

Aging and Rheological Properties of Latex and Crumb Rubber Modified Bitumen Using Dynamic Shear Rheometer

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Highlights:

- Natural rubber latex and crumb rubber improve the conventional properties of bitumen.
- 4% of latex and 8% of crumb rubber was found to be the optimum rubber content to improve the rheology of bitumen.
- Latex modified bitumen showed better aging resistance compared to CR-modified bitumen.

Abstract. Rubberized bitumen technology has been applied for a long time to enhance the performance of the asphalt pavement. In this research, the influence of natural rubber (NR) latex and crumb rubber (CR) on the conventional and rheological characteristics of 80/100 penetration grade bitumen before and after aging was compared. Conventional tests of penetration, ring and ball temperature, and ductility were conducted to evaluate the consistency of base bitumen and rubberized bitumen. A dynamic shear rheometer (DSR) test was carried out to evaluate the viscoelastic characteristics of the base and rubberized bitumen. The results showed that the addition of NR latex and CR reduced the penetration grade and increased the ring and ball temperature and ductility of the rubberized bitumen. This indicates that promising enhancement of the bitumen properties can be expected with the addition of NR latex and CR. The rheological properties analysis results showed that addition of CR up to 8% and NR latex up to 4% improved the complex modulus and rutting resistance of the rubberized bitumen compared to conventional bitumen. This indicates that the application of NR latex and CR in bitumen modification is expected to improve the durability of asphalt pavement.

Keywords: crumb rubber; natural rubber latex; rheological properties; rubberized bitumen; rutting resistance.

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1 Introduction

Fatigue and rutting distresses are the main causes of asphalt pavement structure failure. It is important to carry out investigations to increase the service life of pavement by making it able to sustain higher stresses and strains. Rutting is the most common distress in tropical regions. Permanent deformation of asphalt pavement is due to strain over time under different environmental conditions and traffic loads [1-3], causing deterioration of its service life and structural performance. The characteristics of the pavement material used are among the main factors that influence the rutting performance of pavement. Rutting distresses mainly depend on the stiffness of the bitumen and asphalt mixture [4-6].

Over the last few decades, research on polymer modified bitumen has increased, as reported in several studies [3,7]. The utilization of PMB can enhance the performance of asphalt mixes and significantly improve the service life of highways. NR latex is one of the most common thermoplastic elastomers utilized for bitumen modification [8]. Ramez Al-Mansob carried out an investigation into the physical and rheological properties of epoxidized NR-modified bitumen. The results showed that the temperature susceptibility of the base bitumen was enhanced with the addition of NR. It was also found that the rutting resistance and the fatigue resistance of the bitumen at high and intermediate temperatures respectively were improved and that the optimum NR content is 6% by weight of base bitumen [7].

Another study applied NR powder as modifier of bitumen. It was found that NR increased the viscosity and elastic characteristics of the modified bitumen, which indicates that there is a potential improvement of rutting and fatigue resistance. It was also found that the optimum NR to be utilized as bitumen modifier is 7% of base bitumen by weight [9]. Ashiru Sani studied the rheological and microstructure characteristics of NR-modified bitumen incorporating ZycoTherm additive. The results were in accordance with other researches in that the rutting performance of the NR modified bitumen was improved compared to the base bitumen. It was also found that the ZycoTherm material was uniformly dispersed in the bitumen matrix and the NR-modified bitumen with ZycoTherm exhibited excellent rutting resistance [10].

Crumb rubber (CR) is an important modifier to improve the high and intermediate temperature performance of bitumen binders and mixtures. CR is more economical and environmentally friendly compared to other, more expensive commercial polymer modifiers, since it is produced from recycled rubber waste [11,12]. Hajikarimi, *et al.* have conducted a study to investigate the rheological properties of rubberized bitumen. The results showed that there was a significant

improvement in rutting resistance of the modified bitumen, which agreed with the findings of [13,14]. Another study was carried out to compare the effects of CR and SBS modified bitumen on the rheological performance of bitumen. It was concluded that to achieve the same performance, the CR content required is much higher compared to SBS. 8% CR showed the most suitable rheological properties, which were recommended as optimum [15]. Even though several studies have compared CR-modified bitumen to other modifiers, only a very small body of research compares the effects of CR and NR as bitumen modifiers. Therefore, in this study, the effects of CR and NR on the aging and rheological properties of bitumen were compared.

2 Materials and Methods

2.1 Materials

A base bitumen of 80/100 penetration grade was utilized for sample preparation in this study. The bitumen was supplied by Petronas Refinery, Malacca, Malaysia. NR latex and CR were applied as modifiers to produce the rubberized bitumen. The particles sizes of the utilized CR were 0 to 1 millimeters. Table 1 shows the conventional properties of the base bitumen used in this study.

Test -	Standard		Dogulta
	Max.	Min.	Kesuits
Penetration at 25 °C, 0.1 mm	80	100	87
Softening point, °C			42
Ductility at 25 °C, cm	100	-	148
Specific gravity			1.015
Penetration at 25 °C, 0.1 mm	80	100	87

Table 1Conventional properties of the base bitumen.

2.2 Methods

2.2.1 Preparation of Samples

Rubberized bitumen was prepared with 4%, 6% and 8% concentrations of NR latex and CR by weight of base bitumen. To obtain homogeneous rubberized bitumen, a multi-mix high shear mixer was utilized to mix the CR with the base bitumen at a shearing rate of 3000 rpm for a duration of 1 hour at a mixing temperature of 160 $^{\circ}$ C.

The NR latex was also gradually added to the base bitumen and then mixed at a lower shear rate of 800 rpm for 1 hour and at a mixing temperature of 160 °C.

Finally, the samples were carefully stored to be tested for the conventional short-term aging (STA) and the rheological properties.

2.2.2 Rolling Thin Film Oven (RTFO) Test

An RTFO test was conducted according to the ASTM D2872 standard on the base bitumen and the rubberized bitumen to simulate short-term aging of the bitumen during the production and compaction of asphalt mixture in the field. Samples were prepared by pouring a 35 gram of each binder into RTFO glass containers and an open container directly facing a jet of air after placing in an RTFO carriage. The aging process was continued for 1 hour and 25 minutes at 163 °C and a 15 rpm carriage rotating speed.

2.2.3 Conventional Properties Tests

Penetration testing of the unaged bitumen and the STA-bitumen was carried out according to ASTM D5 to investigate the consistency of the rubberized bitumen and control. The test was carried out at a temperature of 25 °C. The penetration value was recorded as the depth of a penetration needle under the conditions stated in the standard.

A ring and ball temperature test was conducted according to the ASTM D36 standard to evaluate the temperature that causes a phase change of the base bitumen and the rubberized bitumen before and after short-term aging. The softening point is the maximum temperature at which bitumen can still support a standard steel ball. Ductility is the tensile deformation and elongation resistance of the bitumen. A ductility test indicates the performance of bitumen at intermediate temperatures. The test was carried out according to the ASTM D113-07 standard. The stretch of the base bitumen and the rubberized bitumen before and after short-term aging was evaluated at a temperature of 25 °C and 5 cm/min deformation speed.

2.2.4 Rheological Properties Test

A Malvern DSR was used to evaluate the rheological behavior of the base bitumen and the rubberized bitumen in this study. An 8-mm diameter parallel plate geometry and a 2-mm gap were utilized. The test temperature range was varied at 20 °C to 40 °C. A temperature sweep test at a controlled strain mode and standard loading frequency of 10 rad/s was performed for all base and rubberized bitumen samples in unaged and STA conditions.

3 Results and Discussion

3.1 Conventional Properties

3.1.1 Penetration

Figures 1 and 2 show the results of the penetration testing of the base bitumen and the rubberized bitumen at unaged and STA conditions. Both the rubberized bitumen modified with CR and NR showed lower penetration values compared to the binder penetration value of the unmodified bitumen. However, CR was more effective for penetration reduction in unaged and STA conditions compared to NR.



Figure 1 Penetration values of CR-modified bitumen in unaged and STA conditions.



Figure 2 Penetration values of NR-modified bitumen in unaged and STA conditions.

In more detail, 8% of CR improved the penetration grade of the base bitumen from 80/100 to 50/60 penetration grade, while 8% of NR improved the grade to 60/70 penetration grade for unaged conditions. However, after STA both showed 40/50 penetration grade. This indicates that CR and NR have a significant influence on the penetration grade of rubberized bitumen. This decrease of penetration can be beneficial for enhancing the performance of rubberized bitumen at intermediate temperature and indicates that the durability and performance of asphalt mixture will increase. The results are in agreement with the literature [7,16].

3.1.2 Softening Point

Figures 3 and 4 illustrate the ring and ball temperature results of the CR and the NR modified bitumen in unaged and STA conditions. In general, it can be stated that all rubberized bitumen showed a ring and ball temperature better than the unmodified bitumen in both unaged and aging conditions. This indicates that the application of CR and NR in bitumen modification is significant in improving the softening point of bitumen. It was also found that the softening point of CR-modified bitumen increased up to 4% CR and then decreased as the CR content continued to increase. On the other hand, the softening point of NR-modified bitumen kept increasing as the NR content increased.



Figure 3 Softening point of CR-modified bitumen in unaged and STA conditions.

The largest softening point improvement was found at 8% NR, i.e. 54.5 °C compared to 43.5 °C at 8% CR for unaged condition, whereas at the softening point for STA condition at 8% for CR and NR was 47.5 and 55 °C respectively. These findings are consistent with [7,16]. It can be seen that a small difference in softening point before aging and after aging for both the CR and the NR modified

bitumen indicates lower temperature sensitivity and higher aging resistance of rubberized bitumen compared to unmodified bitumen. However, the NRmodified bitumen showed better aging resistance compared to the CR-modified bitumen. Therefore, it can be stated that the high-temperature performance of the rubberized bitumen and especially the NR-modified bitumen was improved compared to the base bitumen's performance.



Figure 4 Softening point of NR-modified bitumen in unaged and STA conditions.

3.1.3 Ductility

Figures 5 and 6 present the ductility values of the rubberized bitumen in unaged and aged conditions. It was noticed that as the CR content increased up to 6% the ductility value decreased.



Figure 5 Ductility values of CR-modified bitumen in unaged and STA conditions.



Figure 6 Ductility values of NR-modified bitumen in unaged and STA conditions.

In contrast, the addition of NR improved the ductility values of the bitumen and as the NR content increased the ductility value increased as well. This indicates that the resistance of the NR modified bitumen against deformation at intermediate temperature was better than that of the CR modified bitumen. Therefore, bitumen modified with NR is expected to have an acceptable or better intermediate temperature performance compared to CR modified bitumen.

3.2 Rheological Properties

3.2.1 Isochronal Plots

Isochronal plots of complex modulus (G*) for the CR and the NR modified bitumen in unaged and aged conditions are shown in Figures 7 and 8. It can be seen that the G* of the rubberized bitumen increased compared to the unmodified bitumen. It was also observed that the CR modified bitumen showed a significant increase in complex modulus in unaged and aged conditions compared to NR, however, the NR-rubberized bitumen also showed an increase in complex modulus in the unaged bitumen and then decreased in the aged bitumen. That can be attributed to the good chemical interaction between the rubber and the asphalt binder [17]. It was also found that the lower G* of the NR modified bitumen after aging indicates the ability of NR to improve the aging resistance of bitumen compared to CR [18].



Figure 7 Complex modulus of CR-modified bitumen versus test temperature.



Figure 8 Complex modulus of NR-modified bitumen versus test temperature.

The increase in G^* observed in the rubberized bitumen indicates that the viscoelastic performance of the unmodified bitumen was enhanced due to the addition of CR and NR latex. This enhances the hardness of rubberized bitumen. This increase in complex modulus can be translated to an increase in stiffness, which further enhances the rutting performance [9]. It was also observed that there was a high increase in G^* of the rubberized bitumen at high additive contents and higher temperatures, which also indicates that an increase of rubber content in the bitumen could improve the permanent deformation resistance of

conventional bitumen. This finding agrees with other results reported in the literature [9,19].

The isochronal plots of phase angle (δ) for the CR and the NR modified bitumen are presented in Figures 9 and 10.



Figure 9 Phase angle of CR-modified bitumen versus test temperature.



Figure 10 Phase angle of NR modified bitumen versus test temperature.

It can be seen that the phase angle of all base and rubberized bitumens increased as the test temperature was increased. It is also noted that the rubberized bitumen had a lower δ compared to control. Furthermore, the NR modified bitumen had a

lower δ compared to the CR modified bitumen before and after short-term aging, which can be attributed to the special nature of NR. This reveals that the addition of rubber in terms of CR or NR significantly reduces the phase angle of bitumen, which indicates an enhancement in its elasticity. This decrease in the phase angle of the rubberized bitumen also indicates that the improvement of viscoelastic characteristics of rubberized bitumen at intermediate temperatures will most probably enhance its performance. This result is consistent with what was found in previous studies [9,18].

3.2.2 Rutting Resistance

To predict the rutting performance of the unmodified and the rubberized bitumen, a rutting parameter (G*/sin δ) was utilized based on the specifications of Superpave. The minimum requirements from the specifications of Superpave for the rutting factor are G*/sin $\delta \ge 1$ kPa for unaged bitumen and G*/sin $\delta \ge 2.2$ kPa for STA bitumen. The rutting parameter for unaged and aged conditions of control and the rubberized bitumen at different test temperatures are shown in Figures 11 and 12.



Figure 11 Rutting parameter of CR modified bitumen versus test temperature.

In general, both the CR modified and the NR modified bitumen had high rutting parameters compared to the base bitumen. This can be due to their high compatibility and the interaction of the rubber in the bitumen matrix. It is also noted that the CR modified bitumen showed a significant increase of the rutting parameter in unaged and aged conditions compared to the rutting parameter of the NR modified bitumen. This indicates that rubberized bitumen modified with CR most probably has a higher permanent deformation resistance. This finding can be attributed to the particle size and dissolution of the CR particles in the asphalt matrix. It also indicates that the fatigue performance of NR modified bitumen can be expected to be better than that of CR modified bitumen.

All modified bitumens with CR and NR fulfilled the Superpave requirements for the rutting parameter at the testing temperatures in unaged and aged conditions. This means that all modified bitumens had $G^*/\sin\delta \ge 1$ kPa before aging and $G^*/\sin\delta \ge 2.2$ kPa after aging. It was also found that the rutting factor of the NR modified bitumen after aging was lower compared to that of the CR modified bitumen, which indicates better aging resistance of the NR modified bitumen. These findings are in agreement with previous studies [9,14,18].



Figure 12 Rutting parameter of NR modified bitumen versus test temperature.

4 Conclusions

CR and NR latex were applied as bitumen modifiers to evaluate the conventional and rheological properties of conventional 80/100 penetration grade bitumen. It was concluded that the addition of CR and NR to the base bitumen caused a decrease in the penetration value and an increase in the ring and ball temperature of the rubberized bitumen. Furthermore, the ductility of the bitumen was decreased with the addition of CR and increased with the addition of NR. This indicates an enhancement in hardness and consistency. The viscoelastic characteristics of the bitumen were improved with the addition of CR and NR at various temperatures. It was also found that the NR modified bitumen showed less aging sensitivity compared to the CR modified bitumen and the base bitumen. The rutting resistance of the CR modified bitumen improved as the content of CR increased, whereas the rutting resistance of the NR modified bitumen was enhanced up to 4% NR and then reduced as the NR content kept increasing. In general, it can be stated that rubberized bitumen is expected to perform better in pavement applications compared to conventional bitumen.

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