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Laboratory Evaluation of Storage Stability for CRM Asphalt Binders

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Abstract: This paper conveys the laboratory investigation of the storage stability of CRM binder as a basic study. The CRM binder was produced through the wet process in the laboratory. The percentages of crumb rubber used for rubberized binder were 5%, 10%, 15% and 20%. The samples were prepared according to ASTM D7173. In order to evaluate the properties of each part of the binders, tests were carried out through the rotational viscosity and viscoelasticity, and the separation index was assessed with the G*/sin δ and %rec. In general, the results of this study revealed that (1) the conditioned CRM binders appeared to have higher viscosity in the bottom part compared to the middle and top parts.; (2) similar to the viscosity results, the CRM binders after conditioning showed the highest G*/sin δ value in the bottom part; (3) from the MSCR test, J_{nr} and % rec values are observed to have a similar trend with G*/sin δ results, although some of the data were not measured due to the higher load than the DSR test; and (4) it was discovered that the SI from G*/sin δ generally used was suitable for evaluating the storage stability of CRM asphalt binders, compared to the SI from % rec.

Keywords: CRM binder; storage stability; separation index; viscosity; G*/sin δ

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1. Introduction

With the high demand for improvement in the pavement industry to resist aggravative axis loads and prevent damage, such as rutting and cracking [1], polymer modifiers' application is growing. Asphalt binder modification via different polymers has been contemplated to enhance pavement durability [2]. Common modifiers include crumb rubber modifier (CRM), styrene–butadiene–rubber (SBR), and styrene–butadiene–styrene (SBS), which are considered because of their efficient adhesive and cohesive performance [3] and ability to improve the resistance of the asphalt pavements against defects, such as cracking and rutting, under traffic loading. Since CRM has reasonable performance to enhance the resistance of the asphalt binder while reducing the sound of the asphalt pavement, it is more and more popular in application as an asphalt modifier.

The workability and storage stability of modified asphalt binders is a concern during application, which is caused by the interaction among polymers and bitumen liquid phase, and this manner can be effectively alleviated by the application of warm mix asphalt (WMA) technology [4,5]. Asphalt is structured out of continuous three-dimensional polar molecules that are spread in a fluid of nonpolar or low polar molecules [6]. During the mixing process of asphalt modification, the absorption of the low molecular weight oil fraction of the base asphalt by polymer strands occurs [7]. Based on the weak physical interactions and nonchemical interactions between polymers modifiers and asphalt binders, the modified asphalt binders show poor storage stability at high temperatures [8]. The concern of separation tends to occur in elevated temperatures and causes the uneven concentration of polymer in asphalt. Comparing the density of the modifiers and virgin

asphalt, CRM is likely the most susceptible polymer, which tends to sink in the liquid phase of asphalt while SBR and SBS tend to be floating according to their lower density [2]. Considering the compaction and mixing temperature of the CRM asphalt binders compared to the raw asphalt binders, the chance of separation rises significantly [9]. The separation between the asphalt and the modifier, which may occur after storage, affects the rheological and composition of the top and bottom portions of asphalt binders; consequently, the transportation of it and pumping through to the pipelines will suffer difficulties, and the final product would last lesser than expected.

There are many factors that affect the storage stability of CRM binders, such as the density, concentration, additives, and bitumen characteristics. Besides preparation condition controlling, there are generally two concepts to improve the storage stability of CRM asphalt. The first method is to add various types of chemical compounds into CRM asphalt binder to enhance the bonding forces between the binder and polymer networks [10]. The second option is to treat the CRM surface to achieve the desired interaction between the CRM and asphalt binder network [11,12]. Thermomechanical, thermochemical, biological, plasma and microwave are some ways to treat the CRM surface.

The separation index (SI) calculation for storage stability was performed to determine whether a modified binder was suitable for storage stability according to ASTM-D7173. The calculated values are provided for the softening point (D36/D36M), rheological properties (D7175), and results from multiple stress creep tests (D7405). In particular, the softening point and rheological properties were comprehensively used to obtain SI with various equations [13,14]. However, the SI test results by multiple stress creep tests quantified by the specific particle size and content of CRM are incomplete.

The objective of this study is to quantify the storage stability properties of CRM asphalt binders. According to the rubber content used and the test temperature, in order to check how the storage stability changes, CRM binders were prepared using five different contents (0%, 5%, 10%, 15% and 20%), and the properties after treatment were evaluated based on the viscosity, $G^*/\sin\delta$, J_{nr} and % recovery. Figure 1 shows a flow chart of the experimental design.

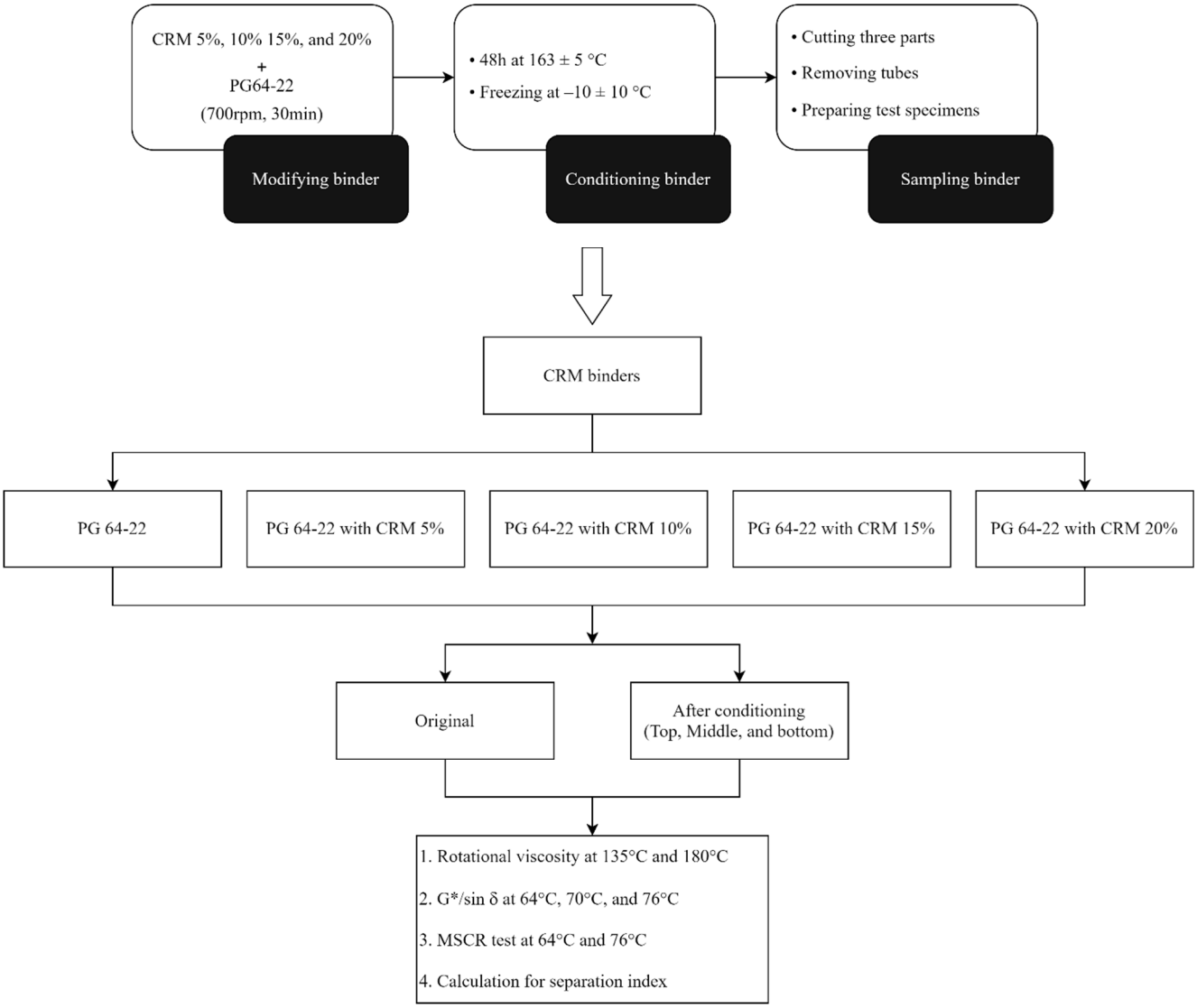


Figure 1. Flow chart of experimental design procedures.

2. Experimental Design

2.1. Materials

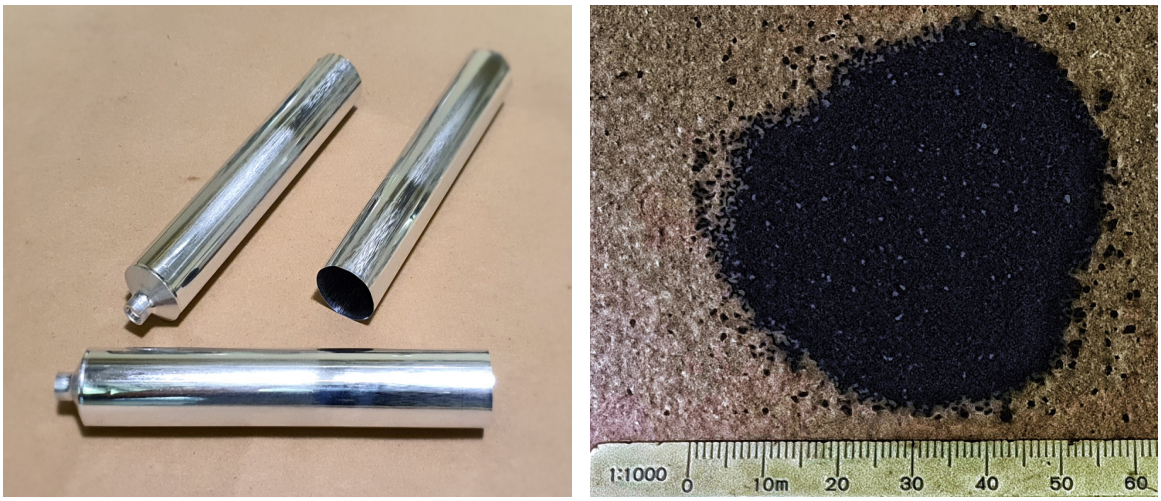
In this study, performance grade (PG) 64-22 asphalt binder was used to make crumb rubber modified (CRM) binders. Table 1 shows the properties of the asphalt binder. Crumb rubber is used in order to modify the binder. Table 2 shows the gradation of material. For the storage stability evaluation, the aluminum tube suggested in ASTM D7173 was used. The aluminum tube and crumb rubber are shown in Figure 2.

Table 1. Properties of base asphalt binder (PG 64-22).

Aging States	Test Properties	Test Result
Unaged binder	Viscosity @ 135 °C (cP) G*/sin δ @ 64 °C (kPa)	538 1.38
RTFO aged residual	G*/sin δ @ 64 °C (kPa)	3.82
RTFO + PAV aged residual	G*sin δ @ 25 °C (kPa) Stiffness @ −12 °C (MPa) m-value @ −12 °C	4402 205 0.323

Table 2. Gradation of CRM used in this study.

Sieve Number (μm) %	% Cumulative Passed
30 (600)	100
40 (425)	91.0
50 (300)	59.1
80 (180)	26.2
100 (150)	18.3
200 (75)	0.0



(**a**) (**b**)

Figure 2. Aluminum tube (**a**) and crumb rubber (**b**).

2.2. Production of CRM Asphalt Binders

CRM asphalt binder was produced through the wet process in the laboratory at 177 °C for 30 min by an open blade mixer at a blending speed of 700 rpm [15,16]. The percentages of crumb rubber used for rubberized binder were 5%, 10%, 15% and 20% by weight of the base binder. In order to ensure that the consistency of the CRM asphalt binder was maintained throughout the study, only one batch of crumb rubber was used in this study.

2.3. Preparation of Test Specimens

The CRM binder sample was thoroughly stirred and poured 50 ± 0.5 g into the vertically held tube according to ASTM D7173 (Figure 3). The modified binder in a sealed aluminum tube was treated in a vertical position for 48 h at a temperature of 163 ± 5 °C. At the end of the treating period, the tube was removed from the oven and placed immediately in the freezer at −10 ± 10 °C, taking care to keep the tube in a vertical position at all times. This tube was placed in the freezer for at least 4 h to completely solidify the sample. After hardening the tube, the tube was removed from the freezer and placed on a hard, flat surface, then cut into three parts of approximately equal length. Each asphalt binder was placed in the 163 ± 5 °C ovens until the asphalt was fluid enough to remove the pieces of aluminum tube, but no more than 30 min. Finally, test specimens were prepared for each test.

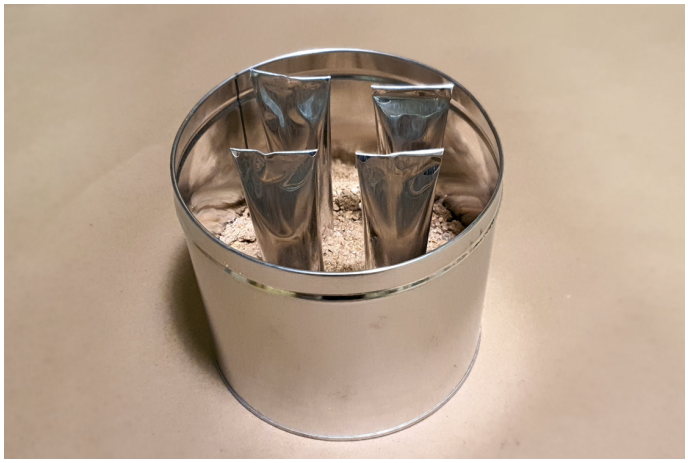


Figure 3. The vertically held tube.

2.4. Binder Evaluation

2.4.1. Rotational Viscosity

In order to evaluate the basic property of CRM binders, a Brookfield rotational viscometer was utilized to measure the viscosity at 135 °C and 180 °C by applying 27 cylindrical spindles and a constant speed of 20 rpm with a weight of 10.5 g of the binder sample. The time taken to acquire data was considered to be 20 min for each sample. Figure 4 shows a set of a rotational viscometer.



Figure 4. The rotational viscometer.

2.4.2. Viscoelasticity

To measure the viscoelasticity of the asphalt binder, dynamic shear rheometer (DSR) was used to result in G*/sin δ, J_{nr}, and % rec. G*/sin δ was calculated from the complex shear modulus (G*) and the sine (δ) of the phase angle at 64 °C, 70 °C and 76 °C temperatures, applying the frequency of 10 rad/s (Test Method D 7175). The multi-stress creep and recovery (Test Method D7405) test was performed in order to draw J_{nr} and % rec according to AASHTO TP 70, loading 3.2 kPa to evaluate the viscoelasticity of the binder at 64 °C and 76 °C temperatures. Figure 5 shows a set of DSR.

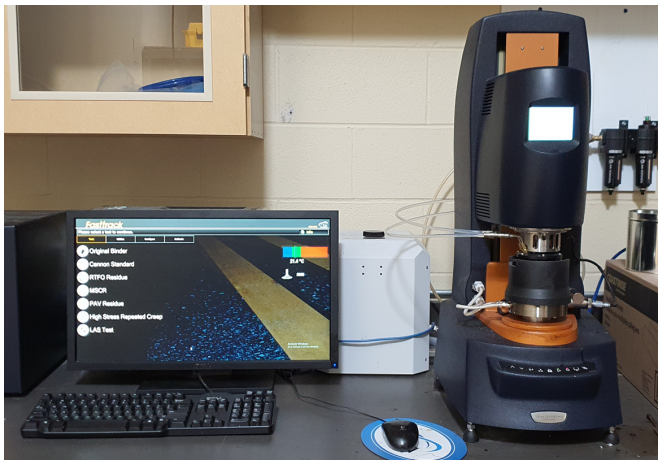


Figure 5. The dynamic shear rheometer.

2.4.3. Separation Index (SI)

It is increasingly common practice to evaluate both fractions in terms of their rheological properties from the DSR test as an alternative to the softening point and penetration. In this study, the G*/sin δ ratio, referencing the Superpave specifications, was used. For instance, the previous research [17,18] mentioned the ratio, using Equation (1), where $(G^*/\sin\delta)_{max}$ presents the higher value between the top and bottom parts and $(G^*/\sin\delta)_{avg}$ is the average value for both parts. In addition, the SI was calculated and evaluated with the %rec, using Equation (2).

$$Separation\ index = \frac{(G*/\sin\delta)_{max} - (G*/\sin\delta)_{avg}}{(G*/\sin\delta)_{avg}} \quad (1)$$

$$Separation\ index = \frac{(\%\ rec)_{max} - (\%\ rec)_{avg}}{(\%\ rec)_{avg}} \quad (2)$$

3. Results and Discussions

3.1. Rotational Viscosity

The viscosity of the asphalt binder affects the workability of the production, delivery and compaction of asphalt mixtures. If the viscosity is too high, it may be difficult to achieve the optimum in-field density, which is also related to the pavement performance life. Figure 6 indicates the viscosity values at 135 °C and 180 °C for the original CRM asphalt binders immediately after blending. It is clear that the viscosity of the CRM binders decreased as the testing temperature increased, and as the CRM content increased, the binder viscosity increased, as expected. At 135 °C, compared to 0% CRM binder (control), the 20% CRM binder showed an increase in viscosity of approximately 7 times. In particular, in the case of 20% CRM binder, it showed a viscosity value higher than 3000 cP at 135 °C, meaning that the production and construction temperatures for 20% CRM mixtures must be increased for proper workability and compaction.

Comparing the viscosity values at 180 °C, compared with the control binder, 20% CRM asphalt resulted in a viscosity increase of about 10 times or more. It was found that the viscosity value at 180 °C decreased, but the viscosity increase rate was higher compared to 135 °C. This result is thought to be because the viscosity of rubber powder is less sensitive to temperature increase.

Figure 7 depicts the viscosity results at 135 °C of CRM binders of top, middle and bottom parts after conditioning. The data of the CRM binders after conditioning for 48 h in an oven at 163 °C showed that the bottom part had a higher viscosity than the top and middle parts in all samples. As expected, rubber particles appear to have sunk to the bottom. In the top part, as the CRM content increased, the viscosity value gradually

increased (0% CRM binder—539, 5% CRM—575, 10% CRM—638, 15% CRM—694 and 20% CRM—2194 cP). Similar to the results in the top part, the middle part showed a tendency to increase in viscosity as the CRM contents increased. In particular, the 15% CRM binder resulted in 11 times higher viscosity, compared to the top part, and the 20% CRM binders showed a viscosity value of 10,000 cP or more.

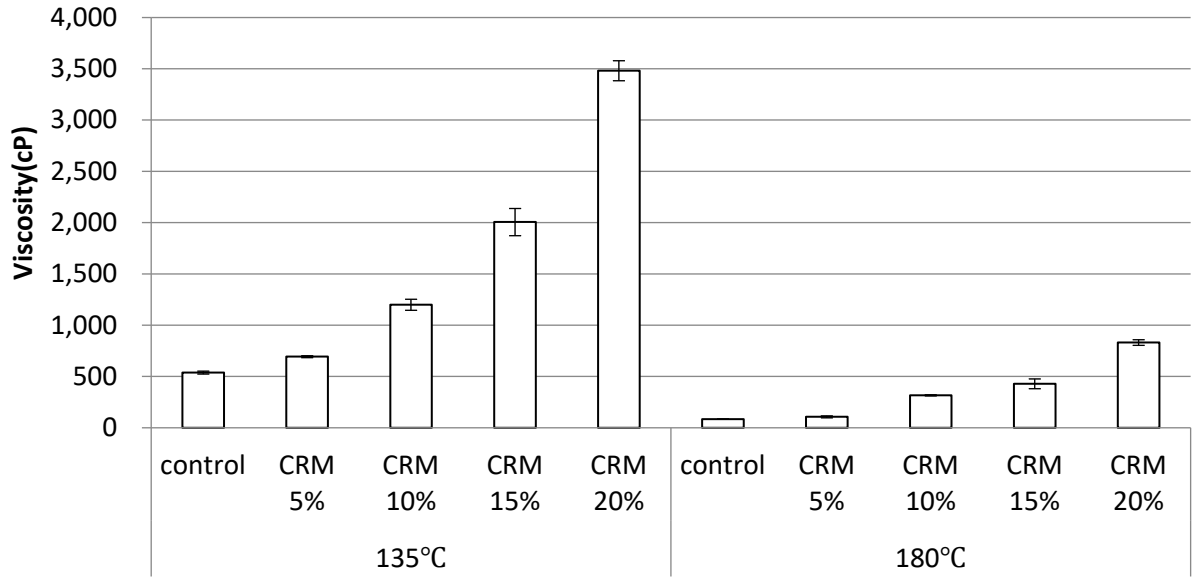


Figure 6. Viscosity of the CRM asphalt binders at 135 °C and 180 °C.

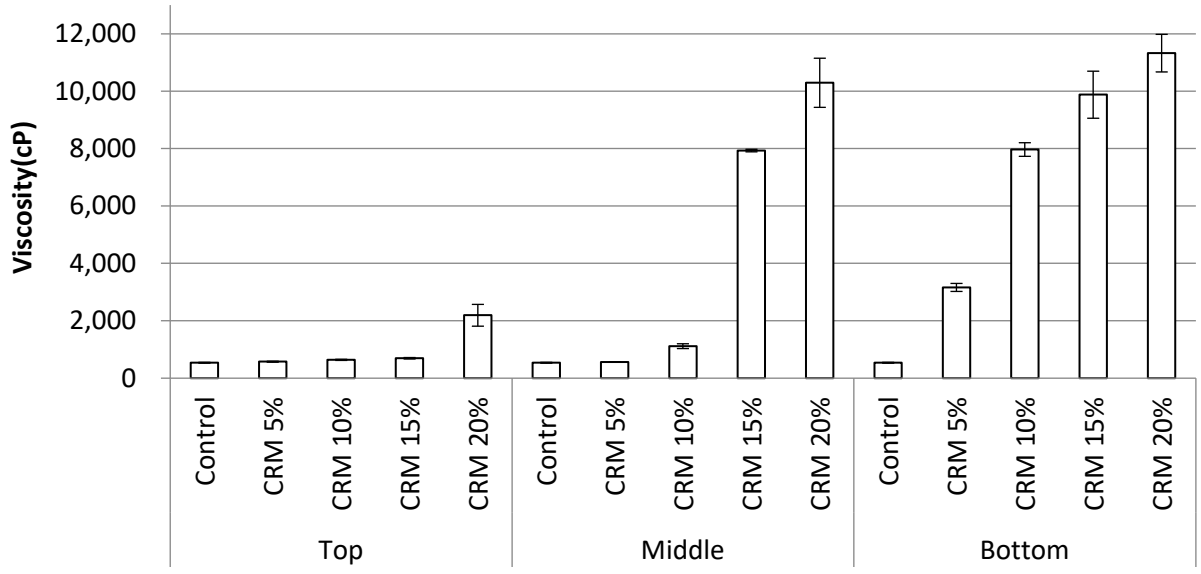


Figure 7. Viscosity at 135 °C of the CRM binders of top, middle and bottom parts after conditioning.

As for the viscosity of the bottom part, it was confirmed that the viscosity of the 20% CRM binder increased by approximately 21 times, compared to that of 0% CRM binder (the viscosity of the CRM binder increased by about 7 times at the same temperature before the conditioning process). This result is considered to have caused the rubber particles to settle to the middle and bottom during the treatment process, resulting in an increase in viscosity.

Figure 8 shows the results of the viscosity test of the conditioned CRM binders at 180 °C. The lowest value appeared in the top part and there was an exhibited tendency to increase in viscosity toward the middle part and the bottom part. In general, in the case of the wet process, the CRM binder is continuously rotated using a propeller until just before

making the CRM asphalt mixture in order to prevent the settlement of the rubber particles. For this reason, the CRM binder should be made immediately prior to the production of the CRM mixture in the asphalt plant.

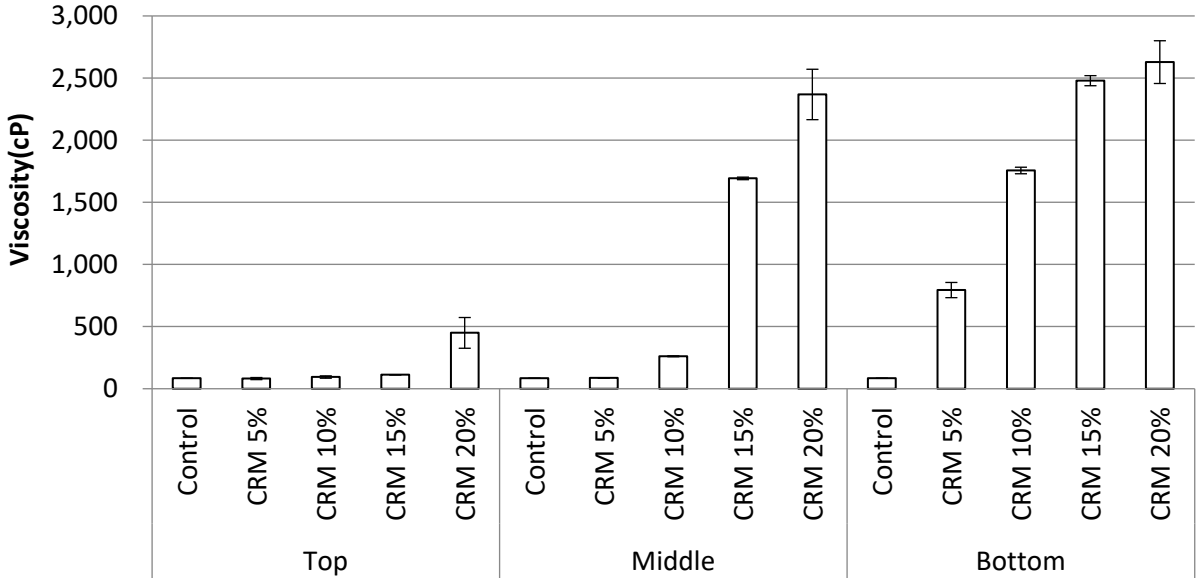


Figure 8. Viscosity at 180 °C of the CRM binders of top, middle and bottom parts after conditioning.

Using one-way analysis of variance, the statistical significance of the CRM binder viscosity values was examined depending on the top, middle and bottom parts after the treatment (Table 3). In the top part, there was no statistically significant difference in viscosity values up to 15% CRM at both temperatures. In the middle part, it was evident that the viscosity values showed a significant difference at 15% or more CRM binders regardless of the measured temperatures. In the bottom part, the viscosity values with statistical significance were confirmed in all CRM contents.

Table 3. Statistical analysis results of the viscosity of CRM binders as a function of top, middle and bottom parts ($\alpha = 0.05$).

		Viscosity at 135 °C																			
		Original (%)					**Top (%)**					**Middle (%)**					**Bottom (%)**				
		0	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**
Original (%)	0	-	N	N	S	S	N	N	N	N	S	N	N	N	S	S	N	S	S	S	S
	5		-	N	S	S	N	N	N	N	S	N	N	N	S	S	N	S	S	S	S
	10			-	N	S	N	N	N	N	S	N	N	N	S	S	N	S	S	S	S
	15				-	S	S	S	S	S	N	S	S	N	S	S	S	S	S	S	S
	20					-	S	S	S	S	S	S	S	S	S	S	S	N	S	S	S
Top (%)	0						-	N	N	N	S	N	N	N	S	S	N	S	S	S	S
	5							-	N	N	S	N	N	N	S	S	N	S	S	S	S
	10								-	N	S	N	N	N	S	S	N	S	S	S	S
	15									-	S	N	N	N	S	S	N	S	S	S	S
	20										-	S	S	N	S	S	S	N	S	S	S

Table 3. *Cont.*

		Viscosity at 135 °C																			
		Original (%)					Top (%)					Middle (%)					Bottom (%)				
		0	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**
Middle (%)	0											-	N	N	S	S	N	S	S	S	S
	5												-	N	S	S	N	S	S	S	S
	10													-	S	S	N	S	S	S	S
	15														-	S	S	S	N	S	S
	20															-	S	S	S	N	S
Bottom (%)	0																-	S	S	S	S
	5																	-	S	S	S
	10																		-	S	S
	15																			-	S
	20																				-
		Viscosity at 180 °C																			
Original (%)	0	-	N	N	S	S	N	N	N	N	S	N	N	N	S	S	N	S	S	S	S
	5		-	N	S	S	N	N	N	N	S	N	N	N	S	S	N	S	S	S	S
	10			-	N	S	N	N	N	N	S	N	N	N	S	S	N	S	S	S	S
	15				-	S	S	S	S	S	N	S	S	N	S	S	S	S	S	S	S
	20					-	S	S	S	S	S	S	S	S	S	S	S	N	S	S	S
Top (%)	0						-	N	N	N	S	N	N	N	S	S	N	S	S	S	S
	5							-	N	N	S	N	N	N	S	S	N	S	S	S	S
	10								-	N	S	N	N	N	S	S	N	S	S	S	S
	15									-	S	N	N	N	S	S	N	S	S	S	S
	20										-	S	S	N	S	S	S	N	S	S	S
Middle (%)	0											-	N	N	S	S	N	S	S	S	S
	5												-	N	S	S	N	S	S	S	S
	10													-	S	S	N	S	S	S	S
	15														-	S	S	S	N	S	S
	20															-	S	S	S	N	S
Bottom (%)	0																-	S	S	S	S
	5																	-	S	S	S
	10																		-	S	S
	15																			-	S
	20																				

N: non-significant, S: significant.

3.2. G/sin δ Property*

G*/sin δ values obtained from DSR equipment are most commonly used to evaluate the storage stability of polymer-modified asphalt (PMA) binders. Figure 9 illustrates the G*/sin δ values measured immediately after making the CRM binders. As expected, the values tended to increase as the CRM content increased. In addition, the value decreased as the testing temperature increased from 64 °C to 70 °C and to 76 °C. It was expected that a high performance grade (PG) of 76 or higher could be obtained when the rubber content was added at more than 10%.

The G*/sin δ values for the top, middle and bottom parts of CRM binders after conditioning at 163 °C for 48 h were measured at 64 °C, 70 °C and 76 °C (Figures 10–12). As expected, as the testing temperature increased, the measured value decreased regardless of the CRM contents. Additionally, as the rubber particle content increased, the G*/sin δ value increased at all testing temperatures. However, the data for top, middle and bottom parts showed a different trend, compared with the original CRM binders before conditioning. First, the top G*/sin δ value maintained a similar level up to the 15% CRM content, and then increased more than twice at 20% CRM. In the middle part, the value was similar to the top part up to 5%, but the value increased from 10% CRM, and the value was approximately three times higher than the top part in 15% or more. The bottom part showed a higher

value compared to the top part and middle part from 5% CRM, and the value increased rapidly at 10% CRM, and thereafter, the value showed a tendency to gradually increase.

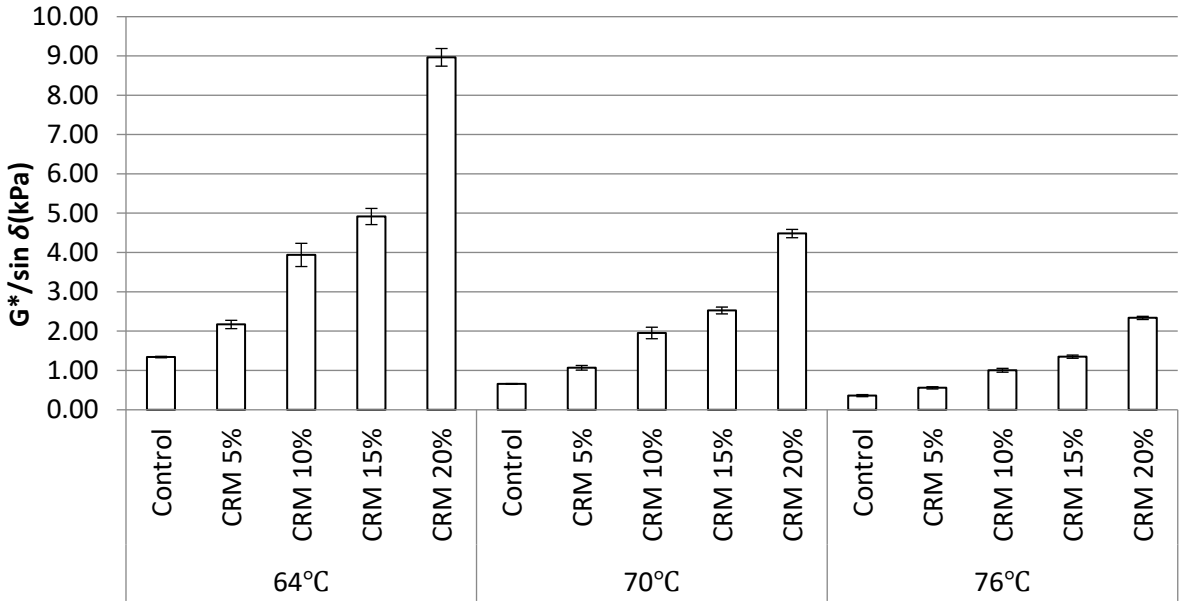


Figure 9. G*/sin δ values of the CRM asphalt binders at 64 °C, 70 °C and 76 °C.

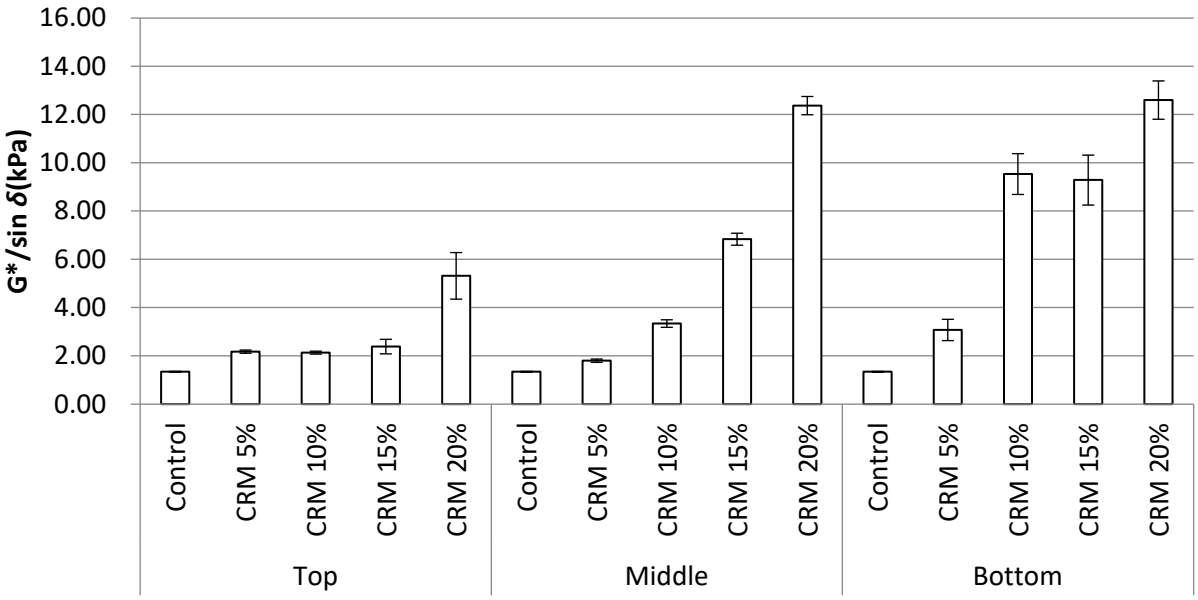


Figure 10. G*/sin δ at 64 °C of the CRM asphalt binders of top, middle and bottom parts after conditioning.

Comparing the middle and bottom parts, the bottom part resulted in a higher value up to 15% CRM but revealed a similar value at 20%. This is thought to be because, at 20% CRM binder, the rubber particles sunk from the top part were distributed at a similar rate in the middle and bottom parts. In general, the measured values for the top, middle and bottom parts at 70 °C and 76 °C demonstrated the same trend as G*/sin δ at 64 °C.

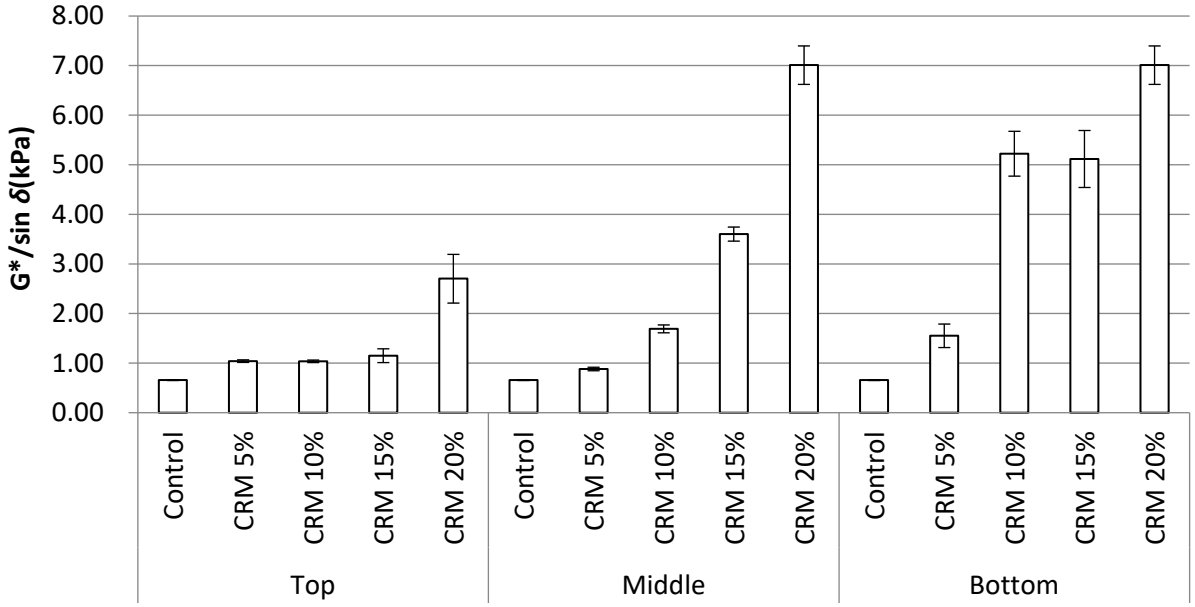


Figure 11. G*/sin δ at 70 °C of the CRM asphalt binders of top, middle and bottom parts after conditioning.

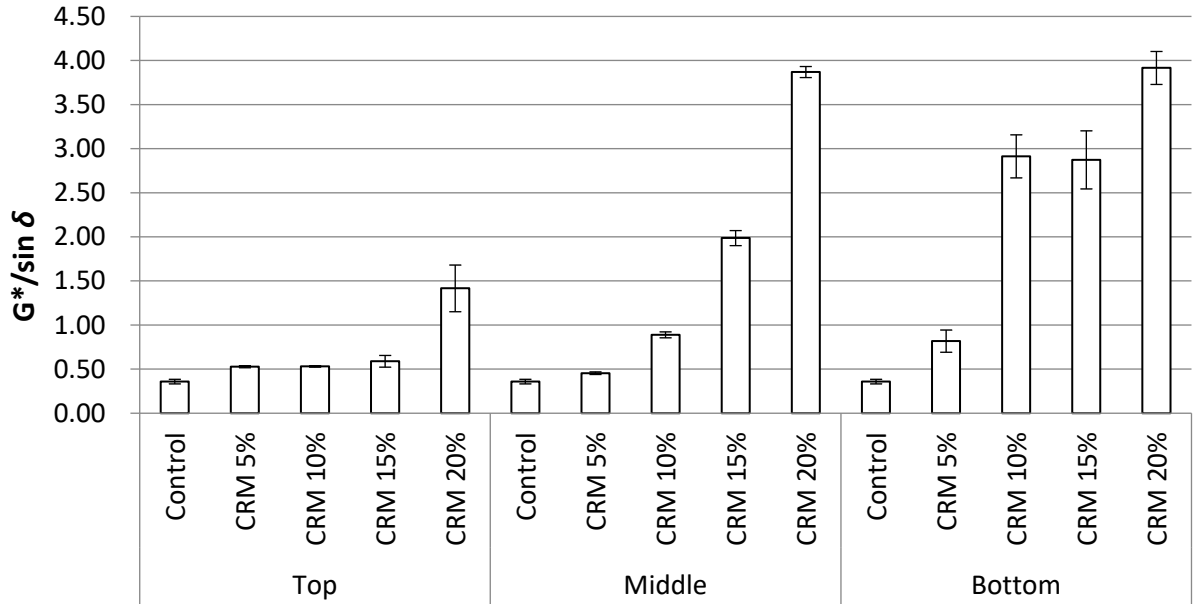


Figure 12. G*/sin δ at 76 °C of the CRM asphalt binders of top, middle and bottom parts after conditioning.

The statistical significance of the change in the CRM contents was examined, comparing the original condition to the top, middle, and bottom parts, using one-way analysis of variance (Tables 4–6). In general, the significant difference within each content of the original condition at all temperatures was observed. In the top part, it was confirmed that the measured values were not statistically significant up to 10% CRM content within each testing temperature. In the bottom part, the values of 10% and 15% CRM binders were statistically similar at the 95% confidence level.

Table 4. Statistical analysis results of the G*/sin δ at 64 °C of CRM binders as a function of top, middle and bottom parts (α = 0.05).

		Original (%)					**Top (%)**					**Middle (%)**					**Bottom (%)**				
		0	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**
Original (%)	0	-	S	S	S	S	N	N	N	S	S	N	N	S	S	S	N	S	S	S	S
	5		-	S	S	S	S	N	N	N	S	S	N	S	S	S	S	S	S	S	S
	10			-	S	S	S	S	S	S	S	S	S	N	S	S	S	N	S	S	S
	15				-	S	S	S	S	S	N	S	S	S	S	S	S	S	S	S	S
	20					-	S	S	S	S	S	S	S	S	S	S	S	S	S	S	S
Top (%)	0						-	N	N	S	S	N	N	S	S	S	N	S	S	S	S
	5							-	N	N	S	N	N	S	S	S	N	S	S	S	S
	10								-	N	S	N	N	S	S	S	N	S	S	S	S
	15									-	S	S	N	S	S	S	S	S	S	S	S
	20										-	S	S	S	S	S	S	S	S	S	S
Middle (%)	0											-	N	S	S	S	N	S	S	S	S
	5												-	S	S	S	N	S	S	S	S
	10													-	S	S	S	N	S	S	S
	15														-	S	S	S	S	S	S
	20															-	S	S	S	S	S
Bottom (%)	0																-	S	S	S	S
	5																	-	S	S	S
	10																		-	N	S
	15																			-	S
	20																				-

N: non-significant, S: significant.

Table 5. Statistical analysis results of the G*/sin δ at 70 °C of CRM binders as a function of top, middle and bottom parts (α = 0.05).

		Original (%)					**Top (%)**					**Middle (%)**					**Bottom (%)**				
		0	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**
Original (%)	0	-	S	S	S	S	N	N	N	S	S	N	N	S	S	S	N	S	S	S	S
	5		-	S	S	S	S	N	N	N	S	S	N	S	S	S	S	S	S	S	S
	10			-	S	S	S	S	S	S	S	S	S	N	S	S	S	N	S	S	S
	15				-	S	S	S	S	S	N	S	S	S	S	S	S	S	S	S	S
	20					-	S	S	S	S	S	S	S	S	S	S	S	S	S	S	S
Top (%)	0						-	N	N	S	S	N	N	S	S	S	N	S	S	S	S
	5							-	N	N	S	N	N	S	S	S	N	S	S	S	S
	10								-	N	S	N	N	S	S	S	N	S	S	S	S
	15									-	S	S	N	S	S	S	S	S	S	S	S
	20										-	S	S	S	S	S	S	S	S	S	S
Middle (%)	0											-	N	S	S	S	N	S	S	S	S
	5												-	S	S	S	N	S	S	S	S
	10													-	S	S	S	N	S	S	S
	15														-	S	S	S	S	S	S
	20															-	S	S	S	S	S
Bottom (%)	0																-	S	S	S	S
	5																	-	S	S	S
	10																		-	N	S
	15																			-	S
	20																				-

N: non-significant, S: significant.

Table 6. Statistical analysis results of the G*/sin δ at 76 °C of CRM binders as a function of top, middle and bottom parts (α = 0.05).

		Original (%)					Top (%)					Middle (%)					Bottom (%)				
		0	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**
Original (%)	0	-	N	S	S	S	N	N	N	S	S	N	N	S	S	S	N	S	S	S	S
	5		-	S	S	S	N	N	N	N	S	N	N	S	S	S	N	S	S	S	S
	10			-	S	S	S	S	S	S	S	S	S	N	S	S	S	N	S	S	S
	15				-	S	S	S	S	S	N	S	S	S	S	S	S	S	S	S	S
	20					-	S	S	S	S	S	S	S	S	S	S	S	S	S	S	S
Top (%)	0						-	N	N	S	S	N	N	S	S	S	N	S	S	S	S
	5							-	N	N	S	N	N	S	S	S	N	S	S	S	S
	10								-	N	S	N	N	S	S	S	N	S	S	S	S
	15									-	S	S	N	S	S	S	S	S	S	S	S
	20										-	S	S	S	S	S	S	S	S	S	S
Middle (%)	0											-	S	S	S	S	N	S	S	S	S
	5												-	S	S	S	N	S	S	S	S
	10													-	S	S	S	N	S	S	S
	15														-	S	S	S	S	S	S
	20															-	S	S	S	S	N
Bottom (%)	0																-	S	S	S	S
	5																	-	S	S	S
	10																		-	N	S
	15																			-	S
	20																				-

N: non-significant, S: significant.

3.3. Multi-Stress Creep and Recovery Property

MSCR (Test Method D7405) is an alternate test to the DSR test method used for the storage stability. The MSCR test was performed at 64 °C and 76 °C according to AASHTO TP 70 by loading 3.2 kPa to evaluate the viscoelasticity of the CRM binders under more extreme conditions than the DSR test.

Figure 13 presents the results of J_{nr} and % recovery of control CRM binders. In general, increasing the CRM contents made it possible to decline the J_{nr} value and enhance % rec, which means the higher the CRM contents, the higher the viscoelasticity of the binder. In more detail, the data from CRM contents of 0% and 5% were not measured due to the samples' low viscosity at 64 °C. The J_{nr} value decreased steadily, with the data reaching 2.52 in 10% CRM, 1.75 in 15% CRM, and 0.41 in 20% CRM, while the data for % recovery increased gradually by up to over 12.3% from 2.5%. In addition, by increasing the testing temperature to 76 °C, the data were not measured until 15% CRM binders as the binders became softer at a higher temperature. The value of 20% CRM binder was only measured.

Figures 14 and 15 show the J_{nr} and % rec of the CRM asphalt binders at 64 °C and 76 °C, respectively. At 64 °C, no values were recorded in the binders, except for 20% at the top part. The values were measured from 10% in the middle part and from 5% in the bottom part. It is considered that the viscosity at 64 °C increased from the top to the middle to the bottom. As the CRM content increased, the J_{nr} value decreased, and the % rec value increased. In the case of 20% CRM binder, the middle part and the bottom part resulted in similar values.

When 76 °C was used as the testing temperature, it was not measured in the top part. The J_{nr} and % rec of the CRM asphalt binders were obtained from 15% in the middle part and from 10% in the bottom part. It is thought that this is because the viscosity of CRM binders became lower compared with 64 °C. The general trend was similar to that at 64 °C.

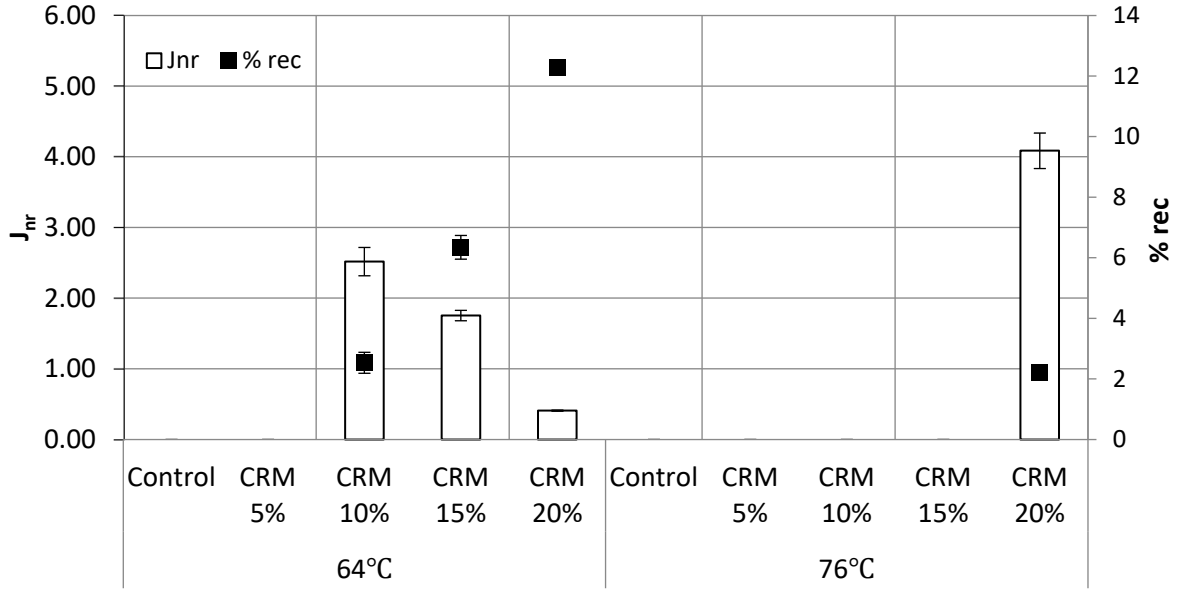


Figure 13. J_{nr} and % rec of the CRM asphalt binders at 64 °C and 76 °C.

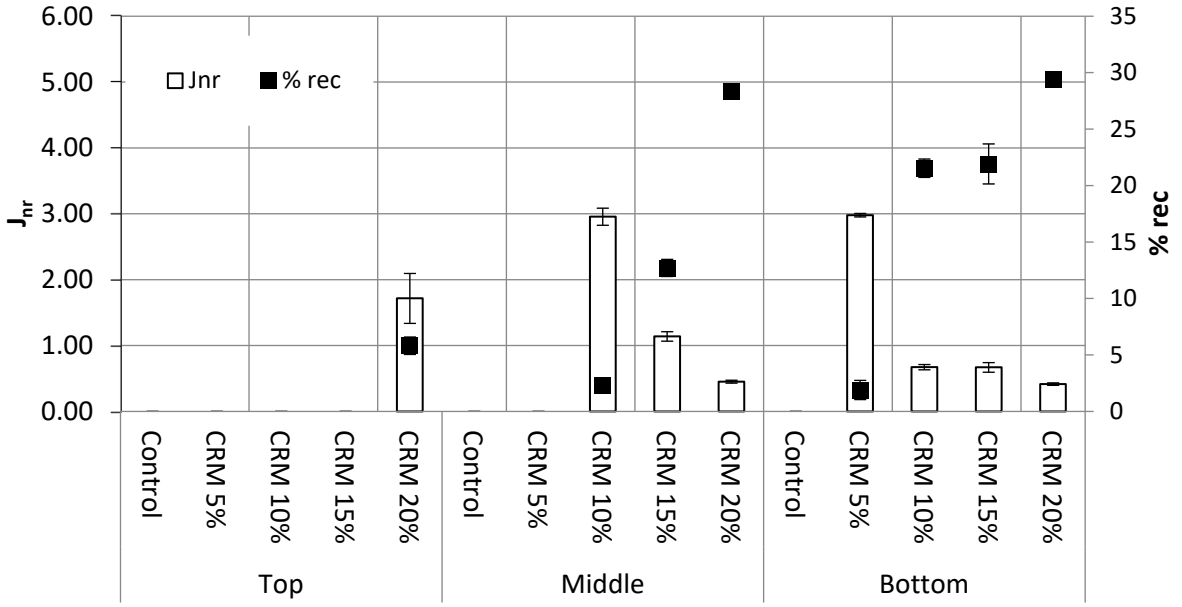


Figure 14. J_{nr} and % rec at 64 °C of the CRM asphalt binders of top, middle and bottom parts after conditioning.

Using one-way analysis of variance, the statistical significance of the change in the J_{nr} and % rec was examined, comparing the original condition to the top, middle, and bottom parts (Tables 7 and 8). In general, the J_{nr} values within the original condition from the MSCR test at 64 °C were significantly different depending on CRM contents. For the conditioned CRM binders of the top, middle, and bottom parts, the significant difference was observed within each part, compared to the original condition from the top to the bottom. In the case of statistical analysis for J_{nr} at 76 °C, there was an insignificant difference within each part of the original, top, and bottom due to the non-measured results. The case of % rec showed a similar trend to the J_{nr} analysis.

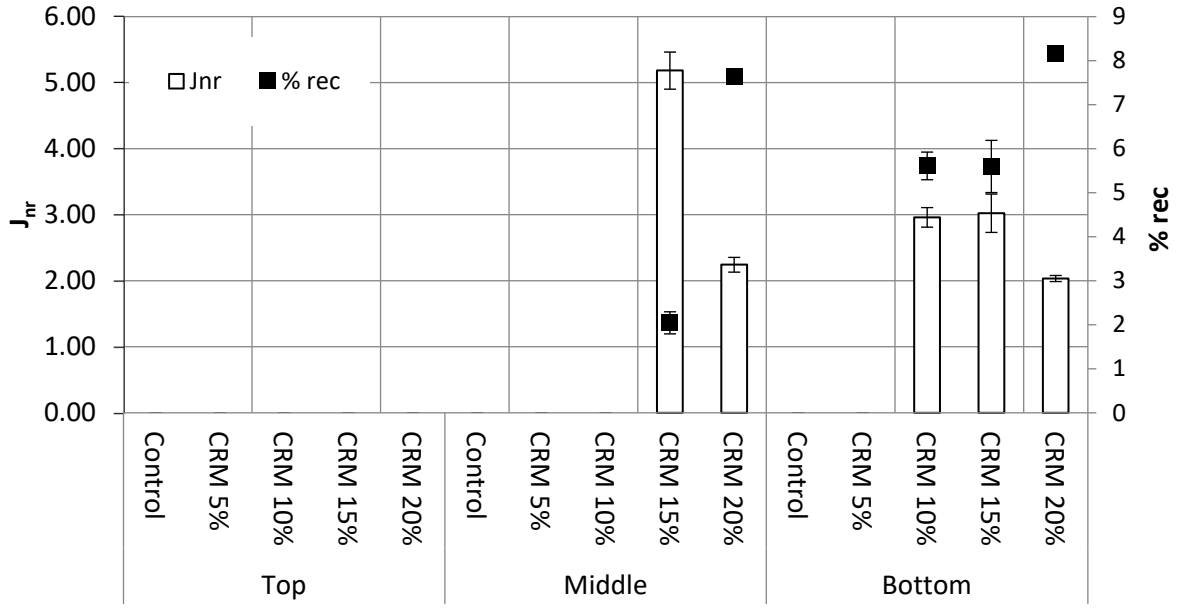


Figure 15. J_{nr} and % rec at 76 °C of the CRM asphalt binders of top, middle and bottom parts after conditioning.

Table 7. Statistical analysis results of the J_{nr} of CRM binders as a function of top, middle and bottom parts ($\alpha = 0.05$).

		J_{nr} at 64 °C																			
		Original (%)					Top (%)					Middle (%)					Bottom (%)				
		0	5	10	15	20	0	5	10	15	20	0	5	10	15	20	0	5	10	15	20
Original (%)	0	-	N	S	S	S	N	N	N	N	S	N	N	S	S	S	N	S	S	S	S
	5		-	S	S	S	N	N	N	N	S	N	N	S	S	S	N	S	S	S	S
	10			-	S	S	S	S	S	S	S	S	S	S	S	S	S	S	S	S	S
	15				-	S	S	S	S	S	N	S	S	S	S	S	S	S	S	S	S
	20					-	S	S	S	S	S	S	S	S	S	N	S	S	N	N	N
Top (%)	0						-	N	N	N	S	N	N	S	S	S	N	S	S	S	S
	5							-	N	N	S	N	N	S	S	S	N	S	S	S	S
	10								-	N	S	N	N	S	S	S	N	S	S	S	S
	15									-	S	N	N	S	S	S	N	S	S	S	S
	20										-	S	S	S	S	S	S	S	S	S	S
Middle (%)	0											-	N	S	S	S	N	S	S	S	S
	5												-	S	S	S	N	S	S	S	S
	10													-	S	S	S	N	S	S	S
	15														-	S	S	S	S	S	S
	20															-	S	S	N	N	N
Bottom (%)	0																-	S	S	S	S
	5																	-	S	S	S
	10																		-	N	N
	15																			-	N
	20																				-

Table 7. *Cont.*

		J_{nr} at 64 °C																			
		Original (%)					Top (%)					Middle (%)					Bottom (%)				
		0	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**
		J_{nr} at 76 °C																			
Original (%)	0	-	N	N	N	S	N	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	5		-	N	N	S	N	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	10			-	N	S	N	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	15				-	S	N	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	20					-	S	S	S	S	S	S	S	S	S	S	S	S	S	S	S
Top (%)	0						-	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	5							-	N	N	N	N	N	N	S	S	N	N	S	S	S
	10								-	N	N	N	N	N	S	S	N	N	S	S	S
	15									-	N	N	N	N	S	S	N	N	S	S	S
	20										-	N	N	N	S	S	N	N	S	S	S
Middle (%)	0											-	N	N	S	S	N	N	S	S	S
	5												-	N	S	S	N	N	S	S	S
	10													-	S	S	N	N	S	S	S
	15														-	S	S	S	S	S	S
	20															-	S	S	S	S	S
Bottom (%)	0																-	N	S	S	S
	5																	-	S	S	S
	10																		-	N	S
	15																			-	S
	20																				-

N: non-significant, S: significant.

Table 8. Statistical analysis results of the % rec of CRM binders as a function of top, middle and bottom parts ($\alpha = 0.05$).

		% rec at 64 °C																			
		Original (%)					Top (%)					Middle (%)					Bottom (%)				
		0	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**	**0**	**5**	**10**	**15**	**20**
Original (%)	0	-	N	S	S	S	N	N	N	N	S	N	N	S	S	S	N	S	S	S	S
	5		-	S	S	S	N	N	N	N	S	N	N	S	S	S	N	S	S	S	S
	10			-	S	S	S	S	S	S	S	S	S	N	S	S	S	N	S	S	S
	15				-	S	S	S	S	S	N	S	S	S	S	S	S	S	S	S	S
	20					-	S	S	S	S	S	S	S	S	N	S	S	S	S	S	S
Top (%)	0						-	N	N	N	S	N	N	S	S	S	N	S	S	S	S
	5							-	N	N	S	N	N	S	S	S	N	S	S	S	S
	10								-	N	S	N	N	S	S	S	N	S	S	S	S
	15									-	S	N	N	S	S	S	N	S	S	S	S
	20										-	S	S	S	S	S	S	S	S	S	S
Middle (%)	0											-	N	S	S	S	N	S	S	S	S
	5												-	S	S	S	N	S	S	S	S
	10													-	S	S	S	N	S	S	S
	15														-	S	S	S	S	S	S
	20															-	S	S	S	S	N
Bottom (%)	0																-	S	S	S	S
	5																	-	S	S	S
	10																		-	N	S
	15																			-	S
	20																				-

Table 8. *Cont.*

		% rec at 64 °C																			
		Original (%)					Top (%)					Middle (%)					Bottom (%)				
		0	5	10	15	20	0	5	10	15	20	0	5	10	15	20	0	5	10	15	20
		% rec at 76 °C																			
Original (%)	0	-	N	N	N	S	N	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	5		-	N	N	S	N	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	10			-	N	S	N	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	15				-	S	N	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	20					-	S	S	S	S	S	S	S	S	N	S	S	S	S	S	S
Top (%)	0						-	N	N	N	N	N	N	N	S	S	N	N	S	S	S
	5							-	N	N	N	N	N	N	S	S	N	N	S	S	S
	10								-	N	N	N	N	N	S	S	N	N	S	S	S
	15									-	N	N	N	N	S	S	N	N	S	S	S
	20										-	N	N	N	S	S	N	N	S	S	S
Middle (%)	0											-	N	N	S	S	N	N	S	S	S
	5												-	N	S	S	N	N	S	S	S
	10													-	S	S	N	N	S	S	S
	15														-	S	S	S	S	S	S
	20															-	S	S	S	S	S
Bottom (%)	0																-	N	S	S	S
	5																	-	S	S	S
	10																		-	N	S
	15																			-	S
	20																				-

N: non-significant, S: significant.

3.4. Storage Stability Results

In order to evaluate the storage stability of CRM binders, G*/sin δ and % rec of the top and bottom parts after conditioning, were used to calculate the separation index (SI) suggested by Superpave.

As shown in Table 9, as the testing temperature increased from 64 °C to 76 °C, the SI value generally increased in all contents of the CRM binders. It showed the lowest value at 5% CRM, and the highest at 10%. The values were slightly lower at 15% and further decreased at 20%. The reason for the highest value at 10% is thought to be that the rubber particles that sunk from the top part were mainly at the bottom part. The relatively lower value at 20% is considered to be due to the high content of CRM used, and there was no more space to sink at the bottom part.

Table 9. Separation index from G*/sin δ of CRM binders.

Binder	G*/sin δ (kPa)			
	Temperature	Top	Bottom	% Separation
CRM 5%	64 °C	2.17	3.08	17
	70 °C	1.04	1.55	20
	76 °C	0.53	0.82	21
CRM 10%	64 °C	2.13	9.53	63
	70 °C	1.04	5.22	67
	76 °C	0.53	2.91	69
CRM 15%	64 °C	2.38	9.29	59
	70 °C	1.15	5.12	63
	76 °C	0.59	2.87	66

Table 9. *Cont.*

Binder	G*/sin δ (kPa)			
	Temperature	Top	Bottom	% Separation
CRM 20%	64 °C	5.31	12.60	41
	70 °C	2.70	7.01	44
	76 °C	1.42	3.92	47

Table 10 shows the SI value for % rec after conditioning. It was observed that the SI for % rec is not suitable for evaluating the storage stability of CRM binders used in this study.

Table 10. Separation index from % rec of CRM binders.

Binder	% rec			
	Temperature	Top	Bottom	% Separation
CRM 5%	64 °C	0.00	1.90	100
	76 °C	0.00	0.00	None
CRM 10%	64 °C	0.00	21.54	100
	76 °C	0.00	5.61	100
CRM 15%	64 °C	0.00	21.91	100
	76 °C	0.00	5.60	100
CRM 20%	64 °C	5.83	29.39	67
	76 °C	0.00	8.15	100

4. Summary and Conclusions

In order to investigate the storage stability of CRM asphalt binders containing 5%, 10%, 15% and 20%, the binders were conditioned for 48 h in the oven at 163 °C. The tests were carried out using the rotational viscosity and the dynamic shear rheometer to determine the properties and separation index (SI) of CRM binders. From these results, the following conclusions were drawn for the storage stability in this study.

(1) The addition of CRM increased the viscosity at 135 °C and 180 °C, as expected. The conditioned CRM binders appeared to have higher viscosity in the bottom part, compared to the middle and top parts. This is caused by the rubber particles filling from the bottom part to the middle part while conditioning.
(2) From the DSR test at high temperatures, it was found that increasing CRM content made it possible to increase G*/sin δ in the original condition. Similar to the viscosity results, the CRM binders after conditioning showed the highest G*/sin δ value in the bottom part.
(3) From the MSCR test, J_{nr} and % rec values are observed to have a similar trend with G*/sin δ results. However, some of the data were not measured due to the higher load than the DSR test.
(4) The SI from G*/sin δ generally increased as the test temperature increased. The SI increased up to 10% CRM, and then decreased as the CRM content was further increased.
(5) It was observed that the SI from G*/sin δ generally used was suitable for evaluating the storage stability of the CRM asphalt binders, compared to the SI from % rec.
(6) The results are limited to the CRM particles and asphalt binders used in this study and are intended to show the change in storage stability according to CRM contents. To draw a more general conclusion, it is recommended to use different types of rubber powder and asphalt binders. Moreover, a study evaluating how the SI varies depending on the binder test method can be considered.

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References

1. Wang, D.; Falchetto, A.C.; Poulikakos, L.; Hofko, B.; Porot, L. RILEM TC 252-CMB report: Rheological modeling of asphalt binder under different short and long-term aging temperatures. *Mater. Struct.* **2019**, *52*, 73. [CrossRef]
2. Ren, Z.; Zhu, Y.; Wu, Q.; Zhu, M.; Guo, F.; Yu, H.; Yu, J. Enhanced Storage Stability of Different Polymer Modified Asphalt Binders through Nano-Montmorillonite Modification. *Nanomaterials* **2020**, *10*, 641. [CrossRef] [PubMed]
3. Yu, H.; Leng, Z.; Dong, Z.; Tan, Z.; Guo, F.; Yan, J. Workability and mechanical property characterization of asphalt rubber mixtures modified with various warm mix asphalt additives. *Constr. Build. Mater.* **2018**, *175*, 392–401. [CrossRef]
4. Akisetty, C.K.; Lee, S.-J.; Amirkhanian, S. High temperature properties of rubberized binders containing warm asphalt additives. *Constr. Build. Mater.* **2009**, *23*, 565–573. [CrossRef]
5. Yu, X.; Wang, Y.; Luo, Y. Effects of Types and Content of Warm-Mix Additives on CRMA. *J. Mater. Civ. Eng.* **2013**, *25*, 939–945. [CrossRef]
6. Wekumbura, C.; Stastna, J.; Zanzotto, L. Destruction and Recovery of Internal Structure in Polymer-Modified Asphalts. *J. Mater. Civ. Eng.* **2007**, *19*, 227–232. [CrossRef]
7. Ragab, M.; Abdelrahman, M. Enhancing the crumb rubber modified asphalt's storage stability through the control of its internal network structure. *Int. J. Pavement Res. Technol.* **2018**, *11*, 13–27. [CrossRef]
8. Pérez-Lepe, A.; Martínez-Boza, F.J.; Gallegos, C. High temperature stability of different polymer-modified bitumens: A rheological evaluation. *J. Appl. Polym. Sci.* **2006**, *103*, 1166–1174. [CrossRef]
9. Wang, H.; Liu, X.; Erkens, S.; Skarpas, A. Experimental characterization of storage stability of crumb rubber modified bitumen with warm-mix additives. *Constr. Build. Mater.* **2020**, *249*, 118840. [CrossRef]
10. Sienkiewicz, M.; Borzędowska-Labuda, K.; Wojtkiewicz, A.; Janik, H. Development of methods improving storage stability of bitumen modified with ground tire rubber: A review. *Fuel Process. Technol.* **2017**, *159*, 272–279. [CrossRef]
11. Hosseinnezhad, S.; Kabir, S.F.; Oldham, D.; Mousavi, M.; Fini, E.H. Surface functionalization of rubber particles to reduce phase separation in rubberized asphalt for sustainable construction. *J. Clean. Prod.* **2019**, *225*, 82–89. [CrossRef]
12. Xiao, F.; Yao, S.; Wang, J.; Wei, J.; Amirkhanian, S. Physical and chemical properties of plasma treated crumb rubbers and high temperature characteristics of their rubberised asphalt binders. *Road Mater. Pavement Des.* **2020**, *21*, 587–606. [CrossRef]
13. Salazar-Cruz, B.; Zapien-Castillo, S.; Hernández-Zamora, G.; Rivera-Armenta, J. Investigation of the performance of asphalt binder modified by sargassum. *Constr. Build. Mater.* **2021**, *271*, 121876. [CrossRef]
14. Wen, Y.; Liu, Q.; Chen, L.; Pei, J.; Zhang, J.; Li, R. Review and comparison of methods to assess the storage stability of terminal blend rubberized asphalt binders. *Constr. Build. Mater.* **2020**, *258*, 119586. [CrossRef]
15. Lee, S.-J.; Amirkhanian, S.; Shatanawi, K. Effects of Crumb Rubber on Aging of Asphalt Binders. *Asphalt Rubber* **2006**, *3*, 779–795.
16. Shen, J.; Amirkhanian, S.; Lee, S.-J.; Putman, B.J. Recycling of laboratory-prepared RAP mixtures containing crumb rubber modified binders in HMA. *Transp. Res. Rec.* **2006**, *1962*, 71–78. [CrossRef]
17. Kim, H.; Lee, S.-J. Laboratory Investigation of Different Standards of Phase Separation in Crumb Rubber Modified Asphalt Binders. *J. Mater. Civ. Eng.* **2013**, *25*, 1975–1978. [CrossRef]
18. Xie, J.; Yang, Y.; Lv, S.; Zhang, Y.; Zhu, X.; Zheng, C. Investigation on Rheological Properties and Storage Stability of Modified Asphalt Based on the Grafting Activation of Crumb Rubber. *Polymers* **2019**, *11*, 1563. [CrossRef] [PubMed]