material addition, and controlled mixing was meticulously executed. Following the preparation of bitumen specimens, experiments were conducted to evaluate properties such as PG and Sharp plus grades (PG$^+$), essential for determining the suitability of the bitumen for various applications [20].

The impact of heat and air on a thin moving layer of bitumen is assessed using the Rolling Thin Film Oven (RTFO) test method, which investigates approximate changes in bitumen properties, including rheological properties, at around 150°C, by conducting tests at 150°C, our goal is to simulate real-world conditions to uncover valuable information about the behavior of bitumen in various scenarios. Bitumen remaining from this test retains similar properties to the bitumen used in hot asphalt mixtures after mixing and can also determine mass changes in bitumen. This test is conducted following the ASTM D2872 standard. The pressurized aging tank (PAV) accelerates the aging of bitumen by exposing them to pressurized air and high temperatures to simulate the changes in bitumen rheology that occur during pavement service, as per ASTM D6521. The ASTM D6521 standard outlines the standard PAV test, with temperatures ranging from 90 to 110 °C (depending on climatic conditions), lasting 20 hours under an air pressure of 2.1 MPa. For this research, the selected temperature was 100 °C, with the test performed once. This choice reflects the conditions typical of a hot climate, specifically at 110 °C, which may influence the material properties under investigation.

The determination of flexural creep stiffness of bitumen is done using the Bending Beam Rheometer (BBR) device that operates in a temperature range between 0 and -36°C [21]. Creep hardness (S) in this test reflects the stress-strain behavior of bitumen at low temperatures, with the rate of creep changes (m-value) serving as criteria for measuring bitumen performance at low temperatures. This test is carried out following the ASTM D6648 standard. The Dynamic Shear Rheometer (DSR) test method is employed for bitumen with a dynamic shear modulus range between 100 Pa and 10 MPa [22]. The tests were conducted using a Dynamic Shear Rheometer (DSR) equipped with a 25 mm plate and a 1 mm gap. Each testing cycle consists of 1 second of loading followed by 9 seconds of rest, with 10 cycles performed at a stress of 0.1 kilopascal, immediately followed by another 10 cycles at a stress level of 2.3 kPa. The total time required to complete all the loading cycles is approximately 300 seconds. The DSR must be capable of recording stress data every 0.1 seconds and should also be able to capture recovery data at least every 0.45 seconds. This testing method is thoroughly described in the ASTM D7405 standard [23]. Typically, an oscillation frequency of 10 radians per second (10 rads per second or 1.59 Hz), which is equivalent to the motion of a vehicle traveling at a speed of 90 km/h, was followed in this study.

The Linear Amplitude Sweep (LAS) test serves a critical function in evaluating the viscoelastic properties of bitumen, particularly in relation to its performance under varying temperature conditions. By measuring the hardness values at specified initial temperatures, this test provides insights into the material's hardness and brittleness at intermediate temperatures, both of which are pivotal in assessing the likelihood of fatigue cracking in bituminous samples. The increased brittleness observed at these temperatures can significantly elevate the risk of fatigue damage, thereby impacting the longevity and structural integrity of asphalt pavements. For standardization in LAS testing,

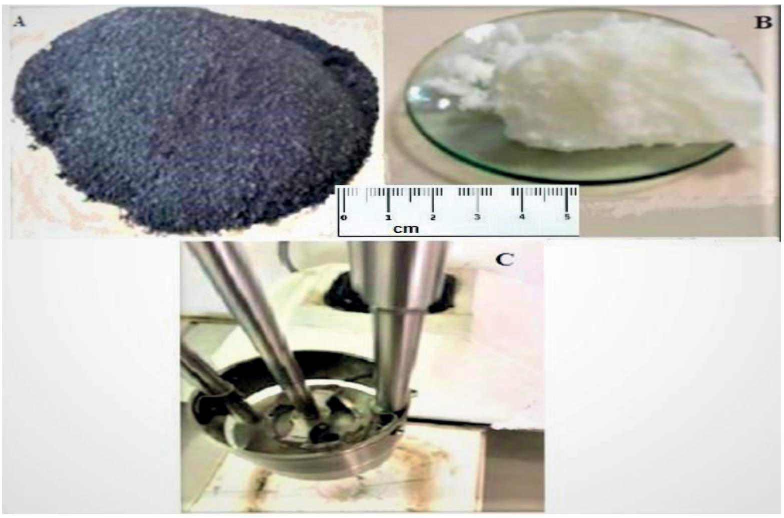

Figure 2 *A) waste rubber powder, B) polyphosphoric acid polymer and C) preparing specimen with a mixer (Silverson L5M-A Laboratory Mixer)*

Table 3 *Characteristics of PPA used in this study*

Product Name	Polyphosphoric acid polymer
Acronym	PPA
Chemical formula	HO[P(OH)(O)O](n)H
Boiling point	530 °C
Melting point	-20 °C
Density (at 20 °C)	2.03 g/cm^3

the relevant international standard is AASHTO TP101, which delineates the methodologies and requirements necessary for consistent and reliable test results.

3 Results and discussion

3.1 Characteristics of bitumen rheology by shear rheometer

Performance degree of bitumen at high temperature

To determine the degree of performance at high bitumen temperature, the rheology properties of bitumen are evaluated by DSR, and the parameter G*/sinδ is a measure of bitumen behavior at high temperature. According to the Superpave guideline (ASTM D6373), this parameter should be greater than 1.0 kPa for virgin, and non-aged bitumen, and greater than 2.2 kPa for aged bitumen after the short-term aging process (RTFO) as shown in Figure 3. In other words, in Figure 3, the column on the left is virgin bitumen and the right column is for aged bitumen.

One aspect to contemplate is the impact of the aging progression on the specimens. The initial phase Base bitumen specimens, and in subsequent phase, specimens altered by discarded CR have marginally reduced efficacy post RTFO; nevertheless, specimens encompassing PPA even displayed enhanced efficacy in certain instances. The addition of PPA to rubber bitumen also improved the performance of the samples after RTFO, and arguably PPA reduces the adverse effects of the aging process.

High temperature creep test

The temperature creep test was used to measure the two parameters G*, and δ, to simulate the conditions of temperature changes, and was performed on non-aging, and short-term aging (RTFO) samples. All samples showed a decrease in G*, and an increase in δ at high temperatures. Bitumen exhibits viscous behavior at high temperatures, and its resistance to deformation is significantly reduced, which is a principle accepted by all researchers. However, additives have reduced bitumen yield at different levels, although this varies from sample to sample (Figure 4).

In addition, all samples showed better performance after aging, which shows that both additives applied in this study have reduced the sensitivity of bitumen to aging. Comparison of G* values in samples containing only one of the two additives, Figure 4, showed that PPA was more capable of increasing the strength of bitumen, and the higher the result of this additive, the higher the G* value. The best performance is related to bitumen containing 2.0% PPA after aging. Additionally, samples containing 1.5%, and 1.0% PPA showed higher resistance than samples modified with rubber powder.

Comparing the phase angle values (δ) of these samples, Figure 5, shows that CR has been more successful in increasing the elasticity of bitumen, and bitumen containing 18% CR has significantly improved the elasticity. Another interesting point that emerges from this comparison is the trend of phase angle changes in bitumen containing 12, and 18% CR; as the temperature rises, the values of δ first increase slightly, and then begin a decreasing trend. Actually, when the base bitumen loses its elasticity with increasing temperature, the particles of CR in the sample help to restore the elasticity of the bitumen, and to some extent preserve the bitumen performance, and structure.

Comparison of G* values for samples containing two additives, Figure 6, showed that the combination of 1.5% PPA with 6%, and 12% CR showed higher G* values at initial temperatures. However, with increasing temperature (around 50 °C), these values drop sharply,

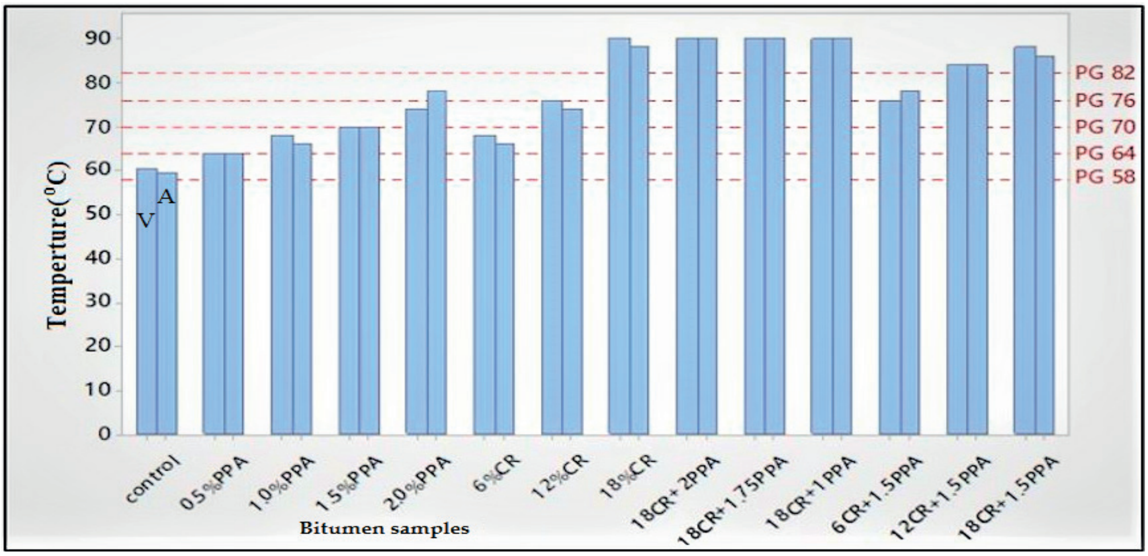

***Figure 3** Performance of high-temperature modified bitumen samples, the left column for virgin bitumen(V) and the right for aged bitumen(A)*

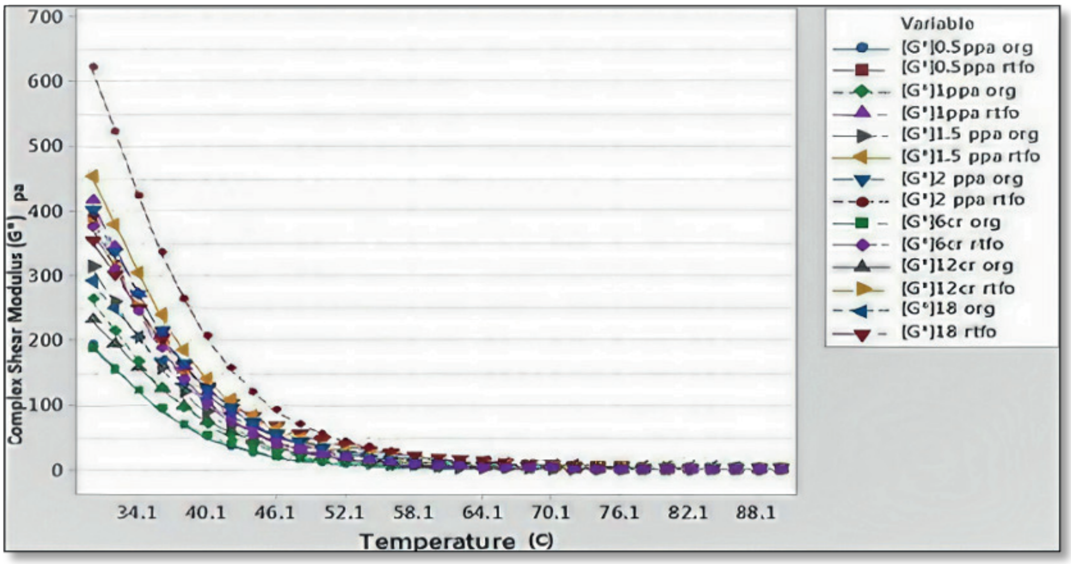

Figure 4 *Complex shear modulus (G*) values for the first-stage samples*

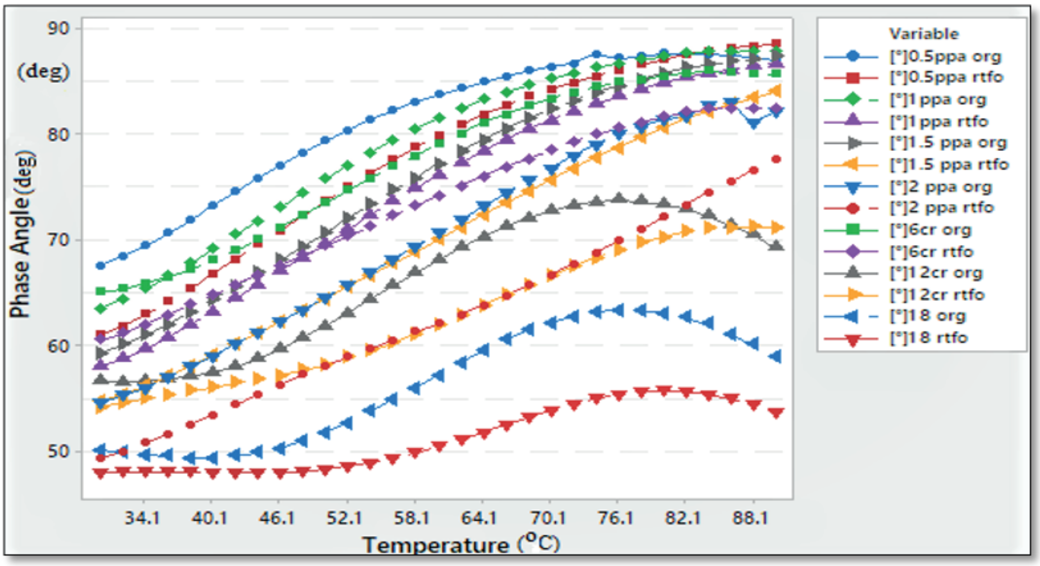

Figure 5 *Phase angle values for the samples containing the first stage*

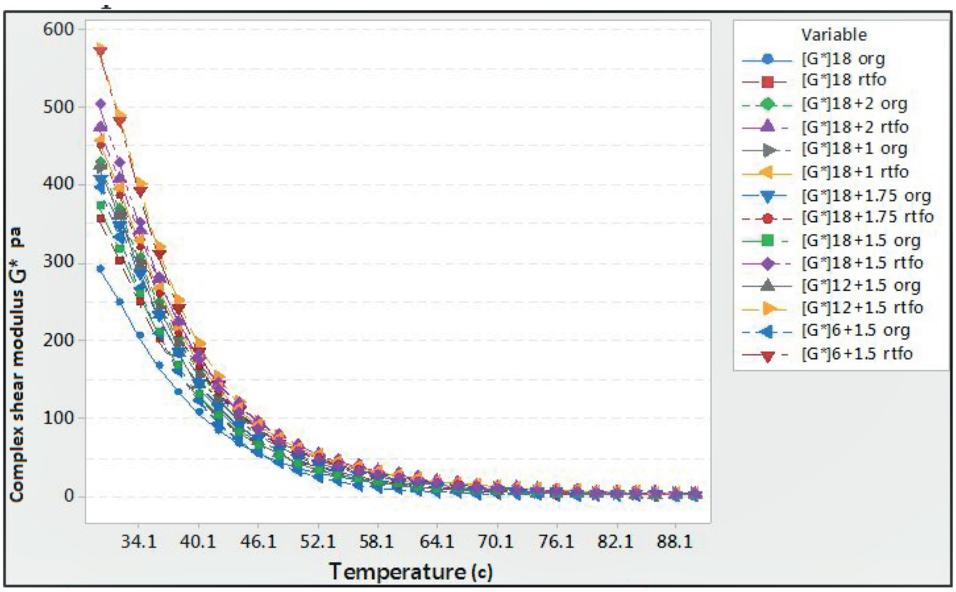

Figure 6 *Complex shear modulus values for the second-stage samples*

and show poorer performance than other samples, In general, it can be said that the combination of two additives has increased the resistance of samples to permanent deformation, and samples (18% CR+1.0% PPA) and (18% CR+2.0% PPA) had a lower rate of G* than other samples.

The best performance for bitumen containing 18% CR with 2.0, and 1.0% PPA is after short-term aging. The bitumen melting point in the sample (18% CR+2.0% PPA) is about 56 °C, and in the sample (18% CR+1.0% PPA) is about 52 °C. Bitumen at previous temperatures has a significant elastic property, and certainly at higher test temperatures, δ shows a slight increase rate. Two samples (6% CR+1.5% PPA), and (12% CR+1.5% PPA), which showed high G* at initial temperatures, showed fragile elastic properties, which reveals the need for simultaneous attention to both G*, and δ parameters.

Results related to the BBR test

One of the objectives of this study was to increase the high degree of performance of bitumen, without affecting the low-temperature performance of bitumen. From this point of view, CR additive seems to be successful, because, in addition to improving the performance of bitumen at high temperatures, it has also improved the low-temperature performance of bitumen, and all levels of this additive have succeeded in doing so, and as the result of CR increased, the performance of bitumen also improved (albeit slightly). The best performance related to bitumen contained 18% of CR which managed to improve the performance of bitumen at low temperatures by one rank. Bitumen containing 1.5% PPA has a similar function to rubber powder, and slightly improves the behavior of bitumen at low temperatures. Samples containing 1.0%, and 2.0% PPA did not show a significant change in m-value, which was confirmed by statistical test (P value > 0.13). However, the sample containing 2.0% PPA caused a decrease in bitumen performance. The m-value for all samples can be seen in Figure 7.

Regarding the combination of two additives, actually the sum of the two additives reduced the m-value. Although this drop in performance varies at different levels, for example, 1.0% PPA caused a slight drop in bitumen yield containing 18% of CR; with increasing PPA, this rate of decline has increased. It seems, the

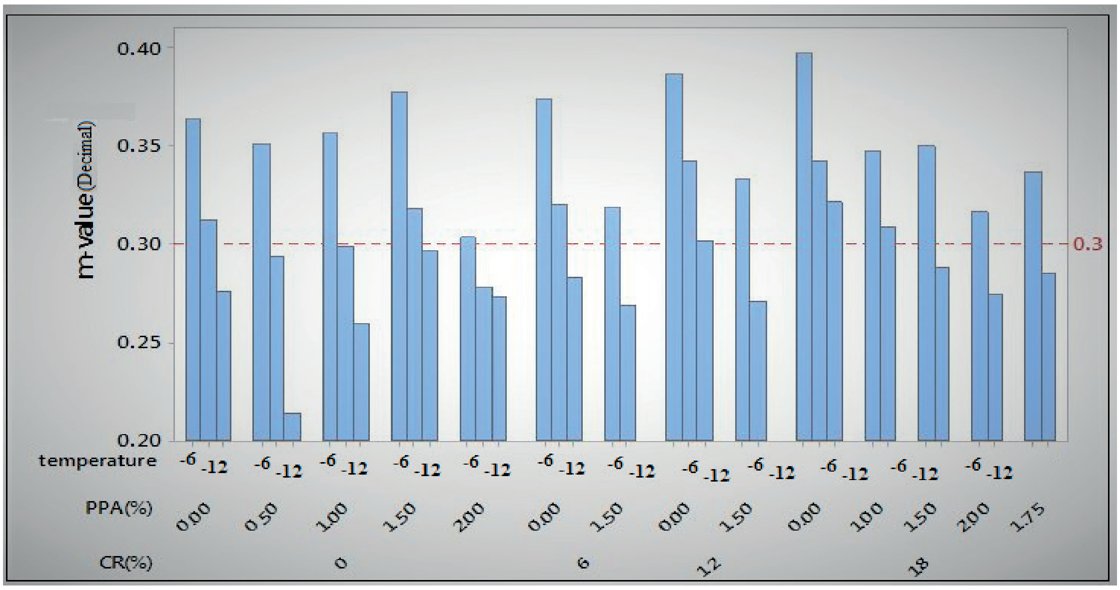

Figure 7 *Creep change rate (m-value) based on other variables*

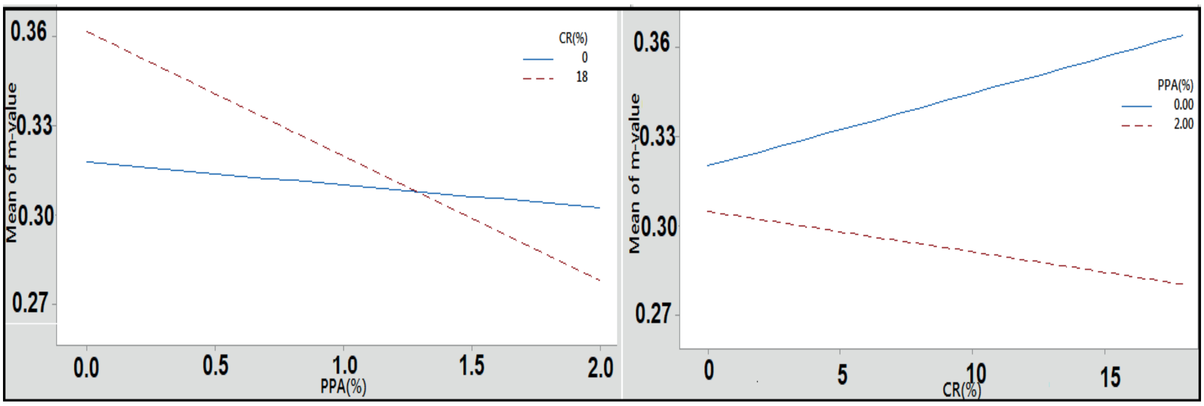

Figure 8 *Effects of PPA(Left), and CR(Right) based on m-value parameter*

reason can be found in the double increase in bitumen hardness due to the combination of two additives, and, of course, the increase in its brittleness, and fragility at low temperatures, Figure 8.

Results related to the LAS test, compliant with the standard [24]

Figure 9 shows some modified samples compared to the base sample 100-85. As it turns out, all the specimens, except the base bitumen, are above the specified mark, and meet the requirements of the fatigue law.

Results related to the Multiple Stress Creep Recovery (MSCR) test

This test provides two parameters of percent recovery (R%), and non-recoverable creep compliance(Jnr) under 0.1, and 3.2 kPa load levels at a test temperature of 58-76 °C. Typically, the higher the bitumen recovery capacity (R%), the more desirable it is. In addition, the closer the non-recoverable creep compliance is to zero, the higher the bitumen resistance to deformation.

At a stress level of 0.1 k Pa, it is observed that the higher the result of CR or PPA, the higher the bitumen recovery rate. Samples that have been modified by only one of the two additives in this study have experienced a decrease in R% with increasing temperature. The samples that combined the two additives also had an increasing R% parameter, albeit a small one. At high levels of the two additives, little change in R% is observed. This level of stress does not seem to be able to distinguish between additive-content samples. The best example of recovery at this stress level is a combination

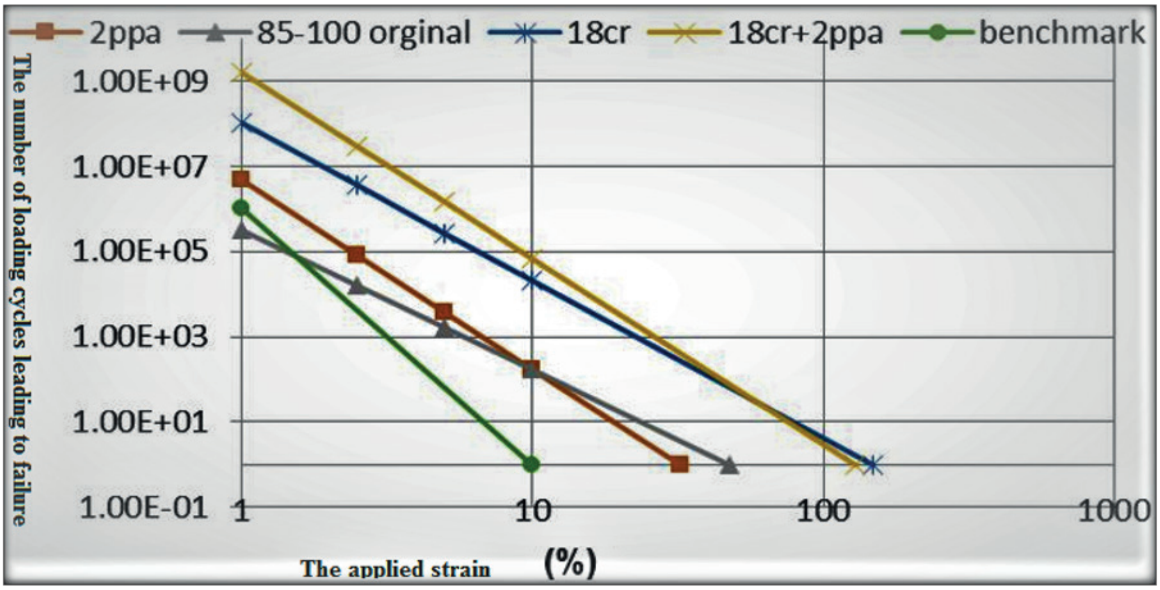

Figure 9 *Comparison of multi-sample fatigue model (at -20 °C after PAV)*

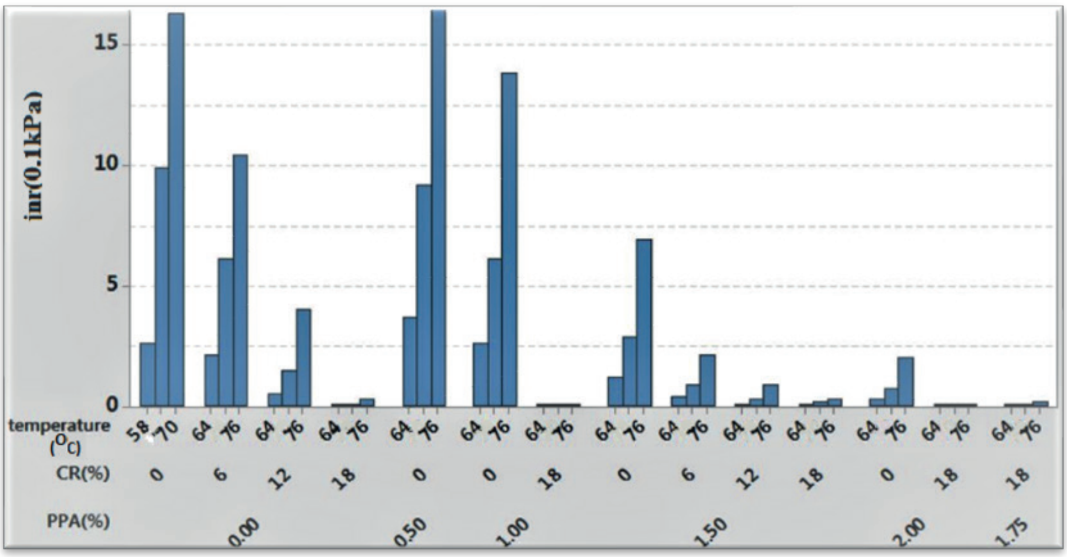

Figure 10 *The effect of test variables on the R% parameter at a stress level of 0.1 kPa*

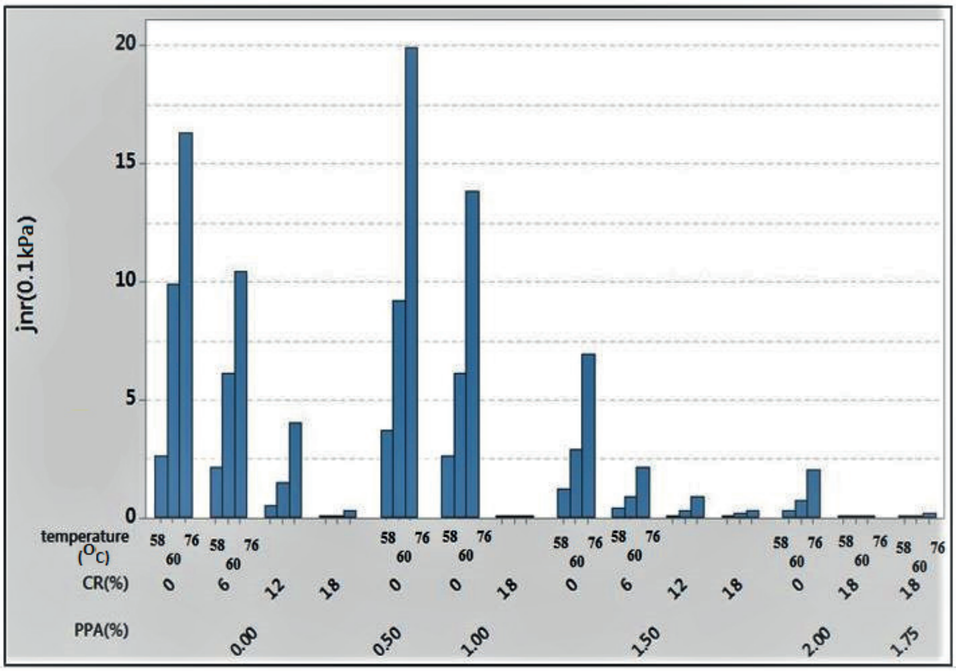

***Figure 11** The effect of test variables on the j_{nr} parameter at a stress level of 0.1 kPa*

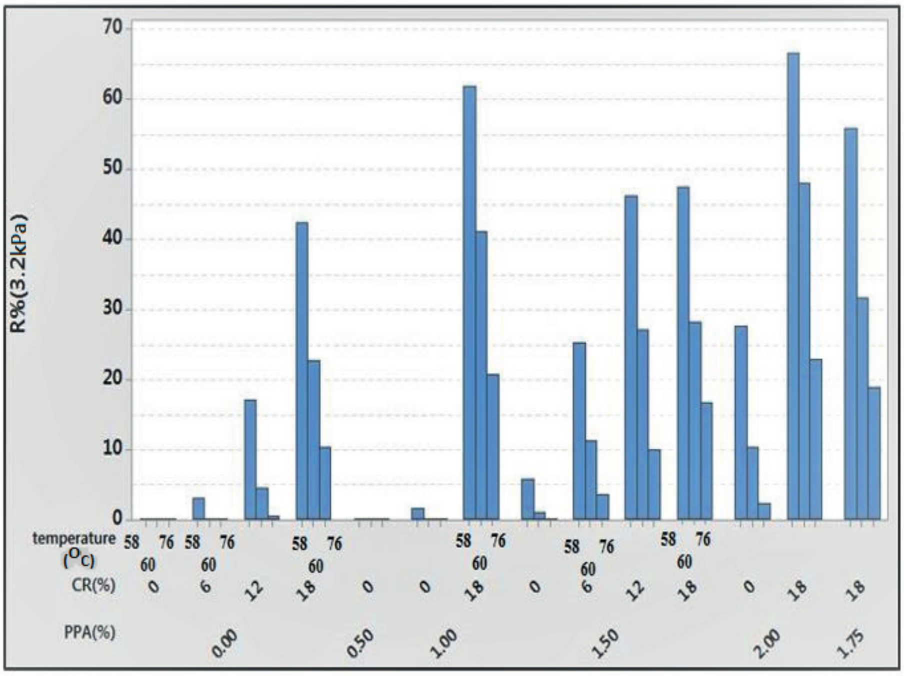

***Figure 12** The effect of test variables on the R% parameter at a stress level of 3.2 kPa*

of 18% CR plus 1.0% PPA. All examples are shown in Figure 10.

The non-recoverable creep compliance at a stress level of 0.1 kPa can be said to be significantly reduced by the addition of additives, and the highest level of CR (18 %), and the highest level of PPA (2.0%) have significantly reduced it. Since this value is directly related to the recovery rate, the best additive levels are the R% parameter. It is noteworthy that the base bitumen at 76 °C, is ultimately flowed, so it has been removed from the comparison. All the points are illustrated in Figure 11.

At a stress level of 3.2 kPa, and for the percent recovery, it can also be said that by adding both bitumen modifiers, the bitumen behavior is improved, and CR seems to perform better than PPA. The sum of these two additives has been successful in increasing the bitumen recovery percentage, and it seems that the best example according to this parameter is the combination of 18% CR with 2.0% PPA, and the combination of

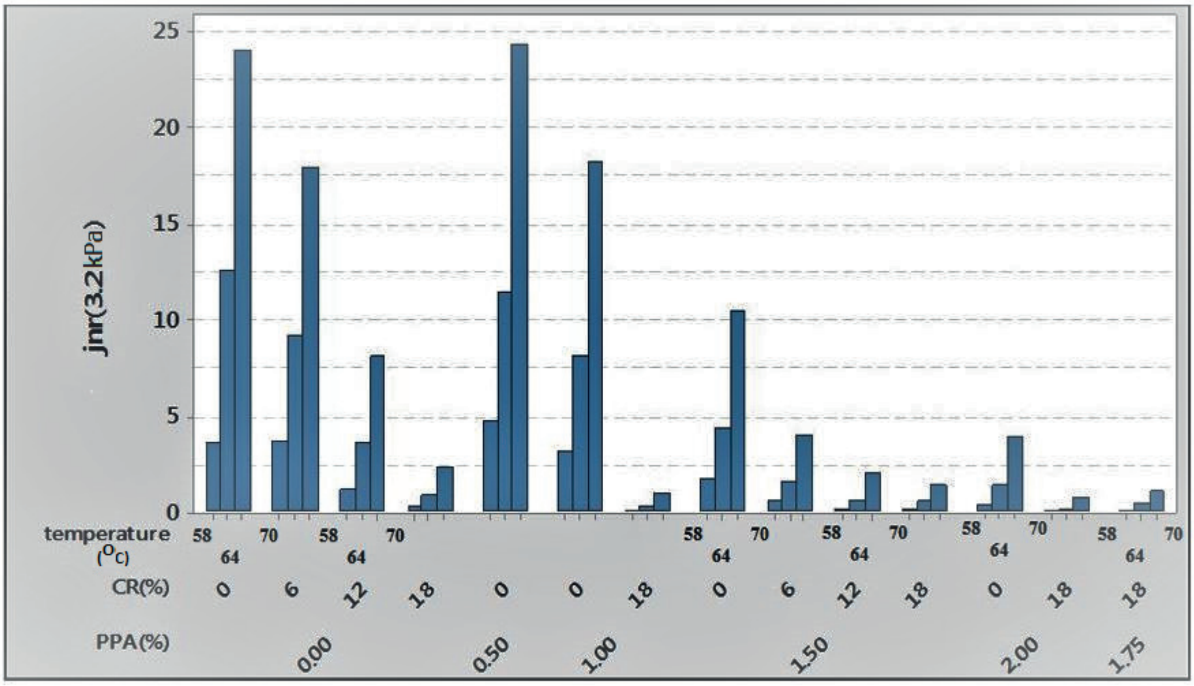

Figure 13 *The effect of test variables on the* j_{nr} *parameter at a stress level of 3.2 kPa*

18% CR plus 1.0% PPA, with a slight difference, is in second place. All the relevant points can be seen in Figure 12.

Finally, the essential parameter obtained from the MSCR test is the non-recoverable creep compliance at the stress level of 3.2 kPa, which has shown a high correlation with the result of rutting in field experiments [25]. A review of several studies in this regard shows that if the result of this parameter is reduced by half after modification, the result of ruts in the pavement will be almost halved [26]. Figure 13 shows that the base bitumen of this study, after the modification process, shows a remarkable improvement in performance.

3.2 Morphological properties of modified bitumen

The images presented in this study were obtained by electron microscopy (TESCAN VEGA 3) with magnifications of 1000 to 4000 times. These images can be seen in Figure 14. The largest size of waste powder particles used in this study is 600 microns, and also the smallest particle size is slightly less than 75 microns, which of course, includes a deficient percentage of particles. In the modified samples, no trace of CR particles of agglomerates is observed, and we see homogeneous, and uniform samples. In addition, in the unmodified sample (Figure 14, left) can be seen the presence of a series of filaments related to the asphalt particles in the bitumen. However, in all the modified samples (Figure 14, right), no trace of these particles is found, what proves that the PPA has succeeded in dissolving the asphalt particles in the bitumen structure, apparently. In general, the combination of waste rubber powder, and PPA can be evaluated for bitumen modification, and from the structural point of view of bitumen, without any problems. However, for more accurate conclusions, more samples need to be evaluated.

4 Conclusions

The objective of this study was to investigate the effect of PPA, and CR on the properties, and performance of bitumen. For this purpose, the two materials were added to the bitumen with different percentages as additives. Then the modified bitumen was tested by classical bitumen tests, bitumen performance grades (PG), and Sharp supplementary grades (PG^+). The results of this research are as follows:

The use of CR at the highest possible level (18% in this study) in the bitumen sample is allowed, and no adverse effect of this additive was observed. The determining condition for its maximum result is the integrity of the sample, and its proper mixing.

- PPA at the level of 1.0% showed positive effects without increasing the weaknesses of bitumen. Given the relatively high price of the product, it can be said that, both from a technical, and economic point of view, the use of 1.0% PPA seems to be suitable for bitumen modification, and of course, it is strongly recommended.

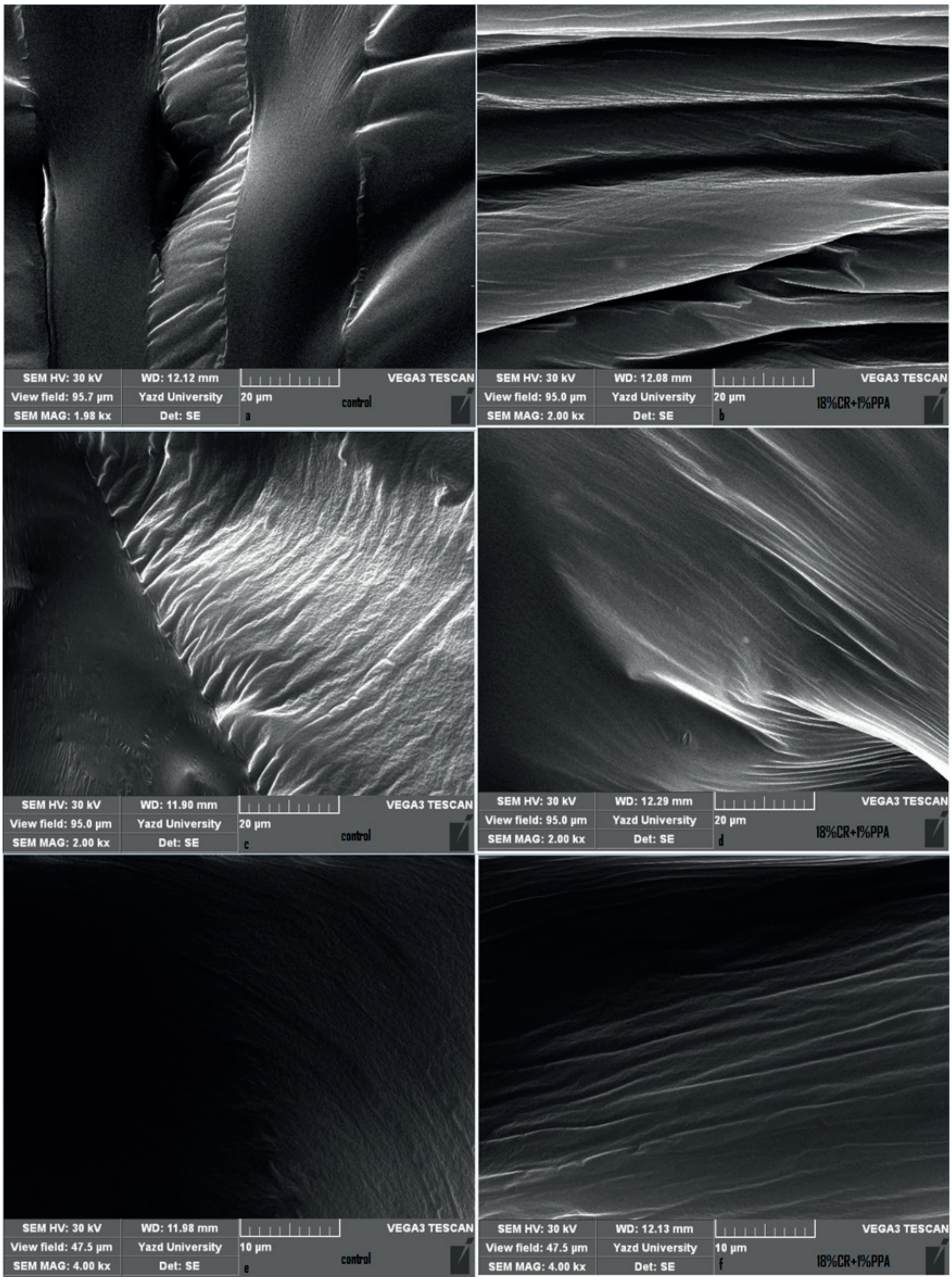

Figure 14 *Scanning electron microscopy images of magnifications of 1000 to 4000 (left), control sample, and (right) of 18% CR+1.0% PPA sample*

- Both additives in this study have succeeded in reducing the sensitivity of bitumen to temperature, and aging. The combination of these two additives is also synergistic, and has highly improved the performance of bitumen. The combination of these two additives has increased the performance class of bitumen at high temperatures up to 4 degrees.
- The samples (18% CR+2.0% PPA), and (18% CR+1.0% PPA) experienced substantial improvement in their rheological properties., and showed remarkable strength, and elasticity. These two samples remained in the range of elastic behavior of bitumen up to 56, and 52° C, respectively, and with further increase in temperature, they have experienced a decline in performance at a meager rate.

- Only the sample containing 18% CR was able to improve the performance of bitumen at low temperatures by one rank, and the combination of bitumen with percentages of PPA content, such as 2.0, 1.75%, and even 1.5%, caused a decrease in performance at low temperatures. Of the samples containing both additives, only the sample (18% CR+1.0% PPA) had no adverse effect on the low-temperature performance of bitumen.
- The results of the LAS test showed that both additives increased the resistance of bitumen to cracking. The test results also revealed the need to pay special attention to the fatigue and cracking of bitumen at the moderate temperature, which is the most common failure related to pavement. The criterion of fatigue at medium temperatures, in addition to the high, and low temperatures of bitumen, should be considered in naming, and introducing the characteristics of bitumen.
- The MSCR test results also showed that the use of additives significantly reduces the non-recoverable creep compliance (j_{nr}) and also increases the bitumen recovery rate (percentage) at high temperatures.
- Examination of the morphological properties of the modified bitumen also showed that in the selected modified composition (18% CR+1.0% PPA), there is no trace of undissolved particles of CR, and this composition is uniform, homogeneous compared to the base bitumen, and is considered successful from this perspective.

Acknowledgements

We would like to thank NaftJey Oil Refinery Company for their help in carrying out the bitumen experiments for this study.

Conflicts of 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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