CHARACTERIZATION OF RUBBERIZED BINDERS WITH WAX ADDITIVES

by

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DEDICATION

I wish to dedicate this dissertation to God, my parents, and my beautiful wife, In Hoe Lee.

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ABSTRACT

Crumb rubber is widely used as an environment-friendly material in asphalt industry to improve pavement performance properties. Warm mixture asphalt (WMA) refers to the technologies which allow a substantial reduction of mixing and compaction temperatures of asphalt mixtures through decreasing the viscosity of asphalt binders. In general, the crumb rubber modified (CRM) asphalt mixes require higher temperatures compared to conventional hot mix asphalt (HMA). However, if the technologies of WMA are incorporated, it is predicted to reduce the mixing and compaction temperatures of rubberized asphalt mixtures. For utilization of rubberized mixes in WMA industry, it is needed to investigate the properties of CRM binders with warm additives depending on CRM contents, additive types, aging states, and other experiment variables.

This research investigated CRM binders with wax warm additives, and the objectives included: 1) quantifying viscosity changes dependent upon mixing temperature and hauling period through Rotational viscometer test; 2) evaluating the effect of blending time on viscosity; 3) investigating the performance properties of recycled aged CRM binders containing wax additives through Superpave binder tests; 4) evaluating the microstructural properties using an environmental scanning electron microscope (ESEM) and an atomic force microscopy (AFM).

The following conclusions were drawn based on the comprehensive laboratory investigation: 1) crumb rubber contents and wax types were found to have significant

influence on the viscosity properties of CRM binders; 2) CRM binder with wax additive (LEADCAP) showed better rutting and cracking resistance compared to the control CRM binder for all aging states; 3) the wax additive in recycled CRM binder is observed to play an important role in binder properties even after subjecting to long-term aging processes; 4) the microstructures on morphology images seemed to have a good relation with the engineering properties of warm CRM binders.

I. INTRODUCTION

Problem Statement

Approximately 250 million scrap tires are generated each year in the United States (Rubber Manufacturers Association, 2014). The disposal of scrap tires has been a serious issue due to many reasons (e. g., lack of landfill space, air pollution, etc.). A method to recycle the scrap tires is addition into asphalt binder. The recycling of scrap tires with asphalt binder has been of interest to the domestic and international asphalt industry for over 50 years (Kim et al., 2014). Traditionally, scrap tires is ground up into crumb sized pieces to be incorporated into asphalt binders. The application of crumb rubber modifier (CRM) in asphalt binders has proven to be beneficial from many standpoints. Previous studies have concluded that rubberized binders could produce asphalt pavements which result in decreased traffic noise, reduced maintenance costs and improved resistance to rutting and cracking (Huang et al., 2002; Lee et al., 2007; Ruth and Roque, 1995; Shen et al., 2006, Kim et al., 2014). From these benefits, there is an increasing interest in using CRM binders in Hot Mix Asphalt (HMA) pavements in some states in the United States and other countries (Bahia and Davies, 1994; Lee et al., 2006).

Even though the addition of crumb rubber has a lot of advantages, CRM asphalt mixtures are compacted at a higher temperature than conventional mixtures, based on the field experience (Amirkhanian and Corley, 2004). With lower compaction temperatures, the use of rubberized mixture might result in several problems such as inadequate volumetric properties (i.e., high air voids) and poor short-term and long-term

performances. Also, the viscosity increase can negatively affect workability of asphalt mixtures as it requires the higher temperature to maintain the binder viscosity for the proper workability, and other environmental problems can be caused by the use of more fossil fuel.

Warm mix asphalt (WMA) refers to technologies which allow a significant reduction of mixing and compaction temperatures of asphalt mixture through decreasing the viscosity of asphalt binders. Reduced mix production and paving temperatures would decrease the energy needed to produce the HMA, reduce emissions and odors from plants, and make the working conditions better at the plant and paving site. Apart from these obvious benefits, there are other advantages of using warm asphalt, such as longer paving seasons, longer hauling distances, reduced wear and tear of the plants, reduced ageing of binders, reduced oxidative hardening of binders and ability of opening the site to traffic sooner, etc. (Akisetty et al., 2009; Button et al., 2007; D'Angelo et al., 2008; Gandhi and Amirkhanian, 2007; Hurley and Prowell, 2005; Hurley and Prowell, 2006; Kim et al., 2012).

According to Edwards et al., (2006), there is an obvious risk when using wax additives in cold climatic conditions. Although wax crystallization improves rutting resistance, other asphalt properties such as susceptibility to low temperature cracking, resistance to fatigue and adhesion properties may be affected in a negative way. However, the addition of CRM into the asphalt binder increases the asphalt pavement resistance to cracking in low temperature. If the wax additives and the CRM are used together, it is

expected that the general performances of asphalt pavement will be improved such as plastic deformation and cracking.

Research Objectives

The primary objective of this research is to investigate the properties of CRM binders containing wax additives. The specific objectives of this study included:

- Quantifying viscosity changes dependent upon mixing temperature and hauling period through Rotational viscometer test,
- Evaluating the effect of blending time on viscosity of CRM binders containing wax additives,
- Evaluating the influence of rubber contents on cracking resistance of warm rubberized binders,
- Investigating the performance properties of recycled aged CRM binder containing wax additives through Superpave binder tests,
- Evaluating the microstructural property of CRM binder with wax additives using an environmental scanning electron microscope (ESEM) and an atomic force microscopy (AFM).

Scope of Research

The objectives of this study are accomplished through the completion of the tasks described below:

- Evaluating the viscosity property (blending time: 1, 30, 60, and 90 minutes; CRM percentage: 0, 10, 15, and 20% by binder weight) in the laboratory using
    - A rotational viscometer.
- Evaluating the rheological property (CRM percentage: 0, 5, 10, and 15% by binder weight) in the laboratory using
    - A dynamic shear rheometer (DSR), and
    - A Bending beam rheometer (BBR).
- Evaluating the properties of recycled CRM binders with wax additives (recycling content: 0 and 15% by binder weight) using the following treatments and tests
    - Artificial aging of CRM binders by using rolling thin film oven (RTFO) and pressure aging vessel (PAV) methods.
    - Measuring the properties of the recycled CRM binders using a rotational viscometer, DSR, and BBR.
- Evaluating the microstructural properties of CRM binders with wax additives using the following treatments and tests
    - Artificial aging of CRM binders by using RTFO and PAV methods.

- Investigating the microstructural properties of the CRM binders using ESEM and AFM.

Dissertation Organization

This dissertation consists of eight chapters. Chapter I presents the background, objective and scope of the study. Chapter II is a literature review of previous research in the crumb rubber modified (CRM) asphalt, the warm mix asphalt (WMA), the recycling of rubberized asphalt, and the microstructural characterization of asphalt binder. Chapter III summaries the statistical methods used for analyzing data. Chapter IV contains the viscosity properties depending on CRM contents and blending time. Chapter V describes the rheological properties through Superpave binder tests. Chapter VI reports the recycling properties. Chapter VII presents the microstructural characterization of CRM binder with wax additives. Finally, Chapter VIII includes a summary of the investigation, conclusions, and provides recommendations for future research.

II. LITERATURE REVIEW

Crumb Rubber Modified Asphalt

Every year huge amount of scrap tires has been produced in United States. It is considered as a reason of serious environmental problems such as fire dangers and provide breeding grounds for rodents, snakes, mosquitoes and other pests, causing health hazard and environmental problems (Snyder, R.H., 1998; Clark et al., 1993; Jang et al., 1998). The addition of scrap tire into asphalt binder came into the spotlight in asphalt industry several decades ago. It was reported that the asphalt industry can absorb up to 40% of scrap tires (Isayev, 2005).

Types of crumb rubber modified (CRM) asphalt are simply divided by their production process, the wet process and the dry process. In the wet process, fine CRM is blended with the asphalt binder before mixing with aggregate. In the dry process, coarse CRM is used to replace aggregate in the asphalt mixture. It is concluded that the wet process is more efficient in improving the properties of an asphalt mixture (Takallou et al., 1991). Crumb rubber is known to absorb liquids and swell, depending on the temperature and viscosity of the liquids it is absorbing (Gawel et al., 2006). Interaction of the rubber particles with the asphalt binder can be affected by several factors such as temperature, mix type, rubber size and texture, and chemical composition of the asphalt binder (Leite et al., 2003). Scrap tires used for asphalt binder improve the pavement performance and safety by being a cost effective modifier for the highway pavement industry (Amirkhanian, 2003).

Many researchers have performed the use of scrap tires in asphalt industry. The first application of scrap tires with HMA was seen in an open-graded friction course (OGFC) in 1975 (Arizona Department of Transportation, 1989). The applications of CRM into asphalt binder have been increased and incorporated by several states. One of the reasons for this utilization of CRM was included in order to meet the standard suggested by the Intermodal Surface Transportation Efficiency Act (ISTEA) in 1991. The U.S. government mandated the utilization of scrap tire rubber in asphalt industry.

Other studies concluded that CRM binders could produce asphalt pavements with several advantages related to pavement performance, including increased resistance to rutting and cracking, decreased traffic noise, and reduced maintenance costs (Huang et al., 2002; Lee et al., 2007; Liang and Lee, 1996; Ruth and Roque, 1995; Shen et al., 2005). Due to these benefits, the interest of using CRM binders in HMA pavements is increasing in the United States and other countries (Bahia and Davies, 1994; Lee et al., 2006).

Although the use of crumb rubber in modified asphalt is shown good performance, there are several concerns that must be considered. Increase of binder viscosity is considered as one of the primary problems of CRM binder. It was reported that the addition of crumb rubber can significantly increase the binder viscosity, and the temperature needs to be increased with the rise of CRM content to meet the 3 Pa requirement in Superpave specification (Wang et al, 2012). Therefore, rubberized asphalt mixes require higher mixing and compacting temperatures (Amirkhanian and Corley 2004), hence generating potential problems to the environment due to emissions. Because of these issues obtaining the permission for asphalt plant in the city becomes a very difficult task in many parts of

the country. In addition, the increase of rubber content causes a decrease in the value of resilient modulus of mixtures (Xiao et al. 2007).

Warm Mix Asphalt

In general, asphalt mixtures can be divided into three categories, hot mix asphalt (HMA), warm mix asphalt (WMA) and cold mix asphalt (CMA). The CMAs are more beneficial for environmental issues because of low production temperature. However, CMA properties are not as good as the HMA properties because of the poor performances for coating the aggregate and the presence of water in the mix. Due to the curing time to open up to traffic, the CMAs are not used for a higher traffic volume road (Kiran, 2008). On the other hand, WMA and HMA are usually being applied for the higher volume traffic. WMA technology was proposed through a study conducted by Kolo Veidekke to overcome the problems of CMA mixture. The study suggested a specific asphalt mix at somewhat lower temperature than conventional HMA that is called warm asphalt (Koenders et al., 2000). The major difference between WMA and HMA is their production temperature. The production temperature of WMA is in the range of 100-140˚C, whereas for HMA it ranges between 150-170˚C (AAPA, 2001). The WMA production and lay down temperatures are 20 to 30˚C below compared to HMA. If the temperature of the mix at the plant is less than 100˚C, the mix is known as half warm mix, which is in between cold mix and warm mix (Prowell, 2007).

A team of 13 materials experts from the United States visited four European countries (Belgium, France, Germany, and Norway) in May 2007 to assess and evaluate

various WMA technologies. The team learned about a wide variety of technologies through discussion with various agencies, visiting construction sites, and viewed in-service pavements. The development of WMA technologies in Europe identified several kinds of factors affecting the production of WMA. Performances were presented based on the data obtained from three countries considering the laboratory and short-term (3 years or less) field performance records. Generally, WMA mixes appear to provide the same performance as or better performance than HMA. Even though poor performance was observed on limited sections in Norway, it is considered that the poor performance was not directly attributed to WMA use. (D'Angelo et al., 2008).

The main advantage of warm asphalt is the reduction of temperature during production and placement, producing less fumes, less emissions at the plant, less energy consumption, less wear and tear on the plant and less aging of the binder (Hurley and Prowell, 2005; Gandhi and Amirkhanian, 2007). Since WMA was introduced in 2000, it attracted considerable attention of the highway engineering community because of its advantages over both HMA and CMA (NCAT, 2005). The application of WMA in field construction appeared in the mid-1990s (The use of Warm Mix Asphalt., 2010). Thus, it is considered that unknown territories related to the long-term mechanical performance of WMA mixtures are still exist (Kristjánsdóttir, 2006). WMA is expected to show better or similar performances compared to HMA. If the engineering properties of WMA cannot perform as HMA, those advantages will be useless, even though WMA has several advantages mentioned previously (e.g. less fumes and less energy consumption). Though research findings have confirmed that WMA performs similar to HMA so far (Zaumanis

and Haritonovs, 2010; Chowdhury and Button, 2008; D'Angelo et al., 2008), the researches including long-term aging still need to be studied in greater depth. This topic is expected to show significant differences between HMA and WMA (including all WMA technologies) (Rubio et al., 2012).

Reclaimed Asphalt Pavement

Several researches were conducted using Reclaimed Asphalt Pavement (RAP) considering numerous influences such as engineering property and environmental issue. Although several features affect the use of RAP in asphalt pavement, the two primary factors are economic savings and environmental benefits. RAP is useful to save virgin materials because it can replace the use of virgin aggregate and the amount of virgin asphalt binder required in the production of HMA. In addition, the use of RAP conserves energy, lowers transportation costs required to obtain quality virgin aggregate, and preserves resources. Using RAP is beneficial for environment in terms of decreasing the amount of construction debris placed into landfills and saving nonrenewable natural resources such as virgin aggregate and asphalt binder. Ultimately, recycling asphalt creates a cycle that optimizes the use of natural resources and sustains the asphalt pavement industry (Copeland, 2011).

According to the previous studies (Decker 1997; Gardiner and Wagner 1999; Kandhal 1997; Kearney 1997; Terrel 1997; Xiao 2006; Lee 2007), the use of recycled materials has proven to be equal or even better than new materials in quality. Over years, recycling has become one of the most attractive pavement rehabilitation alternatives, and

different recycling methods are now available to address specific pavement distress and structural needs. The NCHRP 9-12 report has developed guidelines for incorporating RAP in the Superpave mix design procedure and prepared a manual for RAP usage (McDaniel et al. 2002; NCHRP 2001) The use of RAP in the past has proved to be economical, environmentally sound, and effective in increasing the rutting resistance of asphalt mixtures. Therefore, a number of states throughout the United States have established a guideline to consume RAP, and the satisfactory mix design procedures have been developed.

Federal Highway Administration (FHWA) published a practice report of RAP conducted through the Long-Term Pavement Performance (LTPP) program (Copeland, 2011). The research was performed focusing on high content of RAP up to 30%. In this research, it is estimated that the average consumption of RAP in the United States is approximately 12 percent. However, according to State transportation department specifications, the overall amount of RAP used in pavement construction can be increased up to 30%. It was concluded that the asphalt pavement containing 30% RAP showed similar performance with virgin pavement without RAP based on a survey of LTPP located throughout the United States and Canada. On the other hand, characterizing RAP material is important to achieve proper mix design purpose. The laboratory mixture design should be established using RAP as a component. This is especially important for State transportation departments considering permitting up to 20% RAP in mixtures without changing a softer grade asphalt binder. When applying high RAP contents greater than 25

percent, it is required to cautiously select the grade of asphalt binder added to the recycled asphalt mixture following State transportation department specifications.

High content of RAP negatively affect the long term durability and workability of asphalt pavement, and the use of some WMA technologies may increase the moisture susceptibility. Therefore, the combination of RAP and WMA technologies can complement both problems that could occur at individual application. Recently, there are some researches to incorporate the RAP and WMA technology. The Maryland State Highway Administration found that the stiffness values of the WMA and HMA control mixtures are statistically similar using an asphalt pavement section of road containing 45% RAP in the base course, Stone Mastic Asphalt (SMA) in the intermediate course, and 35% RAP in the surface course with 1.5% Sasobit by weight of total binder as a modifier (Michael, 2005).

Copeland et al. (2010) in association with the Florida DOT performed a case study aimed to evaluate field performance of WMA and HMA mixtures with high RAP. Asphalt mixture including 45% RAP with WMA technology using water injection method was compared with regularly recycled mixture (45% RAP) without WMA. They evaluated the performance grade of the binders, dynamic modulus, and flow number values in the study. They found consistent results that the application of WMA in high content of RAP made it softer than the high RAP with HMA control mixture.

Lee et al. (2009) conducted test with aged asphalt binder with WMA additives, Sasobit and Aspha-min, on binder properties of asphalt binder including 15% RAP. The research found that the addition of Sasobit reduced the viscosity of the binder while the

addition of Aspha-min showed the opposite effect. The result of Dynamic Shear Rheometer (DSR) test at high temperature showed that WMA additives increase rutting resistance for the same virgin binder grade. However, it is concluded that the addition of WMA additives has negative effects on the cracking resistance based on the data obtained from DSR test at intermediate temperature and Bending Beam Rheometer (BBR) test at low temperature.

According to an experimental study conducted by Tayebali et al. (2015), it was found that workability and Tensile Strength Ratio (TSR) was observed to decrease with increase in RAP content for similar mixtures while the Indirect Tensile Strength (ITS) values increased. In addition, in the case of 40% RAP content, WMA mixtures with standard binder grade performed better than corresponding HMA-RAP mixtures.

Xiao et al. (2016) conducted experiment for the evaluation of higher RAP contents with WMA technologies. Total 36 Superpave mix designs were analyzed using one PG 58-28 binder, two PG 64-22 binders, two RAP sources, and three mix technologies (e. g. HMA, foaming technology, and Evotherm additive) to explore the volumetric characteristics of various mixtures. According to the study, the results indicated that there is no significant change of optimum asphalt content (OAC) by increasing the RAP content. Additionally, a higher percentage of RAP (i.e., over 40%) caused no noticeable increases at the mixing and compaction temperatures for these mixtures. The voids in mineral aggregate (VMA) value in WMA mixture showed specific trend that the VMA reduces as the RAP percentage increases regardless of binder source. Thus, the WMA technology and RAP concentration paly significant role in determining the voids in VMA of WMA mixture.

Microstructural Characterization of Asphalt Binder

Alternatively, application of modern technology has allowed to examine asphalt at shorter length scales using atomic force microscopy (AFM), which has revealed heterogeneous domains in asphalt (Loeber et al., 1996; Jager et al., 2004; Masson et al., 2006, 2007; Pauli et al., 2001; Allen et al., 2012) with potentially different mechanical properties.

First microstructural investigation of asphalt binder using AFM was conducted by Loeber et al. (1996). They used AFM for characterizing the natural surface of the asphalt binder without any disturbance. The research found that specifically rippled features on the microstructure which is called "bee" structure. This bee shape is considered as the change in topography on the asphalt binder surface. They described that the bee structure is related to the resin and asphaltene molecules. Therefore, there have been extensive researches on bee structure since Loeber et al. reported the study at 1996.

According to the research conducted by Pauli et al. (2001), the bee structure is deeply associated with asphaltene content. They observed big bee structure on selected binders containing large amount of asphaltene fraction (approximately 20% by weight), while relatively smaller bee structures were observed on asphalt binder surface with only 5% asphaltene content. However, some asphalt binders containing approximately 16% asphaltene showed no bee structure. They explained that this phenomenon is due to the low viscosity of asphalt binder.

Jäger et al. (2004) performed test to identify the chemical composition of the bee structure through ASTM D 3279 using n-heptane which is a completely non-polar solvent.

The addition of n-heptane precipitates separating the asphaltenes and maltenes, and then the n-heptane and maltenes were removed by distillation. After the distillation, the residue was dried under a vacuum to prepare AFM samples. They found that the maltene phase doesn't exhibits bee structure while the asphalt binder with high polarity asphaltenes showed significant bee structures. Based on these results, it is concluded that the bee structure on binder surface is related to the presence of asphaltenes.

There are three types of wax present in the asphalt binder. The first one is macrocrystalline or paraffin wax, which crystallize in flat, large plates or needles and have n-alkanes with no side branches (Dorset, 2005). The second one is microcrystalline wax which consists of aliphatic hydrocarbons with iso- and cyclo-parafins and normally crystalizes into tiny microscopic needles (Srivastava et al., 1993). The last one contains aromatics and molecules with polar functional groups that crystallize with cooling (Carbognani, 2000). De Moraes et al. (2010) conducted an experiment to investigate the reason behind the formation of bee structure with 30/45 asphalt binder obtained from a blend of Arabian Light deasphalting residue with aromatic extract of Bright Stock. They have used a temperature-controlled AFM and differential scanning calorimetry (DSC) testing, and concluded that the formation of bees appeared in the asphalt binder due to paraffin wax. Das et al. (2013) also did a similar study using the heating affects to see the change in the microstructure properties of asphalt binder. The authors concluded that the wax fraction in the asphalt binder has a strong relation with the formation of bee structures in the asphalt binder. Later two other studies performed thermal investigation to know the

formation of bee structures and both studies concluded that bee structures are wax-induced (Soenen et al., 2013; Schmets et al., 2010).

III. STATISTICAL ANALYSIS METHOD

A statistical analysis was performed using the Statistical Analysis System (SAS) program and Microsoft Excel to conduct an analysis of variance (ANOVA) and Fisher's Least Significant Difference (LSD) comparison with an $\alpha = 0.05$. The statistical design was based on conducting rotational viscosity tests, dynamic shear rheometer (DSR), and bending beam rheometer (BBR) test of control and CRM binders containing wax additives.

The effects of several rubber contents and blending times on the viscosity of asphalt binders in assessed using an ANOVA technique in the SAS program. The wax types, rubber contents, and blending time were considered as treatments in the analysis to evaluate viscosity property by CRM contents and blending time. The ANOVA was performed first to determine whether significant differences among the sample means existed. In the analyses of this study, the significance level was .95 ($\alpha = 0.05$), indicating that each finding had a 95% chance of being true. The calculations for ANOVA analysis were performed using the general linear model (GLM) in SAS. The ANOVA table is shown in Table 3.1. The hypotheses for these tests are as follows:

H_0: Mean viscosity values for all rubber contents (or blending times) are equal

(i.e., $\mu_1 = \mu_2 = \mu_3 = \ldots = \mu_a$)

H_1: at least one $\mu_i \neq \mu_j$

If H_0 is rejected at the 5% confidence level, the LSD test is used to identify which treatments are different. Upon determining that there were differences among sample means using the ANOVA, the LSD was then calculated. The LSD is defined as the observed differences between two sample means necessary to declare the corresponding

population means difference. Once the LSD was calculated, all pairs of sample means were compared. If the difference between two sample means was greater than or equal to the LSD, the population means were declared to be statistically different (Ott 2001).

Table 1. Analysis of Variance (ANOVA) Table

Source	*Sum of squares*	*Degrees of freedom*	*Mean square*	F_0
Treatments	$SS_{Treatments}$	a-1	$SS_{Treatments}/(a-1)$	MST/MSE
Error	SS_E	N-a	$SS_E/(N-a)$	
Total	SS_T	N-1		

Where,

$SS_{Treatments}$=treatment sum of squares in between

SS_E=sum of squares for error

SS_T=sum of squares for total

The primary variables included the wax types and rubber contents for cracking property of CRM binders. To evaluate the recycling properties, warm asphalt additives were considered as primary treatments and long term aged (LTA) binder percentages were considered as secondary treatments.

Correlation analyses were conducted to determine the relationship between the stiffness and roughness measured by the atomic force microscopy (AFM) at original and long-term aged (RTFO+PAV residual) states.

IV. VISCOSITY PROPERTIES

Introduction

Viscosity is an important property of asphalt binders that directly relates to pavement quality. The binder viscosity is sensitively controlled by their managing temperature. In addition, the rubberized binder and warm additive significantly affect the temperature of the asphalt binder. Therefore, the deep investigation of binder viscosity is an essential task in this study.

There are two ways to produce CRM binders, the wet process and the dry process, the wet process is generally used because of easier management for binder quality. In addition, the wet process is known to be more efficient in improving properties of an asphalt mixture (Takallou and Takallou, 1991). In the wet process, the viscosity of CRM binder increases rapidly with mixing time for roughly the first 80–90 min of the measurements. Subsequently, a gradual increase in viscosity is observed to be linear with time (Shuler et al., 1986; Roberts et al., 1989). The gradual viscosity increase can be explained due to the amount of aromatic oil absorption and rubber particle swelling. It was reported that as the percentage of rubber increased the effects the rubber had on the viscosity increased significantly (Lougheed and Papagiannakis, 1996).

The gradual viscosity increase of rubberized binder has negative effects on workability of asphalt mixtures. Since rubberized binder requires higher mixing and compaction temperature to maintain the binder viscosity for the proper workability, other problems can occur due to the use of more fossil fuel. These problems are main obstacles

to impede the use of crumb rubber in asphalt binder. It may be considered that the use of wax warm additives can solve the problems caused from the viscosity increase of asphalt binder.

The blending time of CRM binder is required to interact the crumb rubber with an asphalt binder. It depends on many factors including the chemistry of the asphalt binder and the crumb rubber as well as the particle size and texture of the crumb rubber (Jeong et al., 2010). The binder interaction with crumb rubber is a complex chemical action which is not clearly identified yet. Therefore, the blending time for CRM binder is one of the important variables that can directly affect to the binder property. In addition, insufficient blending time may be considered as one of the main reasons for the gradual viscosity increase of rubberized binder. Moreover, the addition of wax additive into rubberized binder can be a potential solution to reduce the binder viscosity.

In this chapter, the viscosity changes of CRM binders containing wax additive are evaluated through rotational viscometer (RV) test using different testing temperatures (135°C and 120°C) and periods (30, 120, and 240 min) for the analysis of viscosity properties with different CRM contents (0, 10, 15, and 20%) and blending time (1, 30, 60, and 90). Figure 1 shows a flow chart of the experimental design used in this viscosity study.

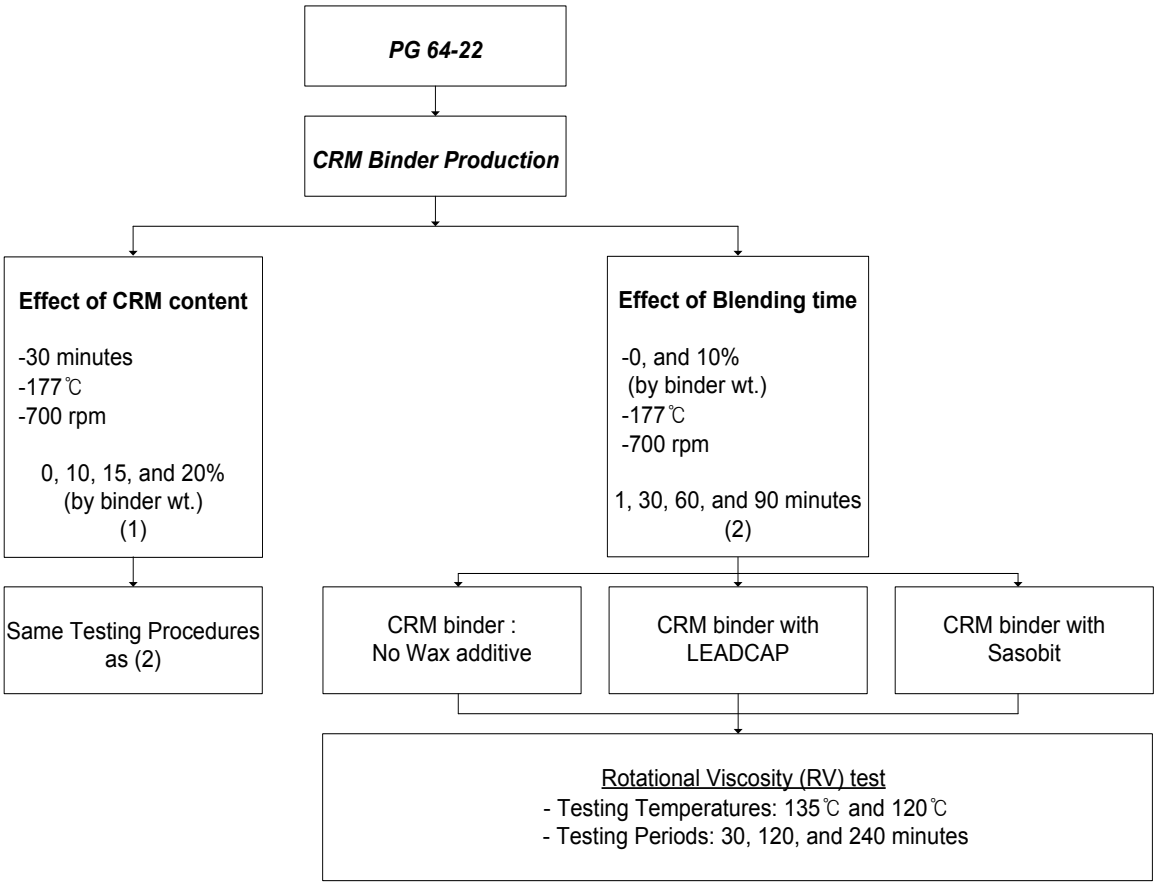


Figure 1. Flow char t of experimental design.

Experimental Program

Materials

Asphalt binder

Performance grade (PG) 64-22 asphalt binder without additives was used as a base binder for this study. The binder properties are represented in Table 2.

Crumb rubber modifier (CRM)

The crumb rubber included in this study was obtained from one source. This source used the mechanical shredding method to process scrap passenger tires into crumb rubber. Table 3 shows a gradation of crumb rubber.

Wax additives

Two commercial wax additives of LEADCAP and Sasobit were selected to add into the rubberized binder. The process included the addition of additive at a specified concentration (1.5% by the weight of the rubberized binder) followed by hand mixing for a minute to achieve a consistent mixing. The concentration of 1.5% was suggested by the manufacturer. The LEADCAP is classified as an organic WMA additive, which is a wax-based composition including crystal controller to adjust crystalline degree of wax material at the low temperature and adhesion promoter to enhance adhesion between asphalt and aggregate (Kim et al. 2013). Sasobit is a product of Sasol Wax. It is a long chain aliphatic hydrocarbon obtained from coal gasification using the Fischer-Tropsch process. After crystallization, it forms a lattice structure in the binder which is the basis of the structure

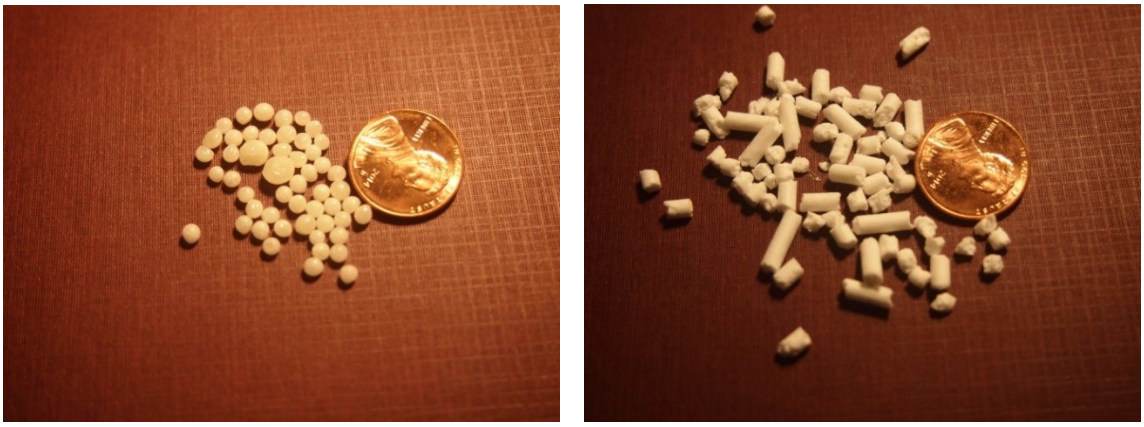
stability of the binder containing Sasobit (Sasol wax). These two wax additives were shown in Figure 2.

Table 2. Properties of base asphalt binder (PG 64-22).

Aging states	Test properties	Test result
Unaged binder	Viscosity @ 135°C (cP)	531
	G*/sin δ @ 64°C (kPa)	1.42
RTFO aged residual	G*/sin δ @ 64°C (kPa)	2.53
RTFO+PAV aged residual	G*sin δ @ 25°C (kPa)	2558
	Stiffness @ -12°C (MPa)	287
	m-value @ -12°C	0.31

Table 3. Gradation of crumb rubber used in this study.

Sieve Number (μm)	% Cumulative Passed
30 (600)	100.0
40 (425)	91.0
50 (300)	59.1
80 (180)	26.2
100 (150)	18.3
200 (75)	0.0



(a) (b)

Figure 2. Wax additives; (a) Sasobit and (b) LEADCAP.

CRM binder

There are two processes to produce the CRM binder, wet process and dry process. The CRM is added into the binder before the mixing with the aggregate in wet process. On the other hand, the CRM is added during the mixing process of the binder and the aggregate in dry process. In general, the wet process is used for laboratory experiment. The reason is that the wet process is easier to manage the binder quality than the dry process. Therefore, the binder mixing used in this study was the wet process. In the wet process, the CRM is added to the base asphalt binder before introducing the binder in the asphalt concrete matrix. The CRM binder was produced in the laboratory at 177 °C for 30 minutes by an open blade mixer at a blending speed of 700 rpm. Figure 3 shows the setup used to manufacture the CRM binder in this study.

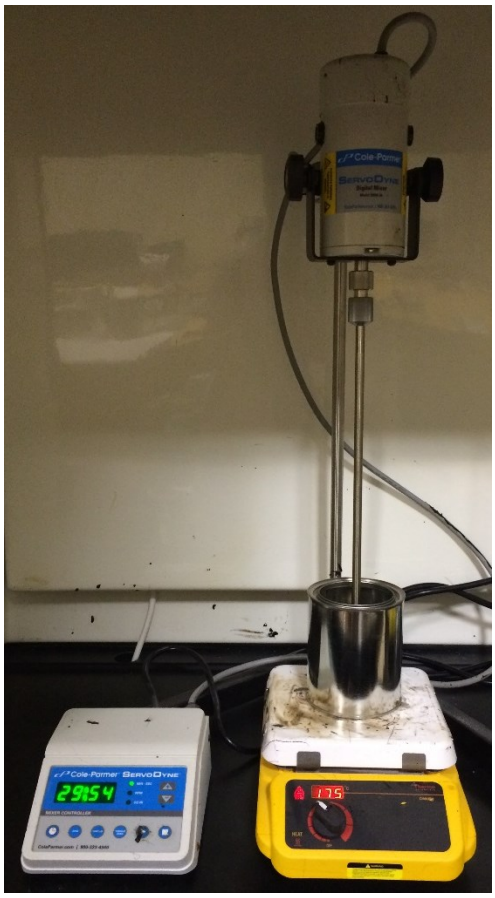


Figure 3. Production of CRM binder.

Rotational viscosity test

The RV test was conducted to evaluate the effect of crumb rubber on the rubberized binders containing wax additives. A Brookfield rotational viscometer was used to measure the viscosity at 135°C (the standard test temperature) and at 120°C (the mixing temperature generally used for warm mix asphalt). Three testing periods (i.e., 30, 120, and 240 minutes) were applied to evaluate the viscosity change by different testing time considerations. An 8.5g sample of the control binders and a 10.5g sample of rubberized binders were tested with a number 21 spindle and with a number 27 spindle in the viscometer, respectively. Three duplicate samples were tested and the results were reported as the average. For all

binders, each specimen was poured just prior to testing. Figure 4 shows a set of rotational viscometer (RV).

Figure 4. Rotational viscometer.

Results and Discussions

Effect of CRM contents

Viscosity

The viscosity of asphalt binders at high temperature is considered to be an important property to decide working temperature because it represents binder's ability to be pumped through an asphalt plant, thoroughly coat aggregate in asphalt concrete mix, and be placed and compacted to form a new pavement surface (Asphalt Institute 2003). Figure 5 shows the standard RV test results for all binders used in this study. The viscosity of 20% CRM binder could not be measured for specific value at 120°C, because the viscosity exceeded a measurable range. It is clear that the addition of crumb rubber into the asphalt binder greatly increases the binder viscosity, which may negatively affect mixing with aggregate in the hot mix asphalt. This potential problem can be solved by addition of wax additive. The viscosity of PG64-22+L and PG64-22+S measured at 135°C was 480 cP and 459 cP, respectively. These values are 90% and 86% of control binder viscosity, respectively. Two wax additives were observed to be effective to reduce the viscosity at both testing temperatures compared to the control binders, as expected. This effect is same for the tests done with all binders containing wax additives. The result indicates that the addition of wax additive results in reducing the mixing and compaction temperatures for the control mixes. In addition, the effect of wax additive in terms of reducing the binder viscosity was evidently showed in the rubberized binder. This phenomenon can be obviously observed in the result at 120°C. The viscosity reduction rate of rubberized binder at 120°C is more than 15%, which is higher than approximately 10% of the PG 64-22 binders containing

wax additives, suggesting that two wax additives into the rubberized binder are more efficient to reduce the viscosity at 120°C. The viscosity values of 15% rubberized binders at 120°C do not meet the current requirement set forth by Superpave (i.e., 3,000 cP). The viscosity data of 20% rubberized binders at 120°C shows immeasurable values, indicating that the mixing temperature needs to be increased for the practical use of 20% rubberized binder.

The statistical significance of the change in the viscosity as a function of binder type and wax additive was examined and the results are shown in Table 4. In general, the data indicated that the CRM content has a significant effect on the viscosity of the warm CRM binders. WMA binders containing wax additives (LEADCAP or Sasobit) were statistically insignificant with the control binders within each CRM content (0, 10, and 15%) at 135°C. Also, it is evident that the viscosity differences between the binders containing LEADCAP and the binders containing Sasobit at both testing temperatures were statistically insignificant for all CRM contents used in this study.

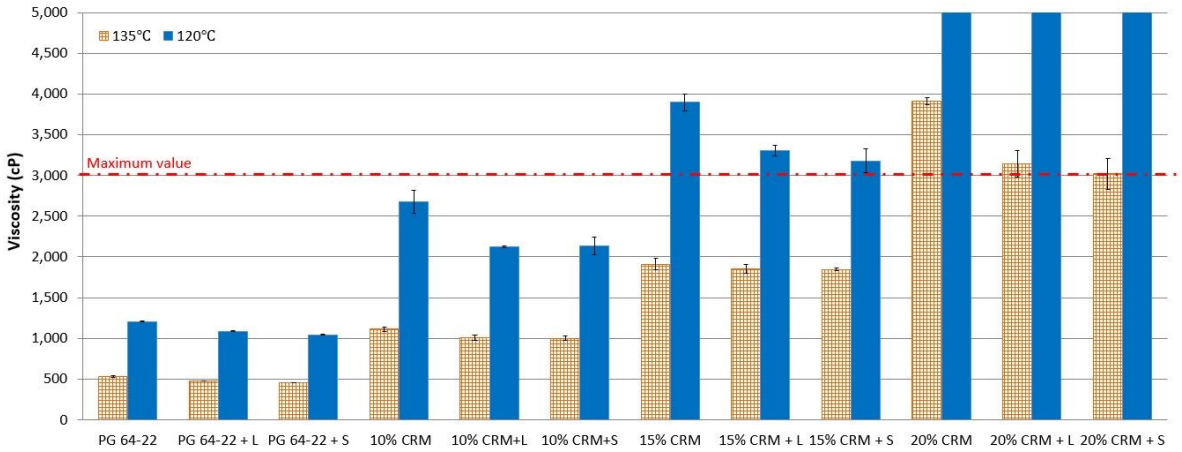


Figure 5. Viscosity of control and CRM binders with wax additive.

Table 4. Statistical analysis results of the viscosity value as a function of binder type and wax additive; (a) 135°C and (b) 120°C

(a)

Viscosity		0% CRM			10% CRM			15% CRM			20% CRM		
		1*	2	3	1	2	3	1	2	3	1	2	3
0% CRM	1	-	N	N	S	S	S	S	S	S	S	S	S
	2		-	N	S	S	S	S	S	S	S	S	S
	3			-	S	S	S	S	S	S	S	S	S
10% CRM	1				-	N	N	S	S	S	S	S	S
	2					-	N	S	S	S	S	S	S
	3						-	S	S	S	S	S	S
15% CRM	1							-	N	N	S	S	S
	2								-	N	S	S	S
	3									-	S	S	S
20% CRM	1										-	S	S
	2											-	N
	3												-

(b)

Viscosity		0% CRM			10% CRM			15% CRM		
		1	2	3	1	2	3	1	2	3
0% CRM	1	-	N	S	S	S	S	S	S	S
	2		-	N	S	S	S	S	S	S
	3			-	S	S	S	S	S	S
10% CRM	1				-	S	S	S	S	S
	2					-	N	S	S	S
	3						-	S	S	S
15% CRM	1							-	S	S
	2								-	N
	3									-

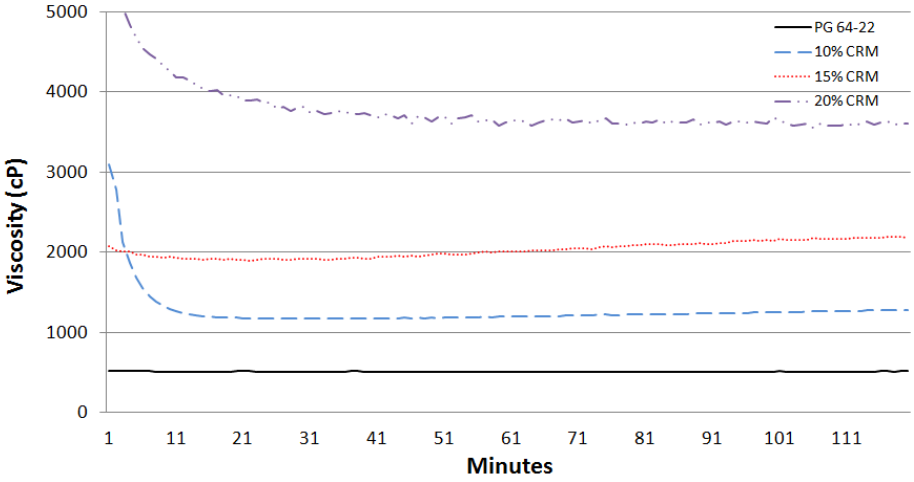
* CRM binder containing wax additive 1: CRM binder (Control)
2: CRM binder + 1.5% LEADCAP
3: CRM binder + 1.5% Sasobit

Rotational viscosity as a function of time at 135°C

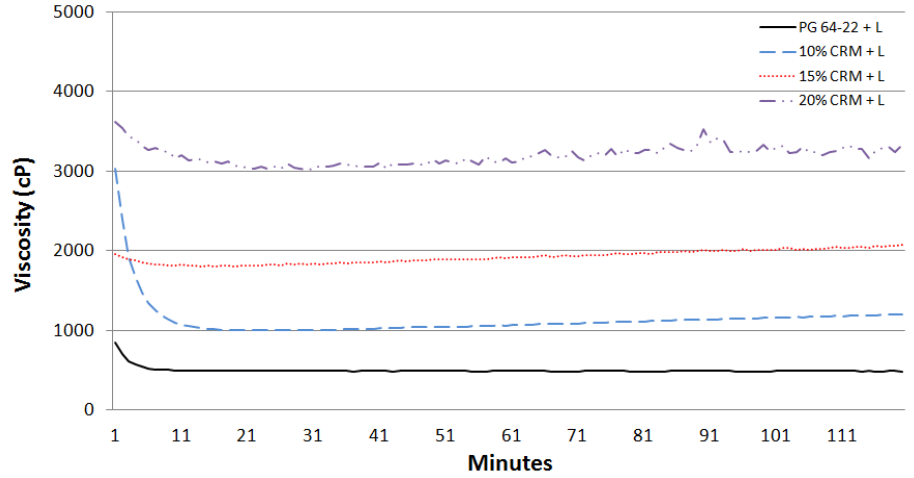
Although the addition of warm additives resulted in a conspicuous decrease of viscosity, the previous study mentioned the gradual increase of rubberized binder viscosity over time (Lougheed and Papagiannakis, 1996). Viscosity change of all binders was measured during 120 minutes and 240 minutes. Figure 6 describes the time versus viscosity curve for 120 minutes at 135°C. As expected, the CRM binders show much higher viscosity values than the PG 64-22 based binders for all testing periods. According to the figures, the viscosity values are stabilized between 20 and 40 minutes. This trend is exhibited in all tests done with PG 64-22 binders. The CRM binders are observed to have a different viscosity change. Generally, asphalt binders containing CRM showed a gradual increase in viscosity after approximately 30 minutes. The viscosity increase of CRM binders tends to be gradual and near linear.

One of benefits of WMA binder is a longer hauling distance and period. The haul management of asphalt mixture usually depends on the binder viscosity. The viscosity test was performed for 240 minutes to evaluate the longer haul management of rubberized binder. This is demonstrated in Figure 7, which depicts the time versus viscosity curve for 240 minutes at 135°C. Similar to the result of viscosity change at 120°C, the PG 64-22 based binders were found to have little viscosity change for the whole testing period. On the other hand, the viscosity of CRM binders increased approximately after 30 to 40 minutes. Although the CRM binders showed relatively higher viscosity and gradual viscosity increase at 135°C, the maximum viscosity values satisfied the current Superpave

requirement (i.e., Maximum 3,000 cP), with the exception of the 20% CRM binder. In general, the higher the rubber percentage, the steeper the viscosity changes over time.



(a)



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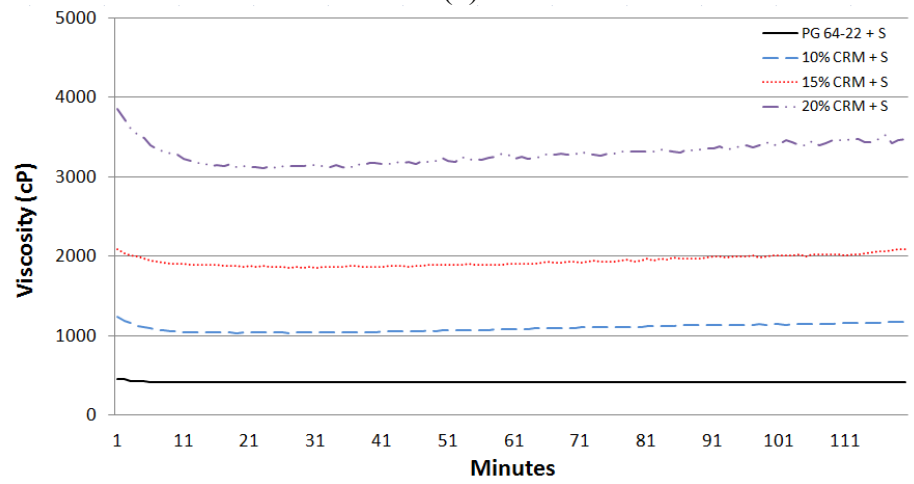
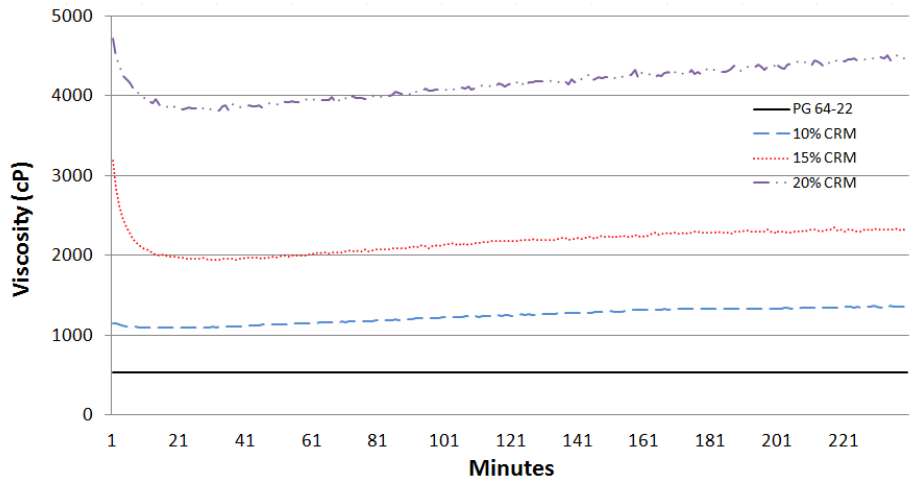
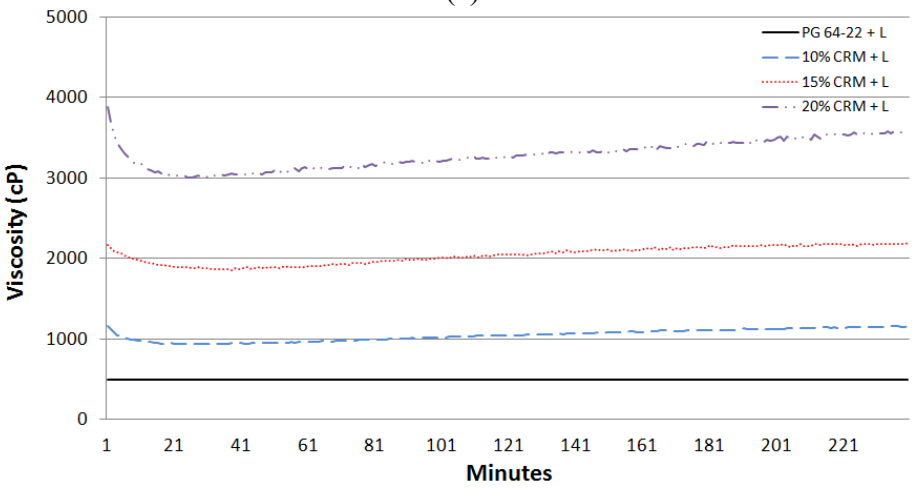


(c)

Figure 6. Viscosity change during 120 minutes at 135°C;
(a) Control binder, (b) WMA with LEADCAP, and (c) WMA with Sasobit.



(a)



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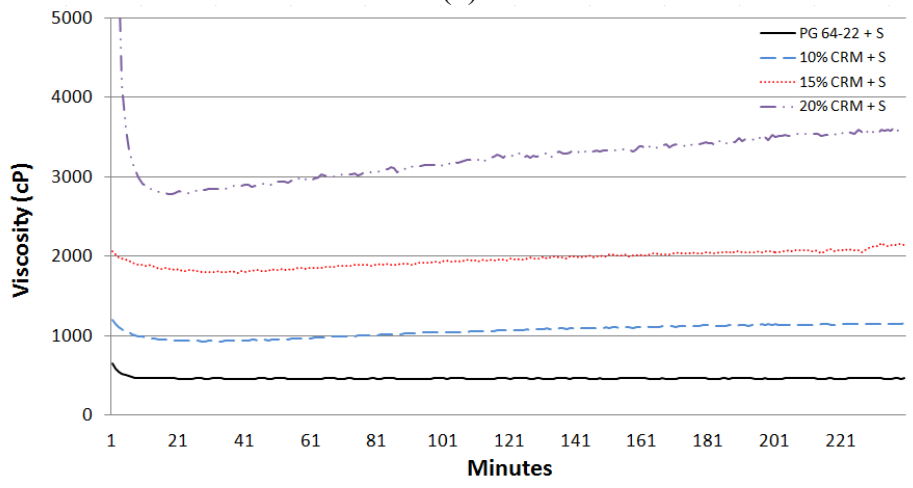


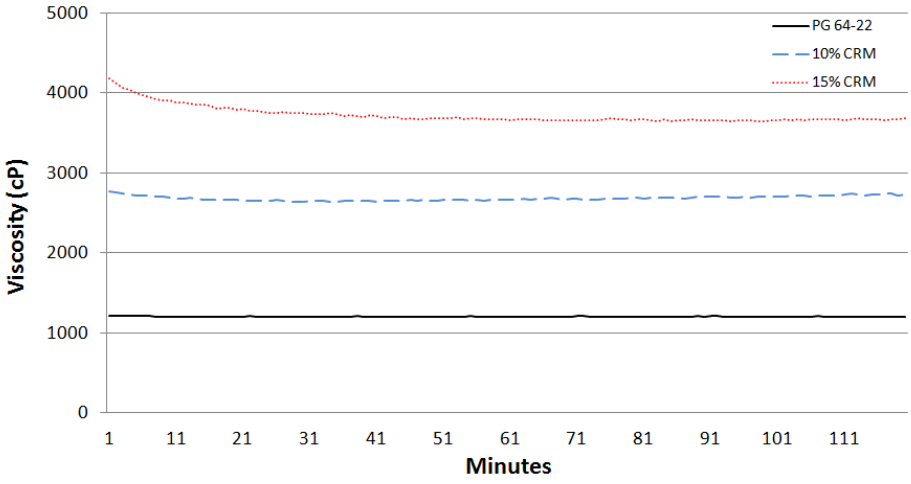
(c)

Figure 7. Viscosity change during 240 minutes at 135°C;
(a) Control binder, (b) WMA with LEADCAP, and (c) WMA with Sasobit.

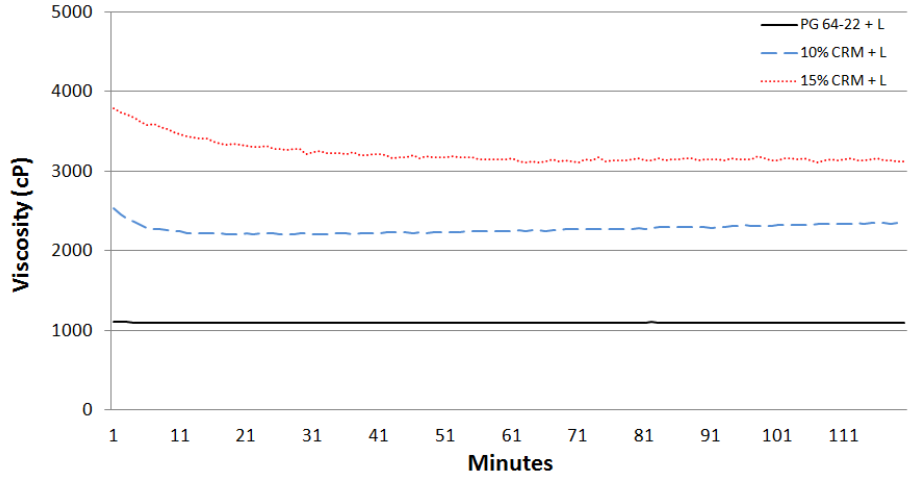
Rotational viscosity as a function of time at 120°C

The viscosity test was performed at the lower temperature of 120°C that is generally used for WMA. Figure 8 depicts the viscosity change of PG 64-22 and rubberized binders with wax additive for 120 minutes at 120°C. The 20% CRM binder cannot be illustrated as the viscosity value exceeded a measurable range, irrespective of wax additive. The result shows similar trends with the findings at 135°C. However, the viscosity values at 120°C exhibited less increase, compared to the result at 135°C, indicating that the wax additives into the rubberized binder are more effective to maintain the viscosity at 120°C than the standard temperature of 135°C. This trend is consistent for all CRM binders. In addition, the rubberized binder is found to be more stable for a longer period at 120°C than at 135°C.

The viscosity changes for 240 minutes were illustrated in Figure 9 to be compared with the result at 135°C. It is observed of similar viscosity change with the result for 120 minutes. Generally, there was no considerable viscosity change at 120°C, compared to 135°C. However, the CRM binders containing wax additives are found to have much higher reduction of viscosity value than the PG 64-22 WMA binders. This means that the effect of wax additive is obvious in rubberized binder, making it possible to reduce the mixing and compaction temperatures of CRM mixes.



(a)



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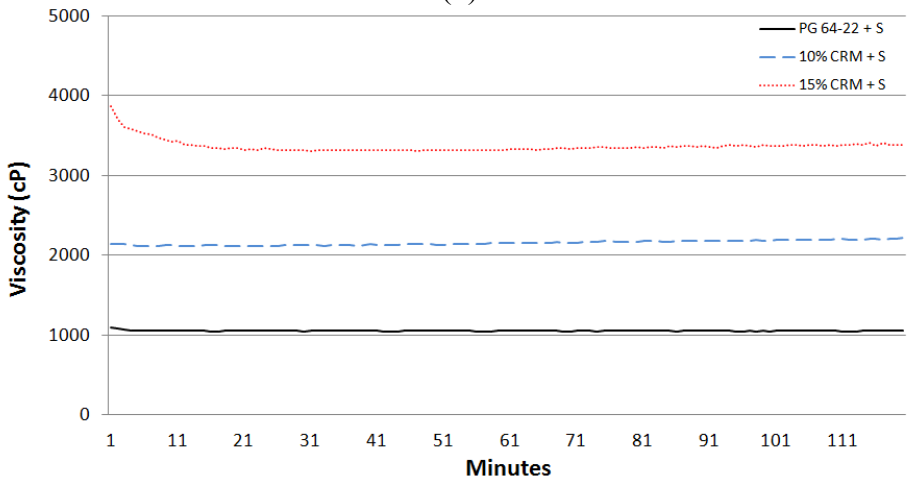
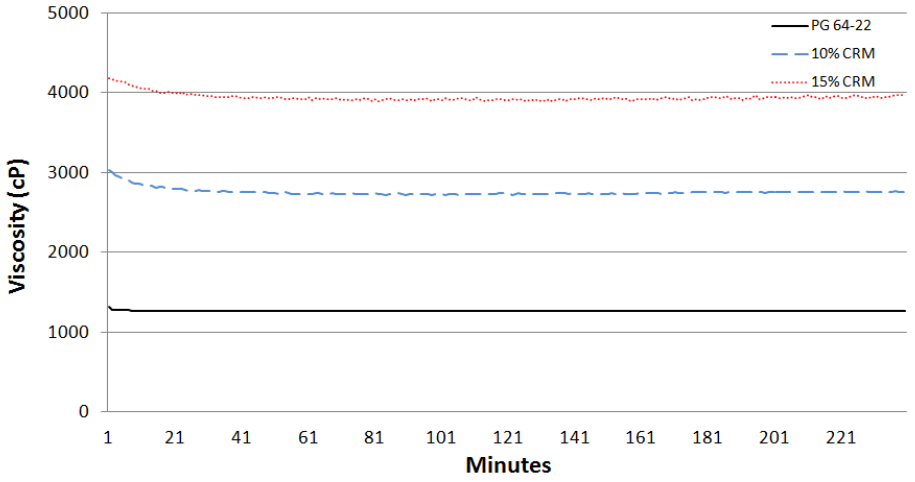
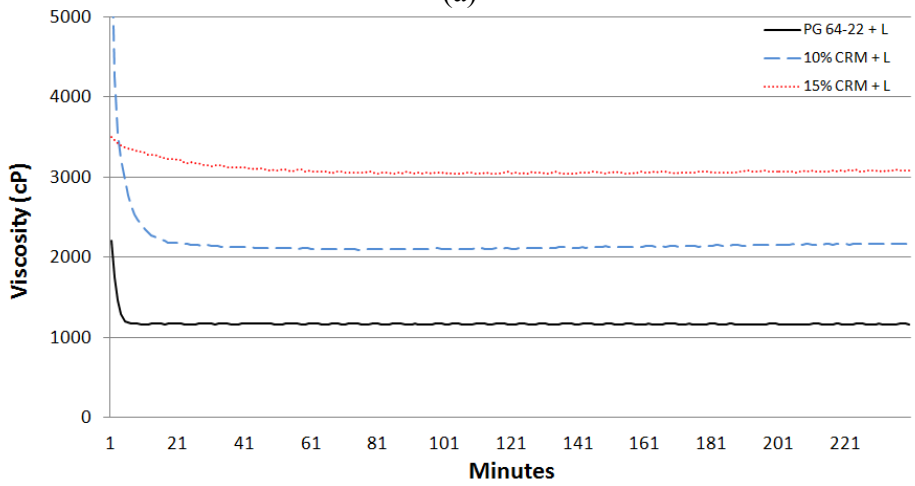


(c)

Figure 8. Viscosity change during 120 minutes at 120°C;
(a) Control binder, (b) WMA with LEADCAP, and (c) WMA with Sasobit.



(a)



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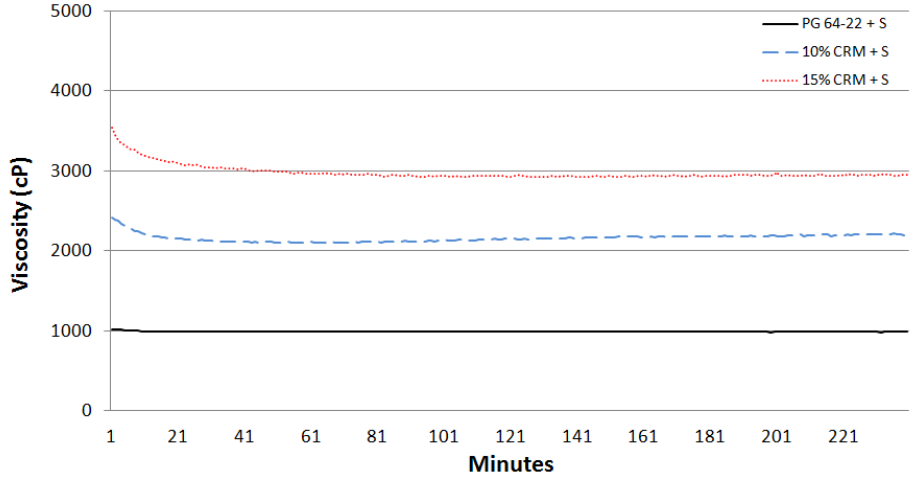


(c)

Figure 9. Viscosity change during 240 minutes at 120°C;
(a) Control binder, (b) WMA with LEADCAP, and (c) WMA with Sasobit.

The viscosity increase rates for the rubberized binders are shown in Table 5. From the results, the following trends are observed:

- At the standard temperature of 135°C, the rubberized binders showed a gradual increase in viscosity for 240 minutes.
- At the lower temperature of 120°C, there was little change in the viscosity of CRM binders over time, suggesting that the use of CRM at 120°C is more effective for the mix stability than at 135°C.
- The CRM mixes with the warm additive can be used for a better haul management, based on the result at 120°C, especially for long-haul projects.
- Generally, the addition of wax additive into the rubberized binders resulted in insignificant effect on the increase rate of viscosity at both testing temperature of 135°C and 120°C.

Table 5. Increase Rate of Viscosity for the CRM binder with wax additive;
(a) 10% rubberized binders with wax additives
(b) 15% rubberized binders with wax additives
(c) 20% rubberized binders with wax additives
(a)

Temperature	Minutes	Increase Rate (%)		
		10%CRM	10%CRM+L	10%CRM+S
135°C	30	0.0*	0.0	0.0
	60	4.4**	2.6	2.6
	90	8.3	6.3	8.5
	120	12.0	9.6	12.8
	150	14.6	13.8	15.7
	180	17.0	15.7	17.6
	210	18.5	17.6	17.6
	240	19.3***	19.4	19.4
120°C	30	0.0	0.0	0.0
	60	-1.8	-1.8	-0.6
	90	-2.3	-2.4	-0.6
	120	-1.4	-1.7	1.2
	150	-1.8	-1.2	1.7
	180	-0.9	-0.6	2.9
	210	-0.5	0.6	3.4
	240	-0.5	1.2	4.0

* $\left\{1-\frac{\text{Viscosity at 30 minutes}}{\text{Viscosity at 30 minutes}}\right\}\times 100\%$

** $\left\{1-\frac{\text{Viscosity at 30 minutes}}{\text{Viscosity at 60 minutes}}\right\}\times 100\%$

*** $\left\{1-\frac{\text{Viscosity at 30 minutes}}{\text{Viscosity at 240 minutes}}\right\}\times 100\%$

Table 5. (Continued)

(b)

Temperature	Minutes	Increase Rate (%)		
		15%CRM	15%CRM+L	15%CRM+S
135°C	30	0.0*	0.0	0.0
	60	3.1**	1.3	2.7
	90	7.1	6.3	5.9
	120	10.3	8.5	7.7
	150	12.8	11.2	10.0
	180	14.8	12.3	12.2
	210	15.7	12.8	13.3
	240	16.6***	14.8	15.8
120°C	30	0.0	0.0	0.0
	60	-1.0	-2.0	-3.0
	90	-1.3	-2.9	-3.4
	120	-1.3	-2.4	-4.3
	150	-1.0	-2.9	-3.8
	180	-1.0	-2.4	-3.8
	210	0.0	-2.4	-3.8
	240	0.0	-2.0	-3.4

(c)

Temperature	Minutes	Increase Rate (%)		
		20%CRM	20%CRM+L	20%CRM+S
135°C	30	0.0*	0.0	0.0
	60	2.9**	3.6	3.4
	90	4.7	5.5	9.2
	120	7.6	7.3	13.0
	150	9.7	9.0	14.6
	180	12.1	12.6	17.1
	210	13.1	13.9	19.4
	240	14.8***	15.4	20.3

Effect of blending time

Rotational viscosity at 135°C (Standard test)

Figure 10 shows the high temperature viscosity of the 10% CRM binder, the CRM binder with LEADCAP, and the CRM binder with Sasobit as a blending time, at 177°C, increases from 1 to 90 minutes. Although the viscosity of 10% CRM binder increases with blending time (except for 90 minutes), the increasing rate is not significant. The increasing rate (0%, 4% and 8% increase for 1, 30 and 60 minutes blending time, respectively) seemed to have linear relationship with blending time. In general, the CRM binders with LEADCAP were found to have similar decreasing rate at all blending times. The similar trend has been observed for the CRM binder containing Sasobit. At 135°C, with 90 minutes blending time, the average decreasing rate of viscosity due to the addition of wax additives is higher compared to the other blending time. The addition of wax additives into the CRM binder was efficient to reduce the viscosity of the binder regardless of blending time. The viscosity of the binders with 1 minute blending time showed relatively low values whereas the binders with 60 minutes blending time have the highest viscosity value among all the binders. In addition, both wax additives exhibited similar viscous performance, indicating that the specific difference depending on the additives was not observed in the viscosity result. All viscosity results showed much less values than the maximum requirement (3,000 cP) at 135°C suggested by Superpave.

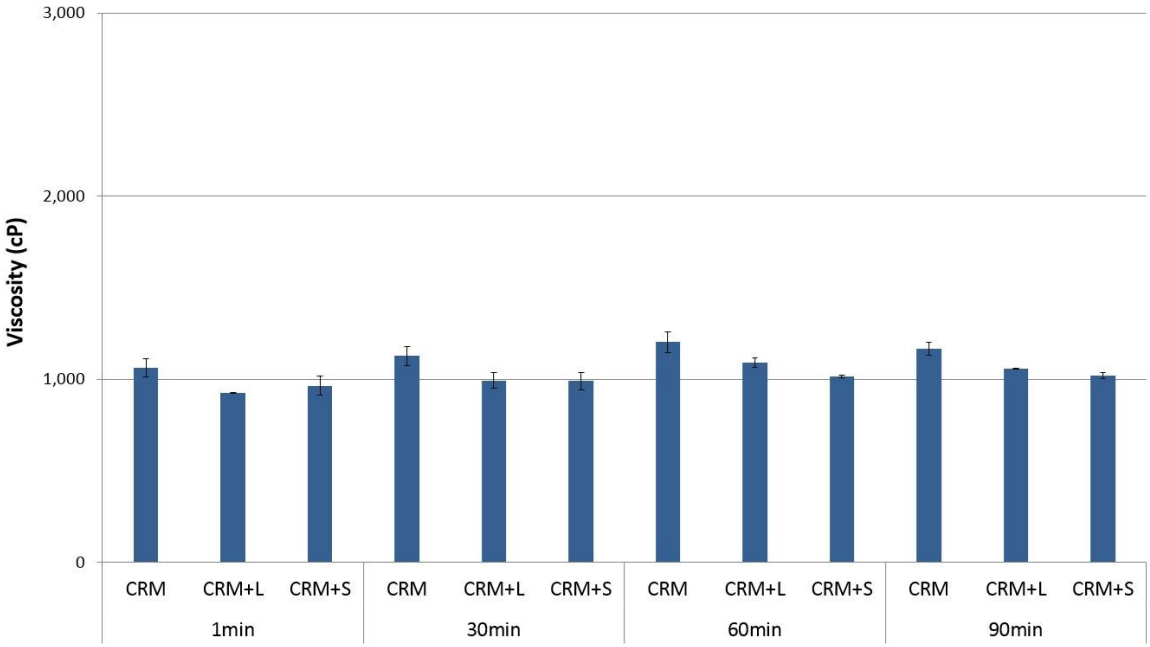


Figure 10. Viscosity of CRM binders with wax additive at 135°C as a function of blending time.

Rotational viscosity as a function of time at 135°C

While the addition of warm additives can significantly decrease the viscosity, the gradual increase of viscosity of CRM binder over time is mentioned in previous study (Lougheed and Papagiannakis, 1996). The viscosity of all the binders was measured and recorded during 120 and 240 minutes to evaluate the change of viscosity over a long period. Figure 11 shows the change of the viscosity as a blending time increases from 1 to 90 minutes. The viscosities of all the binders were found to have gradually increased after 15 to 30 minutes regardless of blending time, as expected. Although the addition of wax additives reduced the viscosity of the binder, the warm CRM binder also showed gradual increase of viscosity over time. The viscosity curves over 120 minutes of CRM binder without wax additives were formed at highest position and the gap between control CRM binder and warm CRM binders was found to have little change over the whole testing period. This result indicates that the addition of wax additives into CRM binder is effective irrespective of blending time of CRM binder. In most cases the gap between the binders containing wax additives was negligible, suggesting that both the additives have similar performance in terms of reducing the viscosity. No significant change has been observed as a function of blending times over 120 minutes.

One of the benefits of wax additive is to increase the hauling distance and period by reducing the viscosity of the binder. The haul management of asphalt mixture usually depends on the binder viscosity. The viscosity change was observed for 240 minutes to evaluate the longer haul management of CRM binder. The results are shown in Figure 12 which presents the time versus viscosity curve over 240 minutes at 135°C. It is clear that

the viscosity of CRM binders at any blending time was increased over time. The viscosity curves were gradually increased approximately after 20 minutes. Also, the difference between the two viscosity curves of CRM binder containing wax additives was insignificant regardless of blending time. In general, the gap between control CRM binder and the warm CRM binder was observed to be gradually increased over 240 minutes.

Table 6 exhibits the increase rate of viscosity of CRM binders over a long period at 135°C. For each blending time, eight different time periods with a 30 minutes interval period over the 240 minutes were selected. The increase rate of viscosity was measured. The viscosity of all the CRM binders is observed to have the increase rate between 16% and 24% at the end of 240 minutes. Even though there was slight difference by the binder type at the same blending time, there was no significant difference due to the blending time. While the additives resulted in the lower viscosity values than the control CRM binder, the viscosity of warm CRM binders steadily increased over the whole testing period.

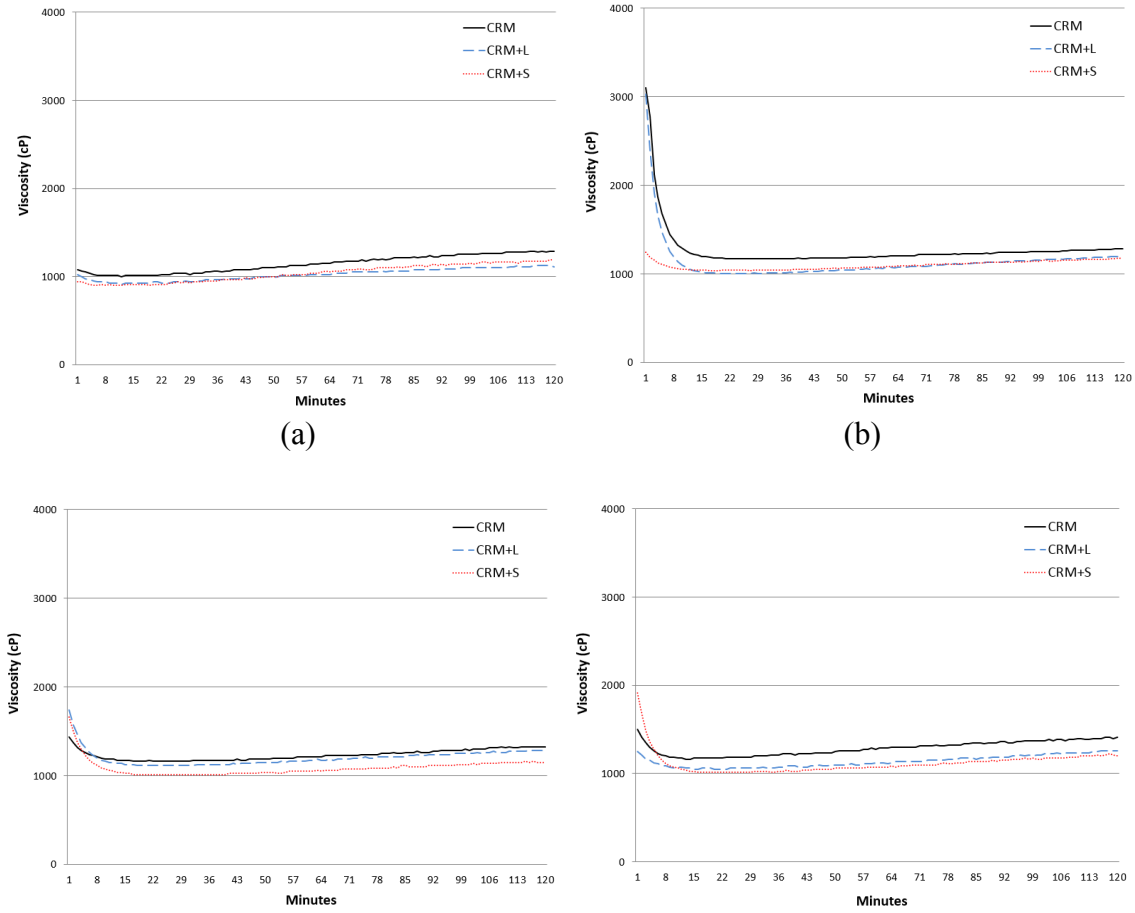


Figure 11. Viscosity change during 120 minutes at 135°C;
(a) 1 minute, (b) 30 minutes, (c) 60 minutes, and (d) 90 minutes

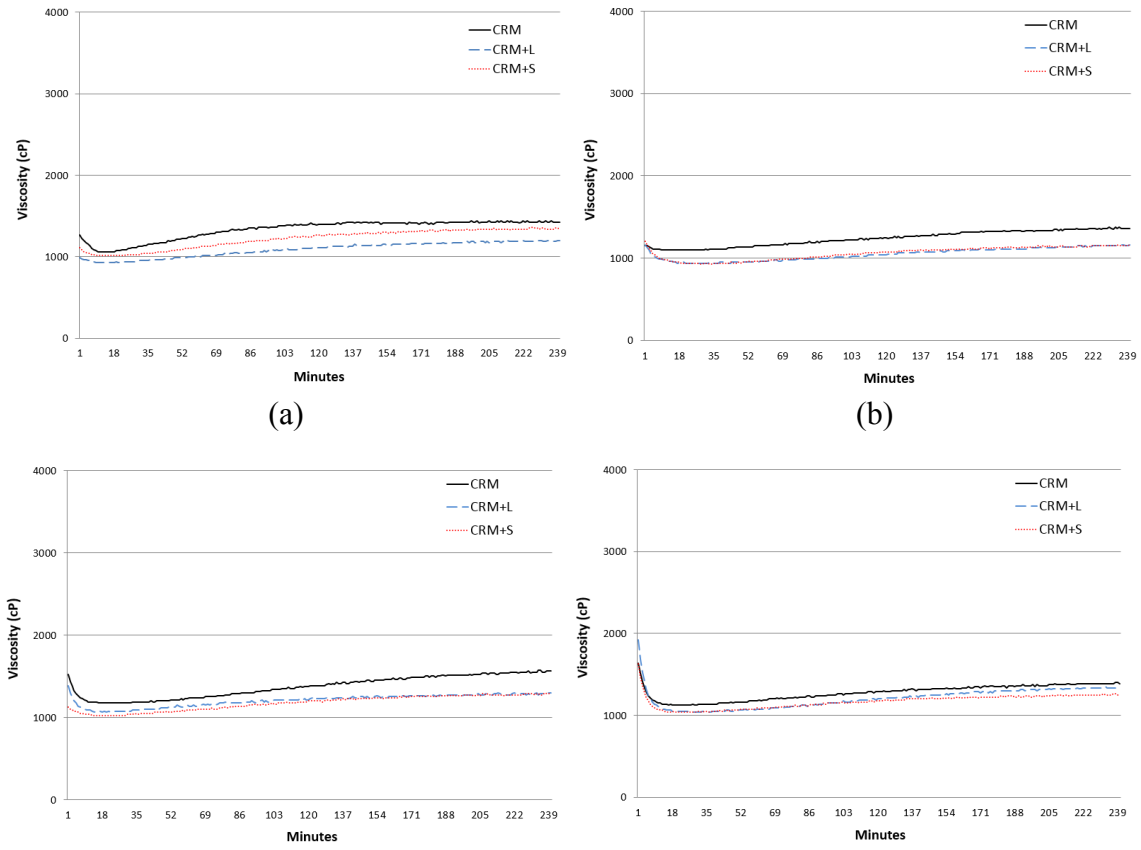


Figure 12. Viscosity change during 240 minutes at 135°C;
(a) 1 minute, (b) 30 minutes, (c) 60 minutes, and (d) 90 minutes

Table 6. Increase Rate of Viscosity for the CRM binder with wax additive at 135°C.

Temperature	Blending time	Minutes	Increase Rate (%)		
			CRM	CRM+L	CRM+S
135°C	1 minute	30	0.0*	0.0	0.0
		60	10.9**	5.0	7.9
		90	17.4***	10.6	14.6
		120	19.6	14.6	18.8
		150	21.1	16.5	21.2
		180	21.1	18.3	21.9
		210	21.1	20.0	23.4
		240	21.1	20.8	23.4
	30 minutes	30	0.0	0.0	0.0
		60	4.3	2.6	2.6
		90	8.3	6.3	8.5
		120	12.0	9.6	12.8
		150	14.6	13.8	15.7
		180	17.0	15.7	17.6
		210	18.5	17.6	17.6
		240	19.3	19.4	19.4
	60 minutes	30	0.0	0.0	0.0
		60	4.1	5.4	4.6
		90	9.6	8.4	9.8
		120	14.5	10.3	13.5
		150	19.0	12.1	15.3
		180	21.0	13.9	17.8
		210	23.6	14.7	18.6
		240	24.8	16.3	20.2
	90minutes	30	0.0	0.0	0.0
		60	4.3	3.5	4.6
		90	9.1	7.8	8.8
		120	12.6	13.5	11.7
		150	15.1	17.0	13.5
		180	16.7	19.4	15.3
		210	18.2	21.7	17.0
		240	18.9	22.4	17.8

* $\left\{1-\frac{\text{Viscosity at 30 minutes}}{\text{Viscosity at 30 minutes}}\right\}\times 100\%$

** $\left\{1-\frac{\text{Viscosity at 30 minutes}}{\text{Viscosity at 60 minutes}}\right\}\times 100\%$

*** $\left\{1-\frac{\text{Viscosity at 30 minutes}}{\text{Viscosity at 240 minutes}}\right\}\times 100\%$

Rotational viscosity at 120°C

The rotational viscosity test was performed at 120°C that is typically a mixing temperature for WMA mixture (Figure 13). With the test temperature decreased, the viscosity of the binders was increased at all blending time. Also, it is clear that the wax additives can significantly reduce the viscosity in the CRM binder. This effect is consistent for the tests done with all CRM binders containing the additives. At 120°C the viscosity of CRM binder increases with the blending time like at 135°C. The CRM binder with LEADCAP was found to have 20.6% decrease in viscosity value with 30 minutes blending time whereas for the other blending times the decreasing rate is in between 9.5% and 15.1%. Similar trend has been observed for the binder with Sasobit where the decreasing rate is 20.3% with 30 minutes blending time. It suggests that the blending period of 30 minutes is effective for decreasing the viscosity value of CRM binders by the addition of wax additives. In addition, the decreasing rate of viscosity at 120°C was higher than at 135°C irrespective of the blending time, suggesting that the addition of wax additive is more efficient to reduce the viscosity of the binder at 120°C. Even though the viscosity of the binders measured at 120°C exhibited higher values than the binders at 135°C, all values satisfied the requirement (maximum 3000 cP) suggested by Superpave.

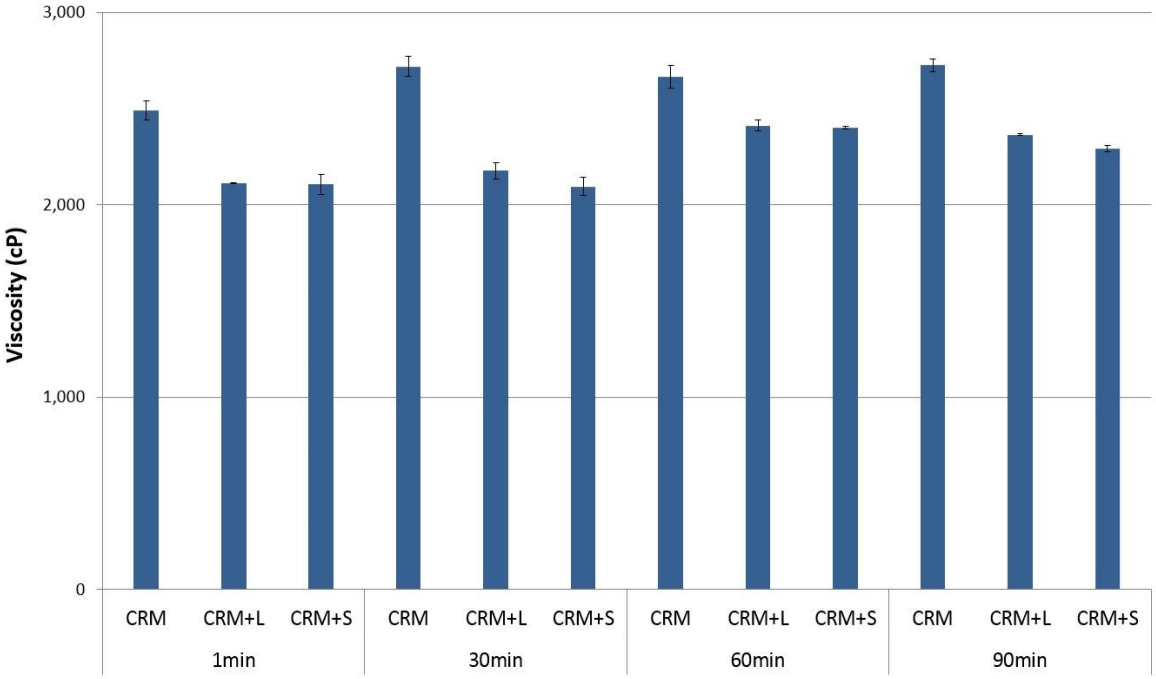


Figure 13. Viscosity of CRM binders with wax additive at 120°C as a function of blending time.

Rotational viscosity as a function of time at 120°C

Figure 14 illustrated the viscosity change of CRM binders over 120 minutes at 120°C. The results showed similar trend with the findings at 135°C. The viscosity seemed to increase approximately after 20 minutes. Unlike at 135°C, the viscosity of the CRM binders increased steadily over 120 minutes at 120°C. Also, the gap between the viscosity curves of control CRM binders and warm CRM binders was higher than the results at 135°C. It suggests that the addition of wax additives into the CRM binder is more influential at lower temperature. In addition, among all the blending times the gap between the CRM binder and CRM binder containing wax additives is found to have bigger with 30 minutes blending periods. There is no significant difference between the viscosity curves of two wax additives. It indicates that the wax additives have maintained similar viscous performance at lower temperature.

The viscosity change for 240 minutes is exhibited in Figure 15. The similar trend of viscosity change during 120 minutes was noticed for 240 minutes. In general, there was no significant change observed due to different blending periods. The decreasing rate of viscosity by the addition of wax additives is observed to be increased when compared to the results at 135°C.

For more detail analysis, the increase rates at 120°C were calculated and the results are shown in Table 7. There was little change in the viscosity of CRM binders over the long period at 120°C, compared to the results at 135°C. In most cases, the maximum increase rate of viscosity of CRM binder was over 20% at 135°C whereas at 120°C the increase rate was less than 10%. This result suggests that the CRM mixes containing the

warm additives can provide better haul management, based on the less increase rate of viscosity over a long period of time at lower temperature.

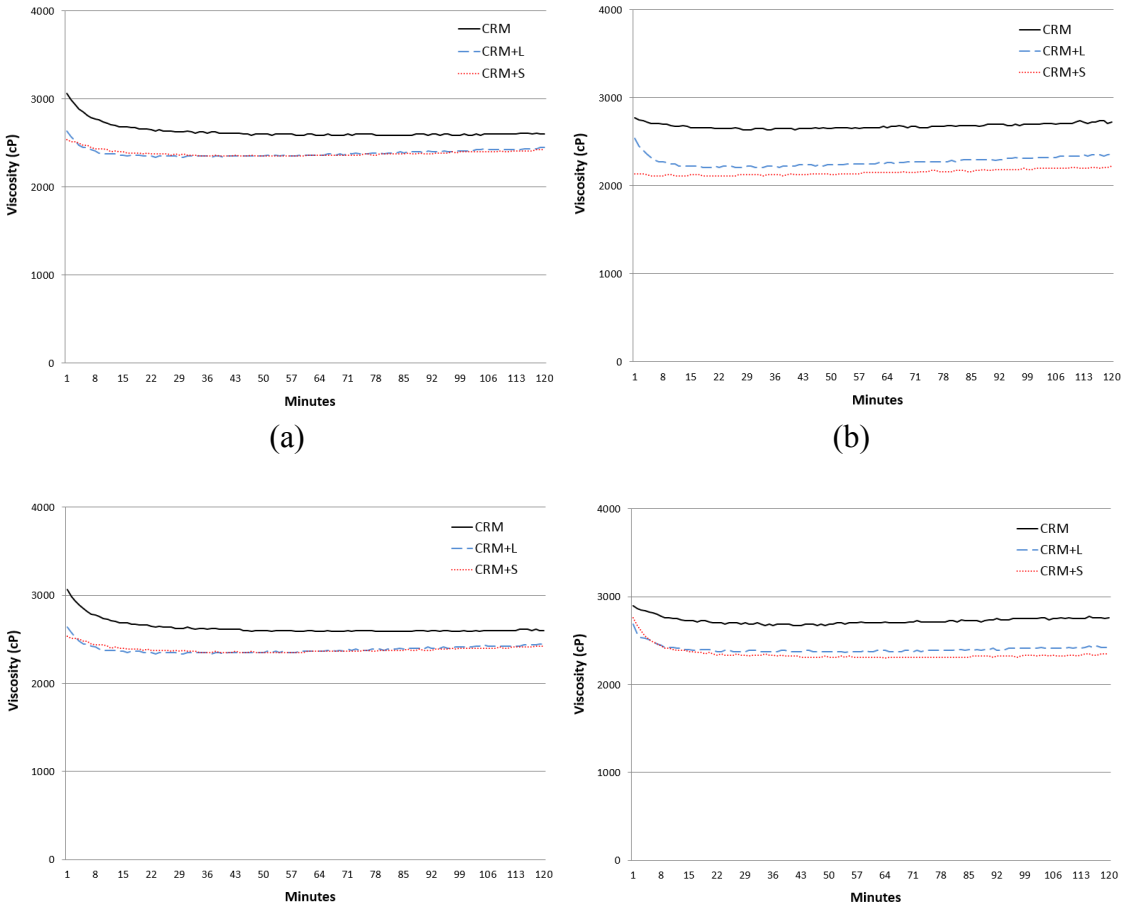


Figure 14. Viscosity change during 120 minutes at 120°C;
(a) 1 minute, (b) 30 minutes, (c) 60 minutes, and (d) 90 minutes

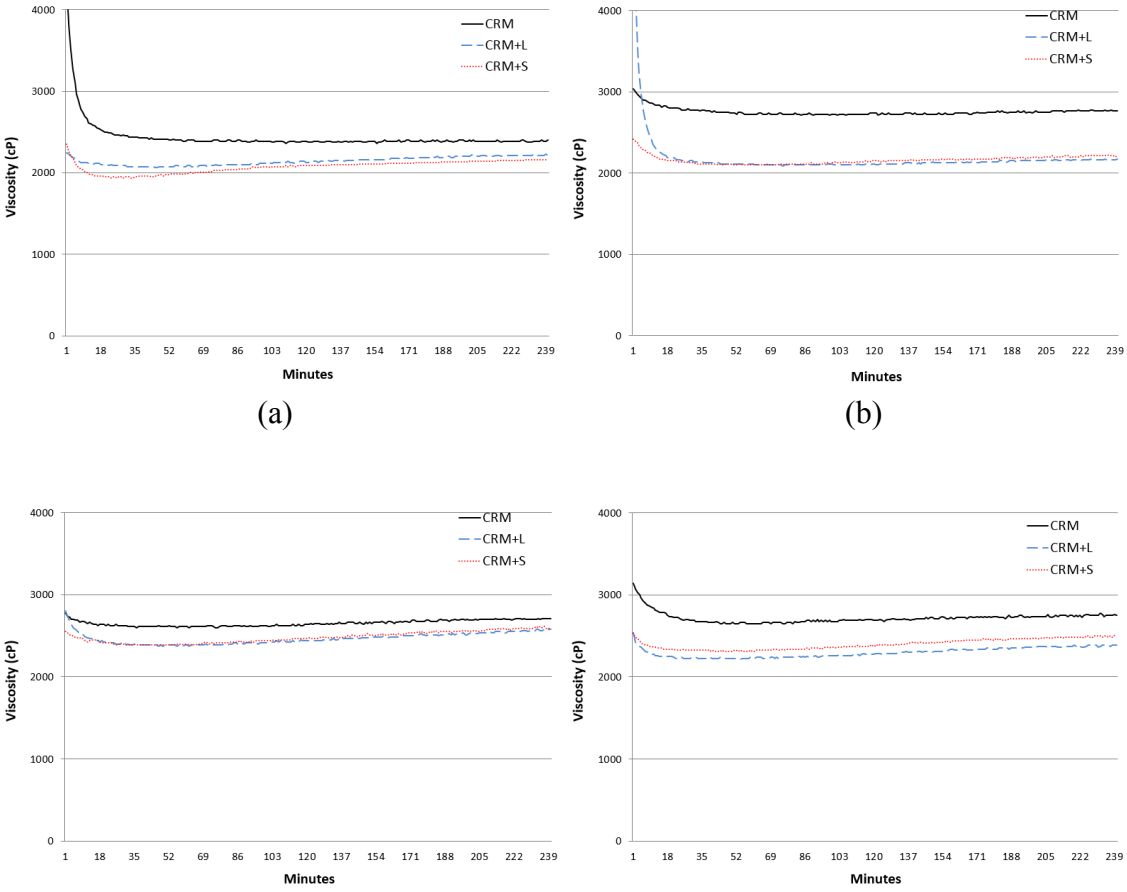


Figure 15. Viscosity change during 240 minutes at 120°C;
(a) 1 minute, (b) 30 minutes, (c) 60 minutes, and (d) 90 minutes

Table 7. Increase Rate of Viscosity for the CRM binder with wax additive at 120°C.

Temperature	Blending time	Minutes	Increase Rate (%)		
			CRM	CRM+L	CRM+S
120°C	1 minute	30	0.0*	0.0	0.0
		60	-2.6**	-0.6	1.9
		90	-2.6***	0.6	4.9
		120	-2.6	1.8	6.6
		150	-3.2	3.5	7.4
		180	-2.6	4.6	8.2
		210	-2.6	5.1	9.0
		240	-2.1	5.6	9.8
	30 minutes	30	0.0	0.0	0.0
		60	-1.8	-1.8	-0.6
		90	-2.3	-2.4	-0.6
		120	-1.4	-1.8	1.2
		150	-1.8	-1.2	1.7
		180	-0.9	-0.6	2.9
		210	-0.5	0.6	3.4
		240	-0.5	1.1	4.0
	60 minutes	30	0.0	0.0	0.0
		60	-0.5	-1.0	0.0
		90	-0.5	0.0	1.0
		120	0.5	1.0	2.5
		150	0.9	2.5	5.0
		180	2.3	4.0	5.4
		210	2.8	5.4	6.8
		240	3.2	6.3	7.7
	90minutes	30	0.0	0.0	0.0
		60	-1.4	-0.6	-0.5
		90	0.0	0.6	1.1
		120	0.0	1.6	2.6
		150	0.9	3.2	3.6
		180	1.8	4.3	5.6
		210	2.3	5.8	6.5
		240	2.3	6.3	7.0

* $\left\{1 - \frac{\text{Viscosity at 30 minutes}}{\text{Viscosity at 30 minutes}}\right\} \times 100\%$

** $\left\{1 - \frac{\text{Viscosity at 30 minutes}}{\text{Viscosity at 60 minutes}}\right\} \times 100\%$

*** $\left\{1 - \frac{\text{Viscosity at 30 minutes}}{\text{Viscosity at 240 minutes}}\right\} \times 100\%$

Statistical Analysis

The statistical significance of the change in the viscosity as a function of binder type and blending time was examined and the results are shown in Table 8. In general, the data indicated that the blending time has an insignificant effect on the viscosity of the warm CRM binders. It is evident that the differences of viscosity between the control CRM binders and two warm CRM binders at both testing temperatures were statistically significant for all blending times used in this study. The viscosity of control CRM binder with 1 minute blending period seemed to have significant difference with other blending period at both testing temperature. At 135°C, with 1 minute blending period, the binders containing Sasobit has insignificant difference with other blending time whereas at 120°C the opposite trend has been observed (except 30 minutes blending time). In general, the warm CRM binders with 30 minutes blending time are found to have significant difference at 120°C with the other blending periods. However, at 135°C the difference was non-significant. In most cases, the differences between the CRM binder containing LEADCAP and the CRM binder containing Sasobit are statistically insignificant within each blending time at both testing temperatures.

Table 8. Statistical analysis results of the viscosity value at 30 minutes as a function of binder type and blending time; (a) 135°C and (b) 120°C

(a)

Viscosity		1 minute			30 minutes			60 minutes			90 minutes		
		1*	2	3	1	2	3	1	2	3	1	2	3
1 minute	1	-	S	S	N	N	N	S	S	N	S	N	N
	2		-	N	S	N	N	S	S	S	S	S	S
	3			-	S	N	N	S	S	N	S	S	N
30 minutes	1				-	S	S	N	N	S	N	N	S
	2					-	N	S	S	N	S	N	N
	3						-	S	S	N	S	N	N
60 minutes	1							-	N	S	N	S	S
	2								-	S	N	S	S
	3									-	S	N	N
90 minutes	1										-	S	S
	2											-	N
	3												-

(b)

Viscosity		1 minute			30 minutes			60 minutes			90 minutes		
		1*	2	3	1	2	3	1	2	3	1	2	3
1 minute	1	-	S	S	S	S	S	S	N	N	S	N	S
	2		-	N	S	N	N	S	S	S	S	S	S
	3			-	S	N	N	S	S	S	S	S	S
30 minutes	1				-	S	S	N	S	S	N	S	S
	2					-	N	S	S	S	S	S	N
	3						-	S	S	S	S	S	S
60 minutes	1							-	S	S	N	S	S
	2								-	N	S	N	N
	3									-	S	N	N
90 minutes	1										-	S	S
	2											-	N
	3												-

* CRM binder containing wax additive 1: CRM binder (Control)
2: CRM binder + 1.5% LEADCAP
3: CRM binder + 1.5% Sasobit

N: non-significant, S: significant.

Summary and Conclusions

To evaluate the effect of crumb rubber and blending time on viscosity of rubberized binders containing wax additives, the rubberized binders were produced using four CRM percentages and four blending times. Two wax additives LEADCAP and Sasobit were used to produce warm asphalt binders. Rotational viscosity tests were carried out for three testing periods of 30, 120, and 240 minutes at two testing temperatures of 135°C and 120°C using the Brookfield viscometer. Based on the result of these tests, the conclusions were drawn for the materials used in this study as following,

1) The addition of crumb rubber into asphalt binder can significantly increase the viscosity of binders. With the rubber percentage increased, the binder viscosity increases at both testing temperatures, which is essential in increasing the binder film thickness for coating aggregates in the hot mixture to maintain the stability of asphalt mixtures.
2) The addition of wax additives was notably observed to reduce the binder viscosity of PG 64-22 and rubberized binders at the both testing temperatures of 135°C and 120°C, indicating that the additives are effective on the rubberized binders, as well.
3) The rubberized binders resulted in a gradual viscosity increase for a long term period, which is different with the PG 64-22 binder. However, the highest value meets the current requirement of maximum 3,000 cP at a standard testing temperature of 135°C, except for the 20% CRM binder.

4) The viscosity of rubberized binders at 120°C exhibited more stable curve, compared to the result at 135°C, suggesting that the rubberized mixes with wax additive are expected to have better haul management.
5) It is evident that the blending time has little influence on the viscosity of CRM binder with wax additives, based on the viscosity increase over a long period.
6) It is recommended that when 20% rubberized binders are used, the mixing temperature should be increased for the practical use.
7) The higher the rubber contents, the steeper the viscosity change over time, indicating that the binder with higher rubber contents is more sensitive for gradual viscosity increase.

This chapter (IV) includes a part of the following publications;

Kim, H. H., Lee, S. J., "Effect of Crumb Rubber on viscosity of rubberized asphalt binders containing wax additives," *Construction and Building Materials*, 95, October 2015, 65-73.

Kim, H. H., Mazumder, M., Lee, S. J., "Effect of blending time on viscosity of CRM binders with wax additives," *Canadian Journal of Civil Engineering* (Submitted)

V. RHEOLOGICAL PROPERTIES

Introduction

Wax additives are mainly used as flow improver in asphalt concrete and mastic asphalt. Wax is known to show a softening effect on the binder and asphalt mix at high temperatures, resulting in improved compaction. Wax crystallization below compaction temperature generally reinforces rutting properties of asphalt pavements. However, the crystallization makes the asphalt binder more brittle and susceptible to cracking at low temperature. Consequently, the use of wax additives may generate a potential threat in asphalt pavements. (Edwards and Redelius, 2003; Hesp, 2004; Edwards et al., 2006). If the WMA technologies are incorporated with the CRM binder, the cracking properties of WMA containing wax additive are thought to be improved as a result of the rubber influence. In addition, it is expected that the high managing temperature of CRM binder can be decreased by the addition of wax additives.

In this study, the performance properties of CRM binders containing wax additives are investigated through Superpave binder tests. The cracking properties are intensely examined considering the wax crystallization. The connectivity of rutting and cracking property is evaluated by aging and CRM contents. Figure 16 shows a flow chart of the experimental design used in this performance property study.

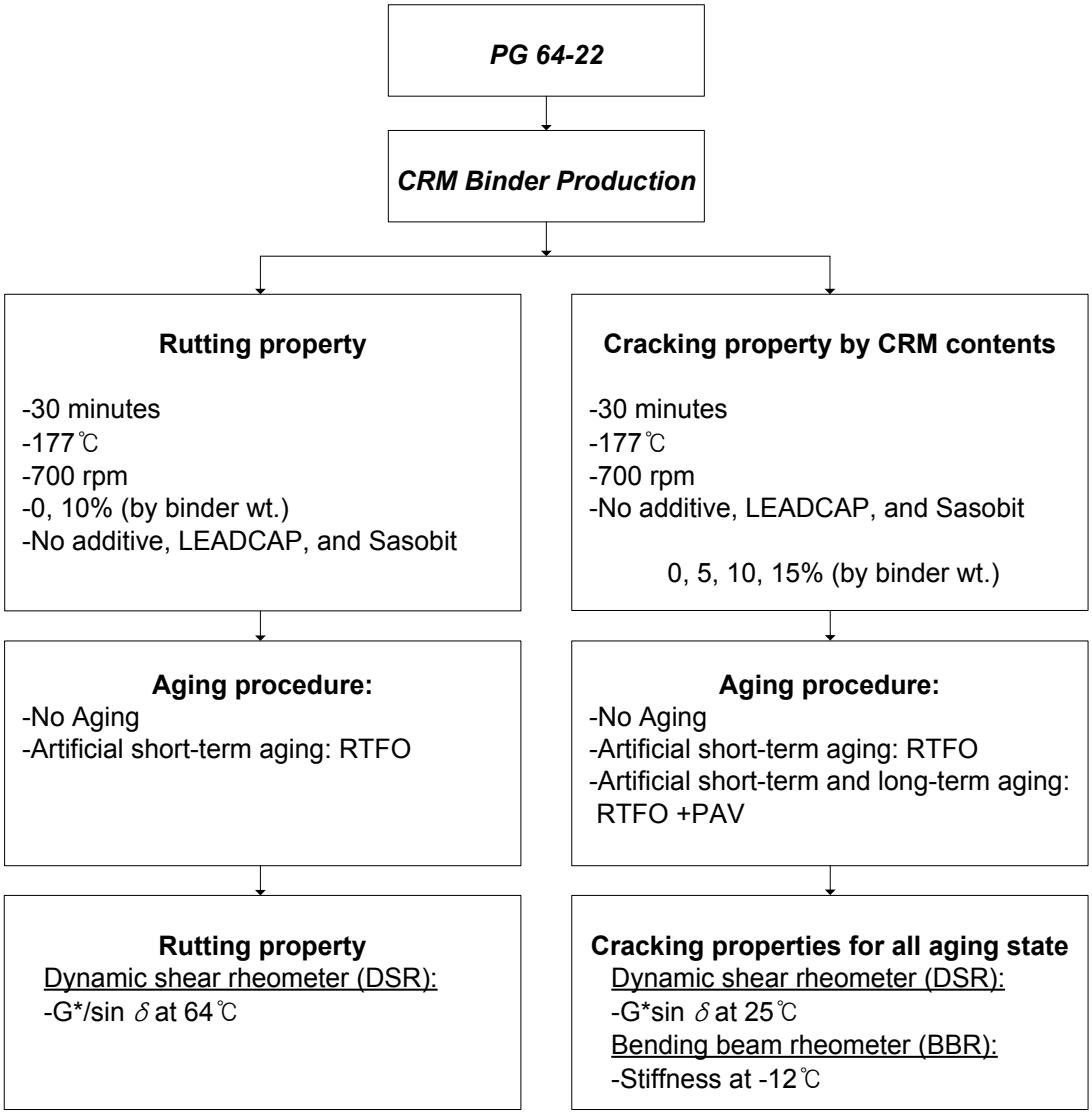


Figure 16. Flow chart of experimental design procedures of the rheology study.

Experimental Program

Materials

Asphalt binder

One PG 64-22 binder was used as a base binder in this study. The details of these base binders are described in Chapter 4.

Crumb rubber modifier (CRM)

The CRM (-40 mesh (0.425 mm)), produced by mechanical shredding at ambient temperature, was obtained from one source. The details of the CRM are described in Chapter 4.

Wax additives

Two commercial wax additives of LEADCAP and Sasobit were used for this research. The details of the wax additives are described in Chapter 4.

Dynamic Shear Rheometer (DSR) test

The high temperature rheological properties of each binder were measured using a dynamic shear rheometer (DSR) per AASHTO T 315. In the DSR test, the original binders and RTFO residual binders were tested with 25mm spindle and parallel plate and the binders (RTFO+PAV residual) were tested using an 8mm parallel plate at 25°C. The complex shear modulus (G*) and phase angle (δ) of binder was measured from 64°C to 88°C, respectively. Binders were tested in all aging condition such as original, short term

aging, and long term aging. The CRM binder was tested using a 2mm gap between the plates, while a 1mm gap was used for the control binders.

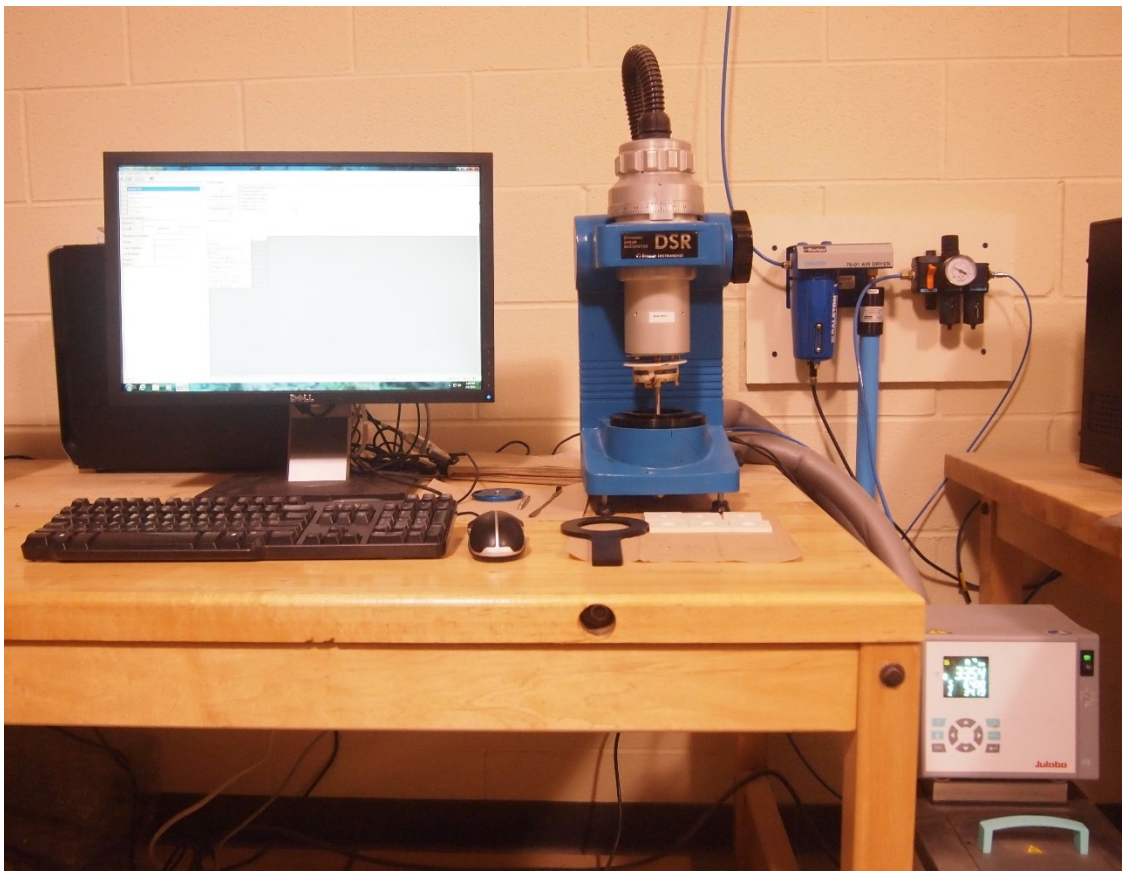


Figure 17. Dynamic Shear Rheometer (DSR).

Bending Beam Rheometer (BBR) test

BBR test was utilized to evaluate crack property at low temperature per AASHTO T 313. The stiffness and m-value was measured from -12°C. Figure 18 shows BBR tester used in this study.

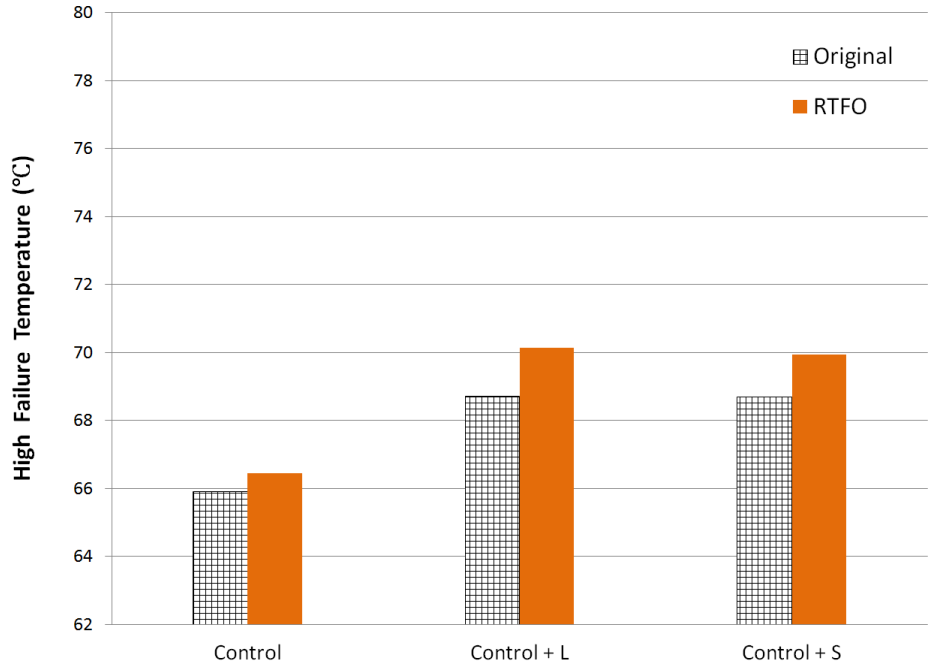
Usually, two aging processes (RTFO+PAV residual) have to be performed to conduct the BBR test. However, samples in original and RTFO condition were tested to evaluate cracking resistance with original and short-term aging state at low temperature.

Figure 18. Bending Beam Rheometer (BBR).

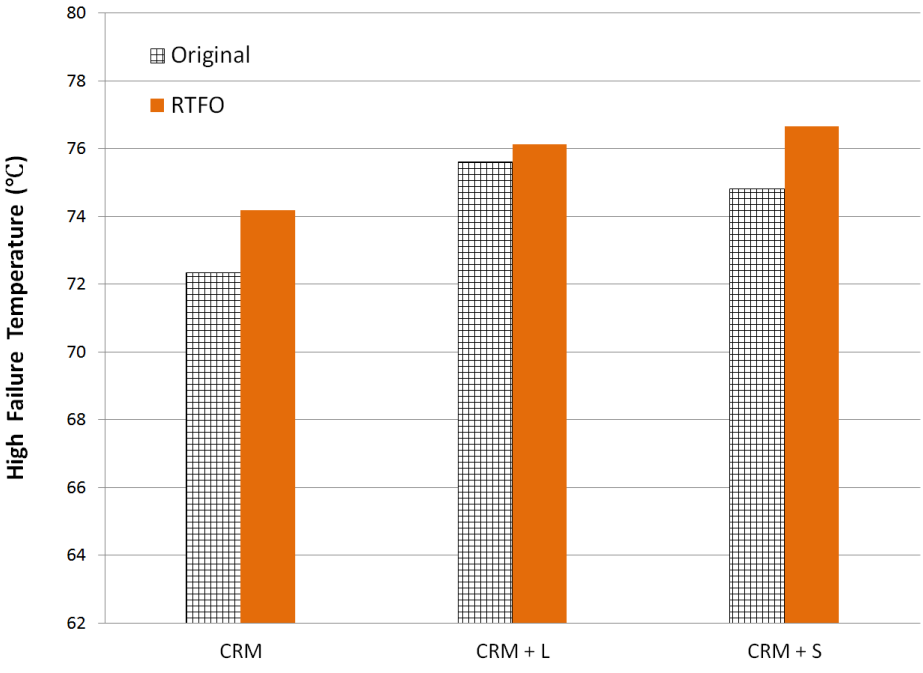
Results and Discussions

Rutting property

The higher failure temperature from the DSR test demonstrates that the binders are less susceptible to permanent deformation at high pavement temperature (The Asphalt Institute, 2003). The high failure temperature of binders in original state and after short term aging was measured and Figure 19 provides the results. In general, the CRM asphalt binder resulted in the higher failure temperature than the control binders regardless of aging state. In addition, the use of wax additive into binders was observed to increase the higher failure temperature in both aging states, indicating that the addition of wax additives positively affect permanent deformation in high temperature. In short, both the wax additives and the crumb rubber particles play a significant role in improving rutting resistance.



(a)



(b)

Figure 19. High failure temperatures; (a) Control binder and (b) CRM binder

Cracking properties by CRM contents

Fatigue cracking property

In general, the lower G*sin δ values are considered to be desirable attributes from the standpoint of fatigue cracking resistance (Asphalt Institute, 2003). The G*sin δ values of the control and CRM binders with wax additives were determined using the DSR at 25°C and the results are illustrated in Figure 20. The G*sin δ values were found to be 2563, 2270, and 2987 kPa for the binders of Control (PG 64-22), Control+L (PG 64-22 + 1.5% LEADCAP), and Control+S (PG 64-22 + 1.5% Sasobit), respectively. This trend, the highest at the addition of Sasobit and the lowest at the addition of LEADCAP, was consistent in the rubberized binders. Form the results, it is predicted that the WMA binder containing the Sasobit has possible lower resistance on fatigue cracking at intermediate temperature compared to the control binder without the additives. Also, the G*sin δ values were found to be reduced with the increase of rubber content. It means that higher content of rubber can improve the fatigue cracking property at certain level.

Using one-way analysis of variance, the statistical significance of the change in the G*sin δ values was examined (Table 9). In general, the data indicated that the CRM content has a significant effect on the G*sin δ of the warm CRM binders. For the CRM contents of 10% and 15%, the differences within each wax type (for example, 10% control vs. 15% control) are statistically insignificant. Also, wax additives were observed to have significantly different influences on the CRM binders.

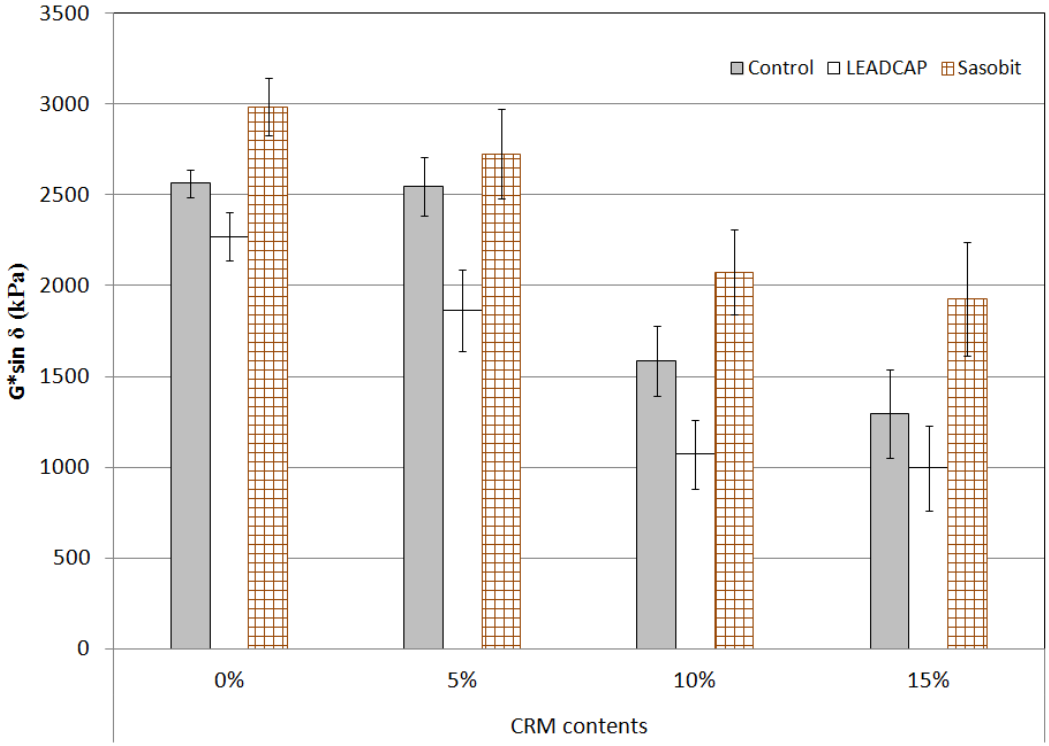


Figure 20. G*sin δ values at 25°C (RTFO + PAV residual)

Table 9. Statistical analysis results of the G*sin δ value as a function of rubber content and wax additive.

G*sin δ		0% CRM			5% CRM			10% CRM			15% CRM		
		1*	2	3	1	2	3	1	2	3	1	2	3
0% CRM	1	-	N	S	N	S	N	S	S	S	S	S	S
	2		-	S	N	S	S	S	S	N	S	S	N
	3			-	S	S	N	S	S	S	S	S	S
5% CRM	1				-	S	N	S	S	S	S	S	S
	2					-	S	N	S	N	S	S	N
	3						-	S	S	S	S	S	S
10% CRM	1							-	S	S	N	S	N
	2								-	S	N	N	S
	3									-	S	S	N
15% CRM	1										-	N	S
	2											-	S
	3												-

* CRM binder containing wax additive 1: CRM binder (Control)
2: CRM binder + 1.5% LEADCAP
3: CRM binder + 1.5% Sasobit

N: non-significant, S: significant.

Low temperature cracking property at original condition (No aging)

The BBR test was carried out at three aging states (original, RTFO aged, and RTFO+PAV aged) to find out the effect of aging level on low temperature cracking. Figure 21 illustrates the BBR test results of control and CRM binders with wax additives at original state (no aging). It is found that the addition of crumb rubber into the asphalt binder significantly decreases the low temperature stiffness. The creep stiffness of CRM binders containing LEADCAP additives was reduced by approximately 25%, compared to the control binder without wax additives. Also, with the percentage of crumb rubber increased, the binder stiffness was observed to be decreased, suggesting that the higher content of CRM decrease the toughness of mixtures at certain point and improve the cracking resistance of asphalt binder at low temperature. A general trend was observed that the addition of Sasobit into CRM binder increased the CRM binder's stiffness and this finding was consistent for all CRM contents. On the other hand, the addition of LEADCAP resulted in decreasing the stiffness, with the exception of CRM content of 5%. These trends were also observed in m-value results depicted in Figure 22. The binder containing LEADCAP exhibited the highest value for all CRM contents, except for 0%. However, the CRM binders with Sasobit showed obviously the lowest value for all CRM contents.

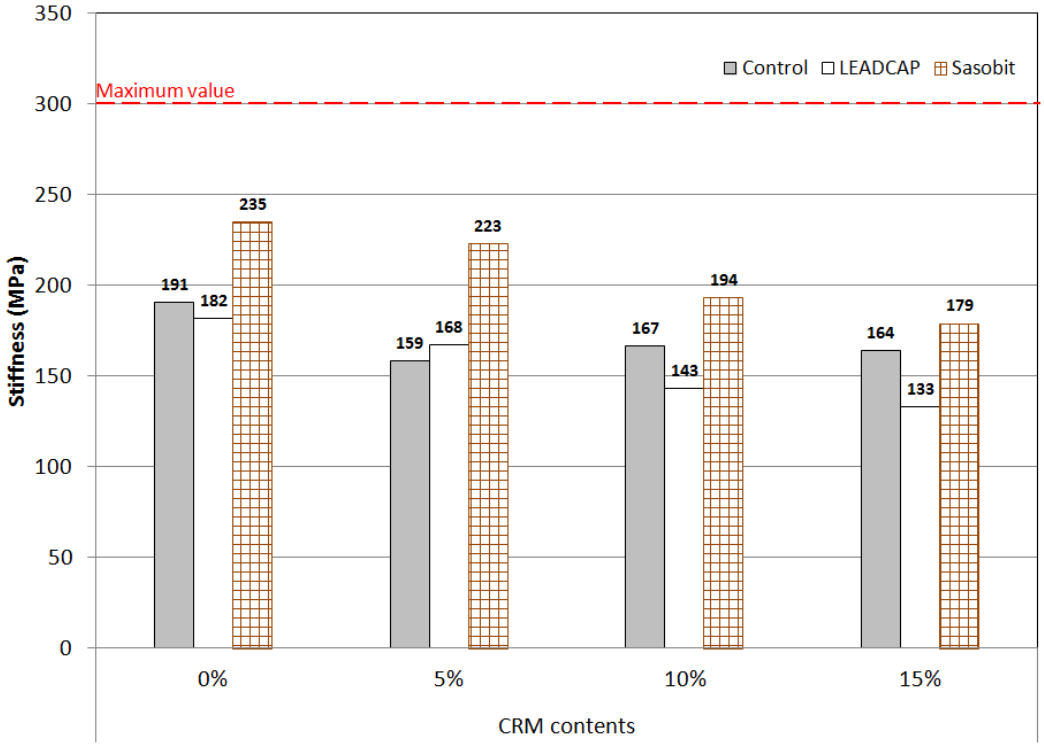


Figure 21. Stiffness at -12°C (Original).

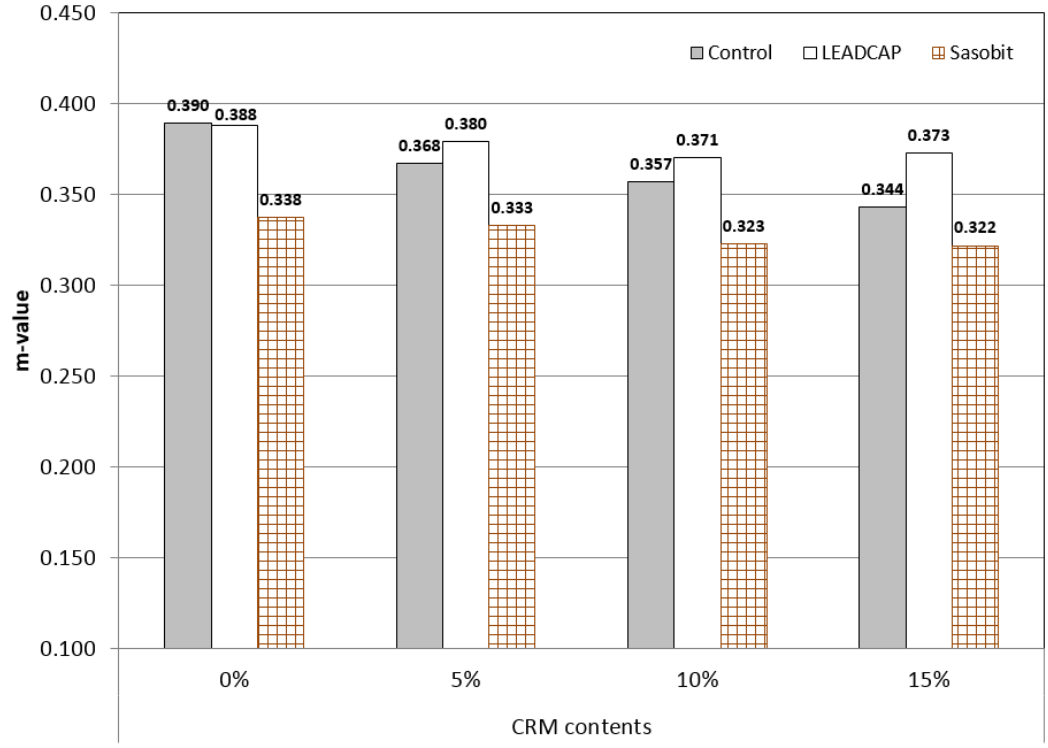


Figure 22. m-value at -12°C (Original).

Low temperature cracking property at short term aging condition

The stiffness values of RTFO residual binders are illustrated in Figures 23 and 24. After the RTFO aging process, the binders exhibited similar trend with the binders at original condition. The higher rubber content in asphalt binder is found to result in the lower creep stiffness of CRM binders. The addition of Sasobit into CRM binder showed the highest stiffness while the asphalt binders with LEADCAP showed the lowest stiffness values. This trend is same for all CRM contents used in this study. When the RTFO residual binders are compared with the binder at original condition, the binders after RTFO aging showed the higher stiffness values, indicating that the aging process made the asphalt binder brittle, as expected. The CRM content of 15% exhibited the lowest stiffness value compared to other CRM contents, meaning that the higher CRM content positively affect low temperature cracking resistance. Similar to the unaged state, the rubberized binders with Sasobit were found to have the lowest m-value within each CRM content. In addition, the short-term aging resulted in the decrease of m-value, regardless of the wax additive.

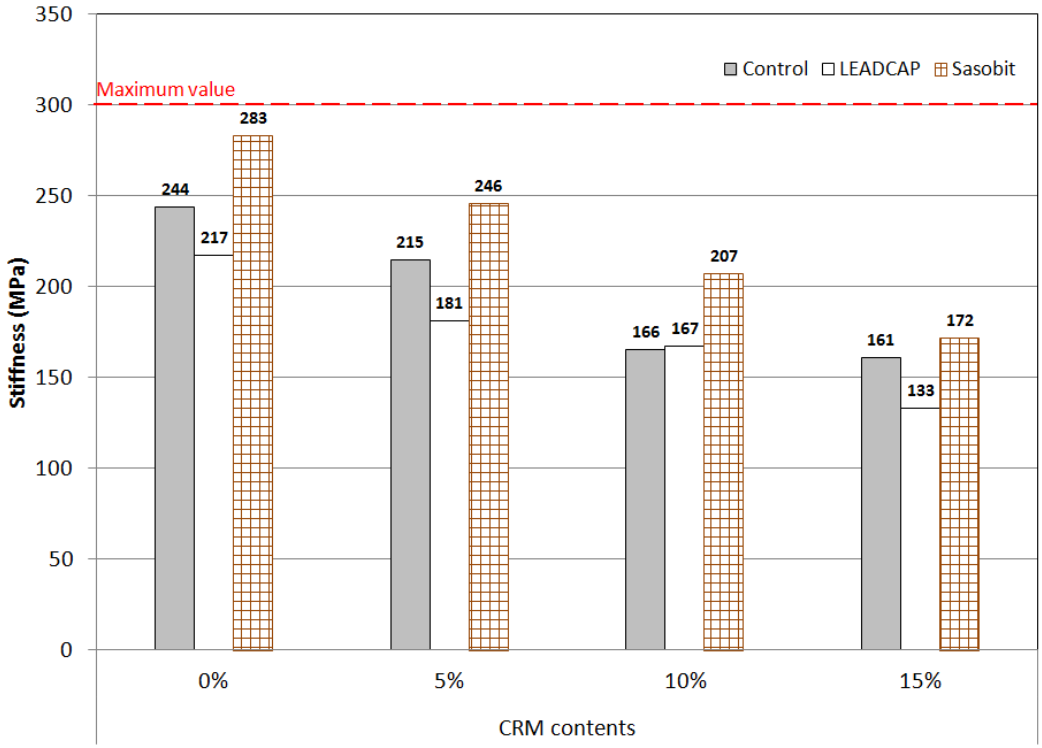


Figure 23. Stiffness at -12°C (RTFO residual).

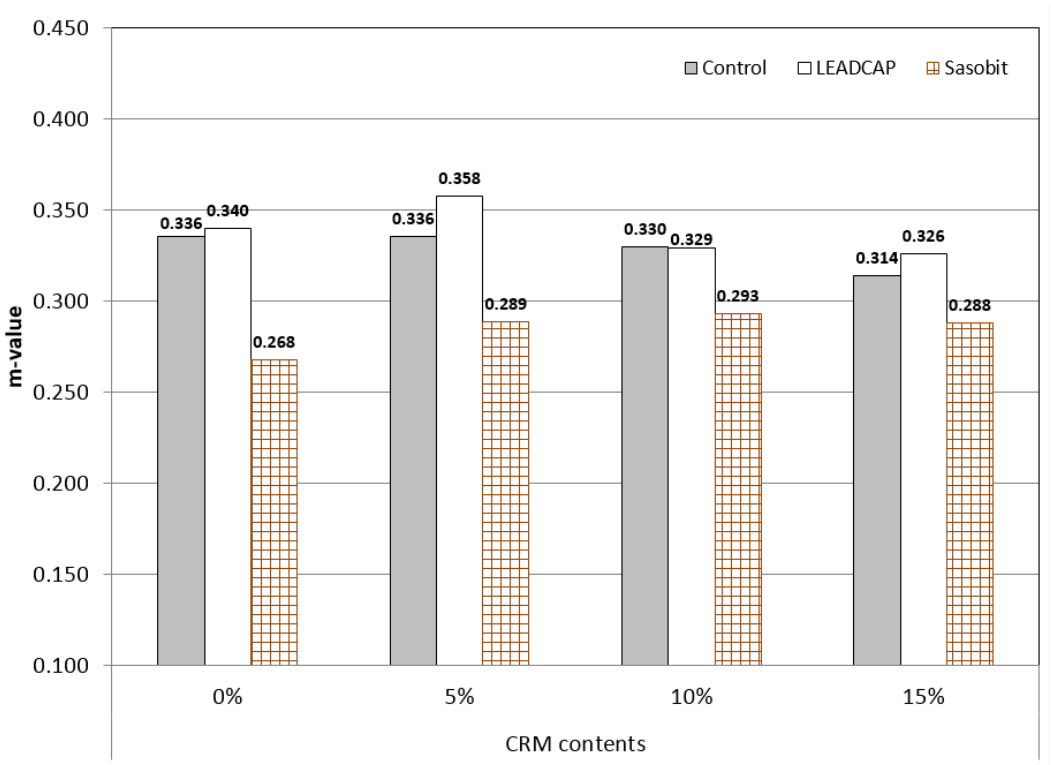


Figure 24. m-value at -12°C (RTFO residual).

Low temperature cracking property at long term aging condition

The BBR test results of RTFO+PAV residual binders are depicted in Figures 25 and 26. Superpave specification requires the creep stiffness to be less than 300 MPa. All the binders satisfied the requirement set forth by Superpave, except for the control binder with Sasobit and the 5% CRM binder with Sasobit, indicating that the addition of Sasobit may result in the binder being less resistant to low temperature cracking. This finding is consistent with previous studies (Edwards and Redelius 2003; Hesp 2004; Edwards et al. 2006). However, the stiffness values of CRM binders decreased with an increase of CRM content. The 15% CRM binder showed approximately 44% lower stiffness value than the control binder. The binders with LEADCAP resulted in similar trend. Especially, the addition of LEADCAP into the 15% CRM binder exhibited the lowest stiffness value (150 MPa), which is 48% lower than the control binder stiffness (287 MPa). The use of CRM to modify the asphalt binder is concluded to be greatly effective to improve the thermal cracking properties (measured from the BBR test at -12°C), and it is expected that 15% CRM binder with LEADCAP will have the best performance in terms of low temperature cracking resistance, among twelve binder types used in the study. In general, the rubberized binders with wax additive satisfied the minimum requirement of m-value of 0.300. Unlike to the stiffness property, the CRM contents were observed to have little influence on m-value results. However, the use of LEADCAP into CRM binders led to increase the value.

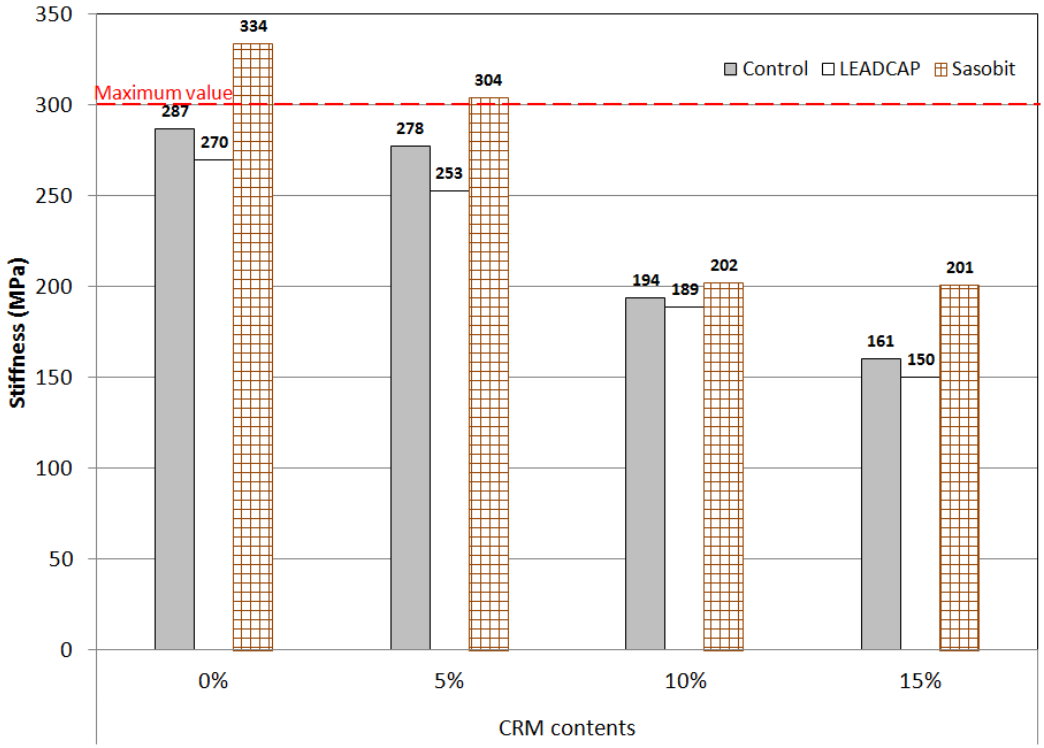


Figure 25. Stiffness at -12°C (RTFO + PAV residual).

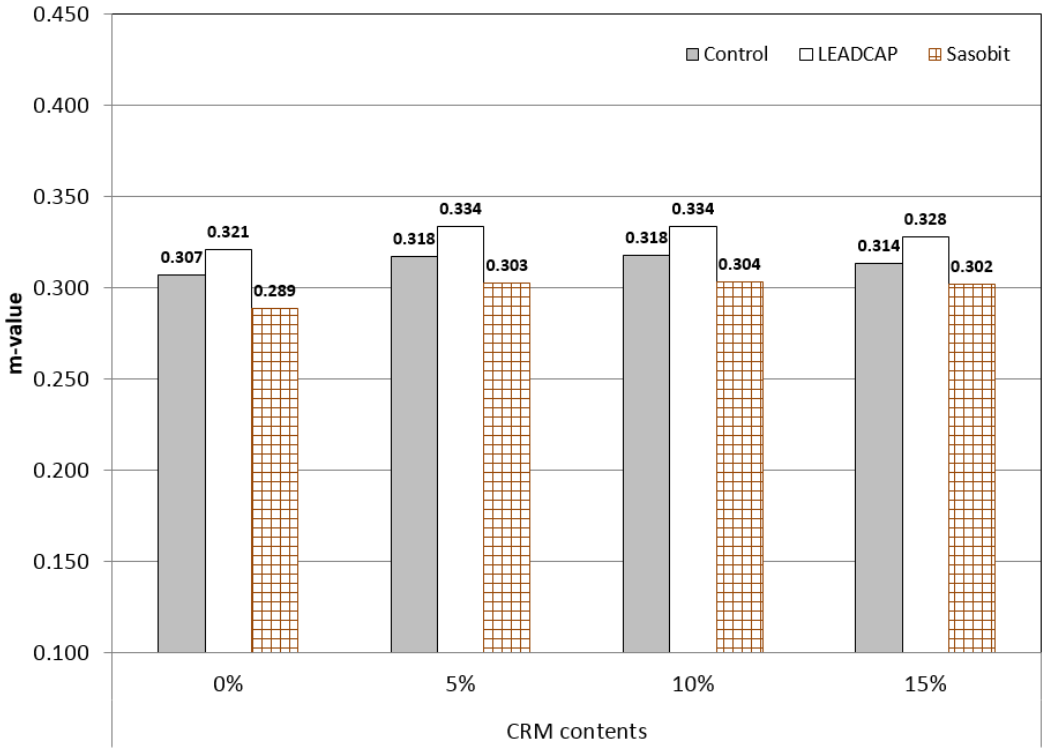


Figure 26. m-value at -12°C (RTFO + PAV residual).

The one-way analysis of variance was applied to investigate the statistical significance of the change in the stiffness and the results are summarized in Table 10. In original state, the CRM contents were found to have a significant difference in the stiffness within each binder type. In general, the differences between the CRM binders with Sasobit and LEADCAP were statistically significant for all CRM contents. The short-term aging (RTFO) state showed similar trend to original state. The data at RTFO state indicated that the CRM content has a significant effect on the stiffness. The stiffness values of control CRM binder and CRM binder containing LEADCAP at long-term aging (PAV) state were observed to be statistically insignificant within each CRM content. On the other hand, the differences between the control CRM binders and CRM binder containing Sasobit were statistically significant for all CRM content, except for 10% CRM.

Table 10. Statistical analysis results of the stiffness value as a function of rubber content and wax additive; (a) Original (b) RTFO aged (c) RTFO+ PAV aged

(a)

Stiffness		0% CRM			5% CRM			10% CRM			15% CRM		
		1*	2	3	1	2	3	1	2	3	1	2	3
0% CRM	1	-	S	S	S	S	S	S	S	N	S	S	S
	2		-	S	S	S	S	S	S	S	S	S	N
	3			-	S	S	S	S	S	S	S	S	S
5% CRM	1				-	S	S	N	S	S	N	S	S
	2					-	S	N	S	S	N	S	S
	3						-	S	S	S	S	S	S
10% CRM	1							-	S	S	N	S	S
	2								-	S	S	S	S
	3									-	S	S	S
15% CRM	1										-	S	S
	2											-	S
	3												-

1* CRM binder containing wax additive 1: CRM binder (Control)
2: CRM binder + 1.5% LEADCAP
3: CRM binder + 1.5% Sasobit

N: non-significant, S: significant.

Table 10. (Continued)

(b)

Stiffness		0% CRM			5% CRM			10% CRM			15% CRM		
		1*	2	3	1	2	3	1	2	3	1	2	3
0% CRM	1	-	S	S	S	S	N	S	S	S	S	S	S
	2		-	S	N	S	S	S	S	N	S	S	S
	3			-	S	S	S	S	S	S	S	S	S
5% CRM	1				-	S	S	S	S	N	S	S	S
	2					-	S	N	N	S	S	S	S
	3						-	S	S	S	S	S	S
10% CRM	1							-	N	S	N	S	N
	2								-	S	N	S	N
	3									-	S	S	S
15% CRM	1										-	S	N
	2											-	S
	3												-

(c)

Stiffness		0% CRM			5% CRM			10% CRM			15% CRM		
		1*	2	3	1	2	3	1	2	3	1	2	3
0% CRM	1	-	N	S	N	S	N	S	S	S	S	S	S
	2		-	S	N	S	N	S	S	S	S	S	S
	3			-	S	S	S	S	S	S	S	S	S
5% CRM	1				-	N	S	S	S	S	S	S	S
	2					-	S	S	S	S	S	S	S
	3						-	S	S	S	S	S	S
10% CRM	1							-	N	N	N	S	N
	2								-	N	N	S	N
	3									-	S	S	S
15% CRM	1										-	S	N
	2											-	S
	3												-

Summary and Conclusions

The rutting properties of CRM binder with wax additives were investigated through measuring G*/sin δ using DSR test at original and RTFO short term aging state. The CRM binders were produced in the laboratory with the percentage of 10% by the weight of binder. On the other hand, the CRM binders were produced using the percentages of 5%, 10% and 15% by the weight of binder in the laboratory to evaluate the effect of crumb rubber on cracking property of CRM binders containing wax additives. Warm CRM binders were made with the wax additives of LEADCAP and Sasobit. Fatigue cracking property was evaluated for RTFO+PAV residual samples at 25°C using the DSR equipment. In addition the BBR test was performed to measure creep stiffness of control and CRM binders containing wax additives at original, RTFO aging and RTFO+PAV aging conditions. The test results obtained from this study suggest an obvious trend and inherent relation of G*sin δ and low temperature stiffness. Conclusions that can be drawn from the research are:

1) Both the additives and crumb rubber particles were observed to be effective on increasing rutting resistance (measured from the DSR test at high temperature). It was found that the additives incorporated with the CRM binder plays a significant role in the resistance for permanent deformation of asphalt pavement.
2) The addition of crumb rubber into asphalt binder can significantly decrease the creep stiffness of CRM binder at low temperature which is helpful for better cracking resistance of asphalt binder.

3) The low temperature stiffness values were increased with the aging processes. This means that the aging negatively affect the thermal cracking resistance of binders, as expected.
4) The G*sin δ values of CRM binders were found to decrease with increasing the rubber percentage, suggesting that crumb rubber is effective in improving the fatigue cracking resistance.
5) The addition of Sasobit significantly increased the stiffness at -12°C of all the CRM binders. However, the stiffness values of control and CRM binders decreased with the addition of LEADCAP.
6) The stiffness differences among control, Sasobit, and LEADCAP are statistically significant within each CRM content. Therefore it is obvious that the use of warm additive into CRM binder significantly influences low temperature performance.
7) The use of CRM in asphalt binder resulted in a better resistance for low temperature cracking, and the 15% CRM binder containing LEADCAP is found to have the lowest stiffness among all the binders tested in this study.
8) The CRM contents are found to have little effect on the m-value property for all the aging stages.

This chapter (V) includes a part of the following publications;

Kim, H. H., Jeong, K. D., Lee, M. S., Lee, S.-J., "Performance properties of CRM binders with wax warm additives." *Construction and Building Materials*, 66(9), September 2014, 356-360.

Kim, H. H., Lee, S. J., "Evaluation of rubber influence on cracking resistance of CRM binders with wax additives," *Canadian Journal of Civil Engineering*, 43(4), February 2016.

VI. RECYCLING PROPERTIES

Introduction

Many research projects have concluded that the use of reclaimed asphalt pavement (RAP) into hot mix asphalt (HMA) pavements can help offset increased initial costs, conserve natural resources, and avoid disposal problems. Furthermore, the properties of properly designed recycled asphalt concrete materials have been proven to be comparable to new asphalt concrete pavements (Xiao 2006; Lee 2007).

The main objective of this research was to investigate the performance properties of recycled aged CRM binder containing wax additives through Superpave binder tests. The CRM binder was produced in the laboratory incorporating one CRM source (ambient) and one CRM percentage (10% by weight of asphalt binder), and the CRM binder was mixed with 1.5% of two wax additives (LEADCAP and Sasobit). The warm CRM binders were artificially aged using rolling thin film oven (RTFO) and pressure aging vessel (PAV), and the aged warm CRM binders were recycled with virgin CRM binder. The recycled warm CRM binders were artificially aged using the same aging processes to perform Superpave binder tests. In addition, warm CRM binders without aged binder were used to be compared with recycled CRM binders. The viscosity properties for the binders in the original state, the rutting properties in the original state and after RTFO aging, the fatigue cracking properties at intermediate temperature after RTFO + PAV aging methods,

and the low temperature cracking properties after RTFO + PAV procedures were evaluated. Figure 27 shows a flow chart of the experimental design used in this study.

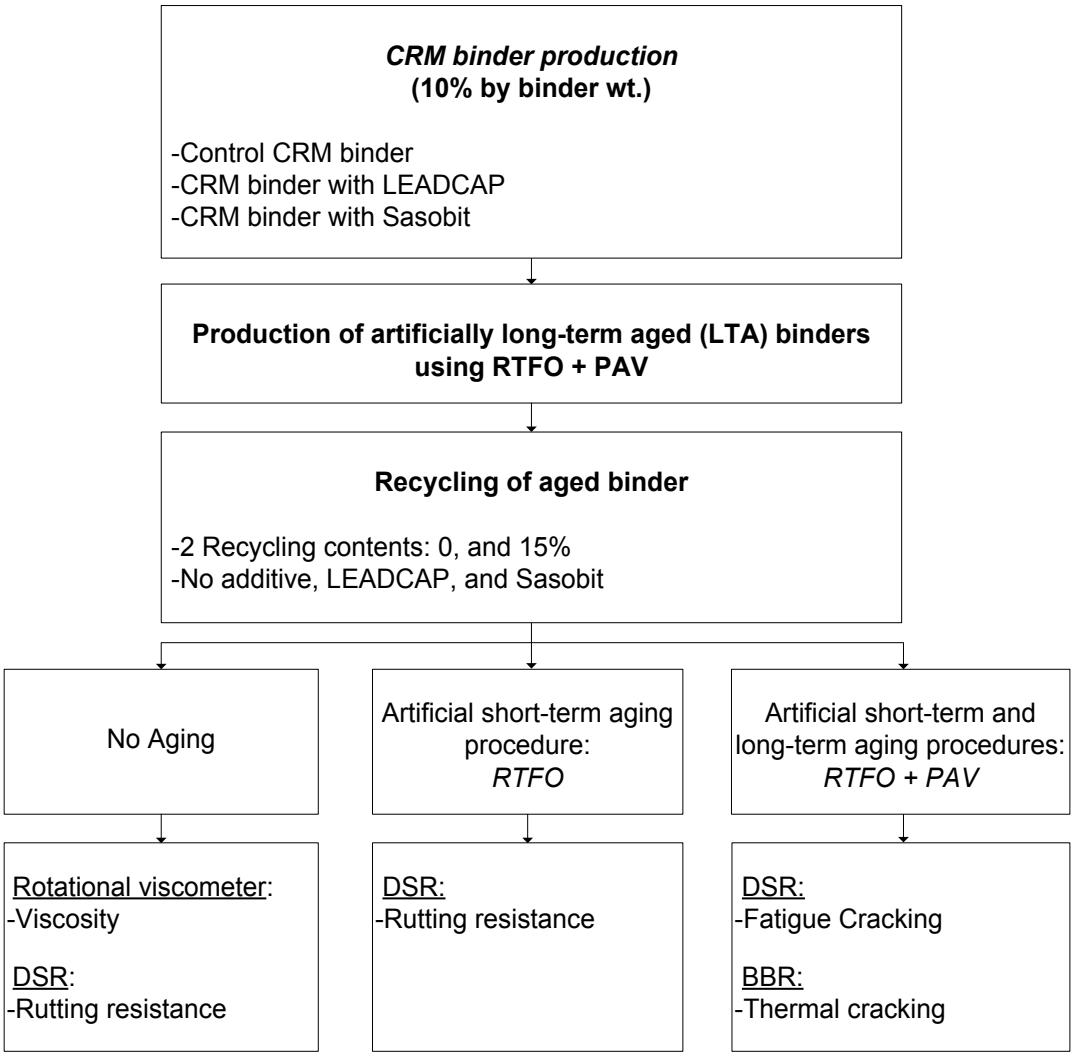


Figure 27. Flow chart of experimental design procedures of the recycling property study.

Experimental Program

Materials

Asphalt binder

One PG 64-22 binder was used as a base binder in this study. The details of these base binders are described in Chapter 4.

Crumb rubber modifier (CRM)

The CRM (-40 mesh (0.425 mm)), produced by mechanical shredding at ambient temperature, was obtained from one source. The details of the CRM are described in Chapter 4.

Wax additives

Two commercial wax additives of LEADCAP and Sasobit were used for this research. The details of the wax additives are described in Chapter 4.

Recycling of aged binders

Virgin CRM binders produced using the base binders of PG 64-22 were used for the recycling of long-term aged (LTA) warm binders. The CRM recycling percentage of 15% was selected because most states in the United States use this percentage in recycling practices, and the control CRM binder was used to be compared with the recycled CRM binder. The binders were then artificially aged through RTFO and PAV processes.

Superpave asphalt binder tests

The properties of these CRM binders were evaluated using selected Superpave binder test procedures including the viscosity test (AASHTO T 316), the DSR test (AASHTO T 315: with the plate gap adjusted to 2 mm), and the BBR test (AASHTO T 313). The details of each test method are described in Chapter 4 and 5.

Results and Discussions

Recycling of CRM binder

Viscosity

Figure 28 shows the experimental values of the viscosities measured by Brookfield viscometer at 135°C. These results clearly demonstrated that the addition of wax additives into CRM binder reduced the binder's viscosity, compared to control binders (i.e., without the additives) at both recycling contents. In addition, the viscosity of recycled CRM binders was increased approximately 45% by the addition of long-term aged (LTA) CRM binders. However, the recycled CRM binders containing wax additives showed lower viscosity than the control recycled CRM binder. The viscosity reduction rate of 13% on average was observed in recycled warm CRM binders, indicating that the wax additives in aged CRM binder are effective to reduce the viscosity of CRM binder, even though the wax additives in aged CRM binder were experienced a series of aging processes (RTFO and PAV). All binders satisfied the maximum limit for viscosity of asphalt binders at 135°C suggested by Superpave (i.e., 3000 cP).

Using one-way analysis of variance, the statistical significance of the change in the viscosity was examined (Table 11). In general, the viscosity values of recycled CRM binders were significantly different depending on the recycling content corresponding to their control virgin binders, as expected. At both recycling content the CRM binders with wax additives were found to be insignificantly different at 5% level in the viscosity, compared within each recycling content.

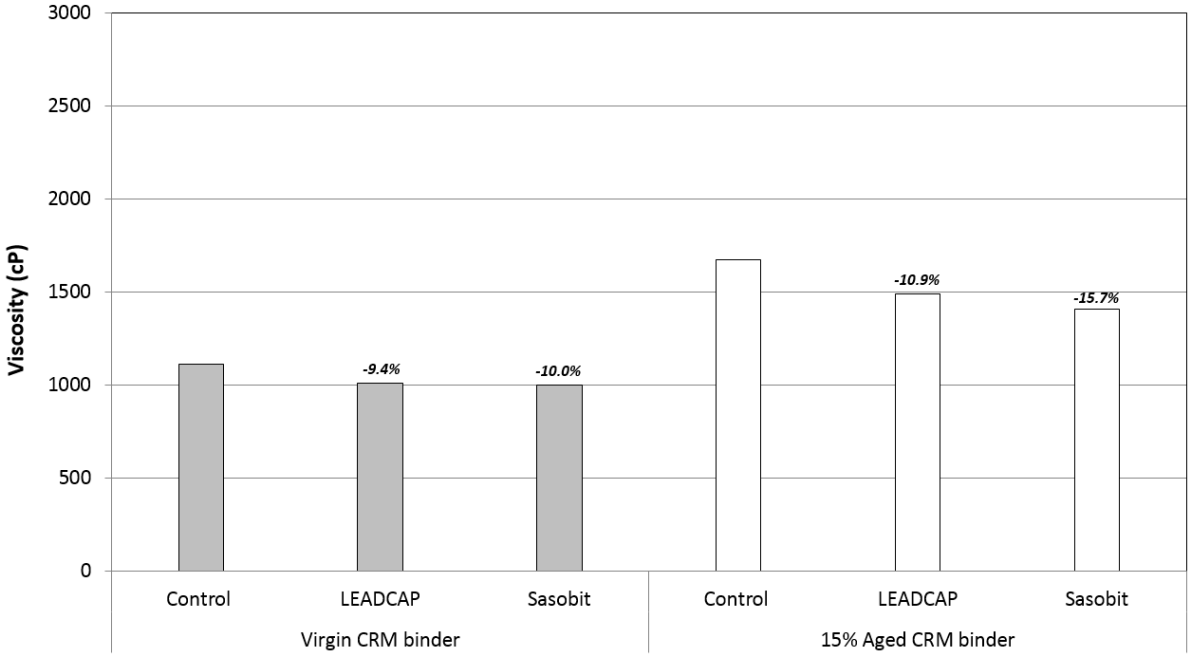


Figure 28. Viscosity of recycled CRM binders at 135°C.

Table 11. Statistical analysis results of the viscosity value as a function of recycling percentage and wax additive (α=0.05).

Viscosity		Virgin CRM binder			15% Aged CRM binder		
		Control	LEADCAP	Sasobit	Control	LEADCAP	Sasobit
Virgin CRM binder	Control	-	S	S	S	S	S
	LEADCAP		-	N	S	S	S
	Sasobit			-	S	S	S
15% Aged CRM binder	Control				-	S	S
	LEADCAP					-	N
	Sasobit						-

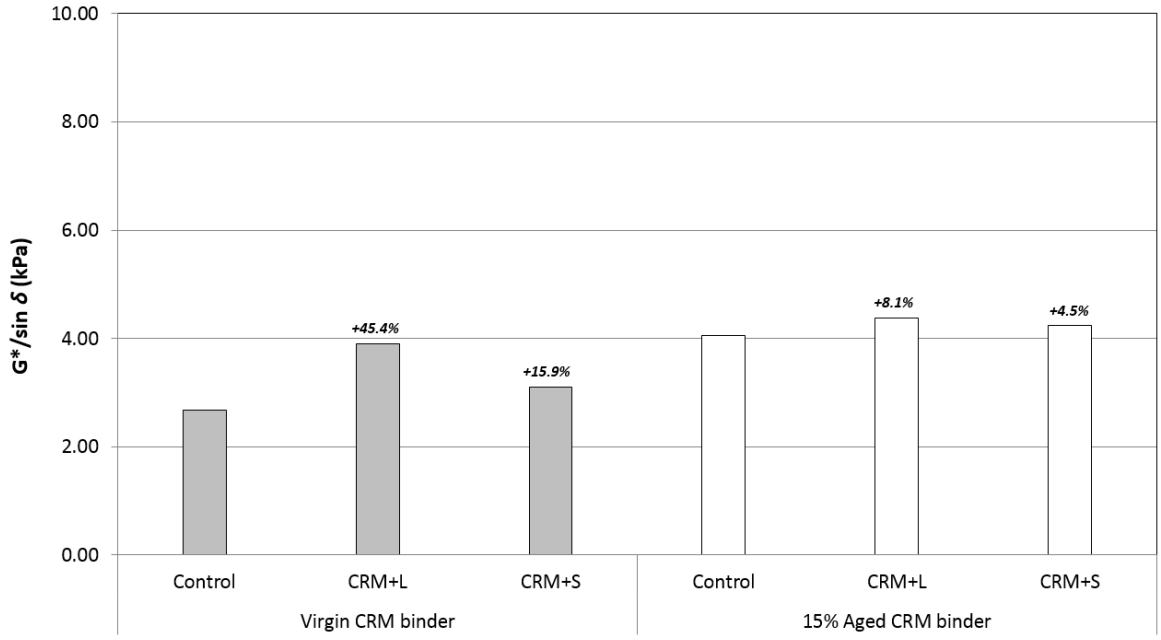
N: non-significant, S: significant.

Rutting property

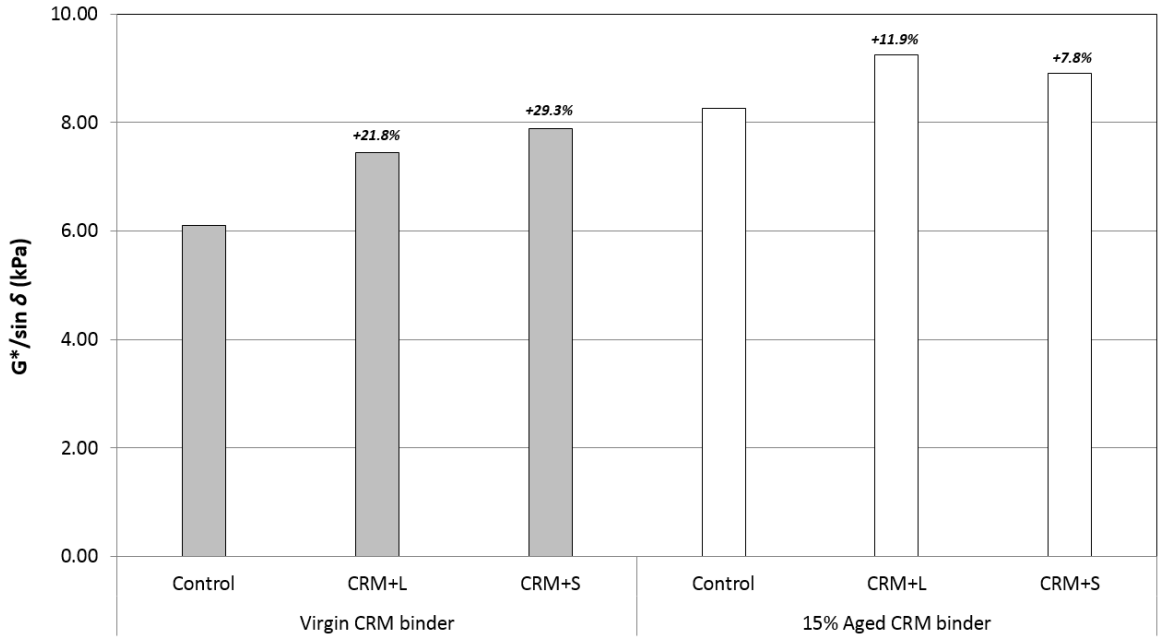
The G*/sin δ values of the recycled CRM binders in original state and short-term aging state were measured and the results are shown in Figure 29. In general, the CRM binders containing the wax additives resulted in higher G*/sin δ than the control CRM binders regardless of aging state, indicating that the addition of wax additives has a positive effect on rutting resistance of the binders. A similar trend was observed for recycled CRM binders. The recycled warm CRM binders showed the increase rate of 6% on average for G*/sin δ value in original state, while the virgin warm CRM binders had the 30% on average. This result is reasonable in terms of substantial amount of wax additive in asphalt binders. The wax content for virgin warm CRM binders was 1.5% by the binder weight (the warm CRM binder of 100g = CRM binder of 98.5g + additive of 1.5g), which is recommended by the manufacturer. The amount of wax additive for recycled warm CRM binders was approximately 0.23% by the binder weight (for example, the 15% recycled warm CRM binder of 100g = virgin CRM binder of 85g + aged warm CRM binder of 15g (the wax additive: 1.5% of 15g, which is 0.23% of 100g)). Therefore, the increase rate of 6% for recycled CRM binder is a rational result considering substantial amount of wax additive in the CRM binder. It means that the wax additives still play an important role in improving the rutting property, even though the additive was short-and-long term aged within asphalt binder.

The statistical significance of the change in the G*/sin δ as a function of recycling content and wax type was examined and the results are shown in Table 12. Generally, the addition of LTA has a significant effect on the G*/sin δ value of the recycled CRM binders.

However, the control CRM binders and warm CRM binders were observed to have an insignificant difference in the rutting resistance at 15% recycling content. In addition, there was no significant difference at $\alpha = 0.05$ level between the $G^*/\sin \delta$ values of warm CRM binders at both recycling content regardless of their aging state.



(a)



(b)

Figure 29. G*/sin δ of recycled CRM binders at 64°C;
(a) Unaged and (b) Short-term aged

Table 12. Statistical analysis results of the G*/sin δ value as a function of recycling percentage and wax additive (α=0.05); (a) Unaged and (b) Short-term aged

(a)

G*/sin δ		Virgin CRM binder			15% Aged CRM binder		
		Control	LEADCAP	Sasobit	Control	LEADCAP	Sasobit
Virgin CRM binder	Control	-	S	N	S	S	S
	LEADCAP		-	N	N	N	N
	Sasobit			-	S	S	S
15% Aged CRM binder	Control				-	N	N
	LEADCAP					-	N
	Sasobit						-

(b)

G*/sin δ		Virgin CRM binder			15% Aged CRM binder		
		Control	LEADCAP	Sasobit	Control	LEADCAP	Sasobit
Virgin CRM binder	Control	-	N	S	S	S	S
	LEADCAP		-	N	N	S	S
	Sasobit			-	N	N	N
15% Aged CRM binder	Control				-	N	N
	LEADCAP					-	N
	Sasobit						-

N: non-significant, S: significant.

Fatigue cracking property

After RTFO and PAV procedures, the G*sin δ values of the recycled CRM binders (RTFO + PAV residual) were measured using DSR at 25°C and the results are shown in Figure 30. A general trend was found from the results that the addition of Sasobit into CRM binder increased the binder's G*sin δ at 25°C by 32.8%, compared to the control CRM binder. However, the addition of LEADCAP resulted in reducing the G*sin δ value significantly by 20.6%. The result indicated that the addition of LEADCAP was effective in improving the fatigue cracking resistance of the control CRM binders. On the other hand, the wax additives in recycled CRM binder were found to have similar trend with virgin CRM binders. The addition of LTA binders into the virgin CRM binders can significantly increase the G*sin δ by approximately 50%. The G*sin δ value of the recycled CRM binder with Sasobit was the highest and that of the recycled CRM binder with LEADCAP was the lowest. At 15% recycling content (substantial amount of LEADCAP is 0.23% by the binder weight), the decreasing rate of G*sin δ is 13.2 % whereas at 0% recycling content (substantial amount of LEADCAP is 1.5% by the binder weight) the decreasing rate is 20.6%. It suggests that the behavior of wax additives into the recycled CRM binders has a significant effect based upon their substantial amount.

Using one-way analysis of variance, the statistical significance of the change in the G*sin δ values was examined (Table 13). The addition of LTA into the virgin CRM binder seemed to have statistical difference with virgin CRM binder. Overall, there was significant difference between control CRM binder and both warm CRM binders within each recycling content. Also, the differences between the binders containing LEADCAP and the

binders containing Sasobit were statistically significant at all recycling contents used in this study.

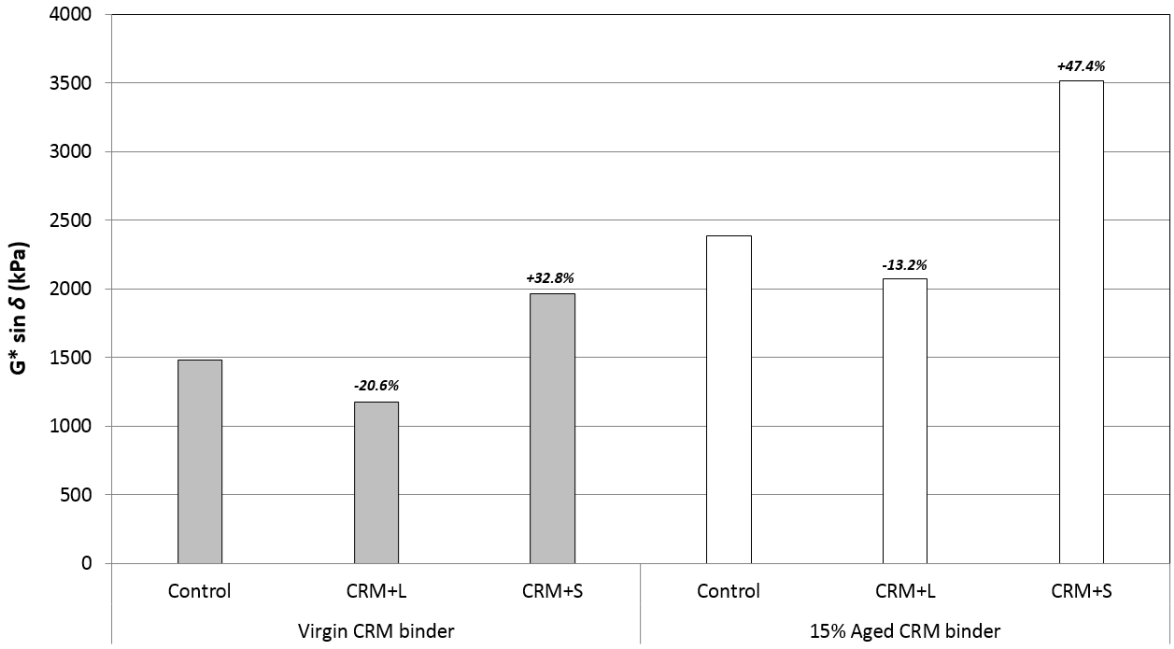


Figure 30. G*sin δ of recycled CRM binders (RTFO + PAV) at 25°C.

Table 13. Statistical analysis results of the G*sin δ value as a function of recycling percentage and wax additive (α=0.05).

G*sin δ		Virgin CRM binder			15% Aged CRM binder		
		Control	LEADCAP	Sasobit	Control	LEADCAP	Sasobit
Virgin CRM binder	Control	-	S	S	S	S	S
	LEADCAP		-	S	S	S	S
	Sasobit			-	S	S	S
15% Aged CRM binder	Control				-	N	S
	LEADCAP					-	S
	Sasobit						-

N: non-significant, S: significant.

Low temperature cracking property

From the BBR tests at -12°C, the stiffness was calculated, and the results are depicted in Figure 31. The stiffness values from the BBR tests showed a similar trend to the DSR values at 25°C. The recycled CRM binder containing Sasobit yielded the highest stiffness, suggesting that the addition of Sasobit may result in the binder being less resistant to low temperature cracking. This finding is consistent to the previous studies (Butz et al. 2001; Edwards and Redelius 2003; Edwards et al. 2006).

On the other hand, the stiffness values of binders containing LEADCAP were reduced by 2.8% and 2.2% at 0% and 15% recycling contents, respectively. Although the substantial amount of LEADCAP at 15% recycling content (0.23% by binder weight) is less compared to 0% recycling content (1.5% by binder weight), no significant difference is observed in terms of decreasing rate. However, the use of LEADCAP as a warm additive provides a positive influence on low-temperature cracking resistance, even in recycled CRM binder. In addition, Sasobit has less substantial amount (0.23% by binder weight) at 15% recycling content compared to 0% recycling content (1.5% by binder weight), the increasing rate of stiffness is higher at 15% recycling content. It indicates that the wax additives have a vital role at the stiffness of recycled CRM binders. Although the addition of long-term aged (LTA) binder increased the stiffness value in recycled CRM binder, all the binders satisfied the requirement set forth by Superpave (maximum 300 MPa).

The statistical results of the change in the stiffness are shown in Table 14. The type of wax was found to have a significant effect on the stiffness of the warm CRM asphalt binders at both recycling contents. The recycling content does not have any significant

effect between each recycled CRM binder corresponding to their virgin CRM binder. In addition, the difference of stiffness values between control CRM binder and CRM binder with LEADCAP was not significant at the 5% level within each recycling content.

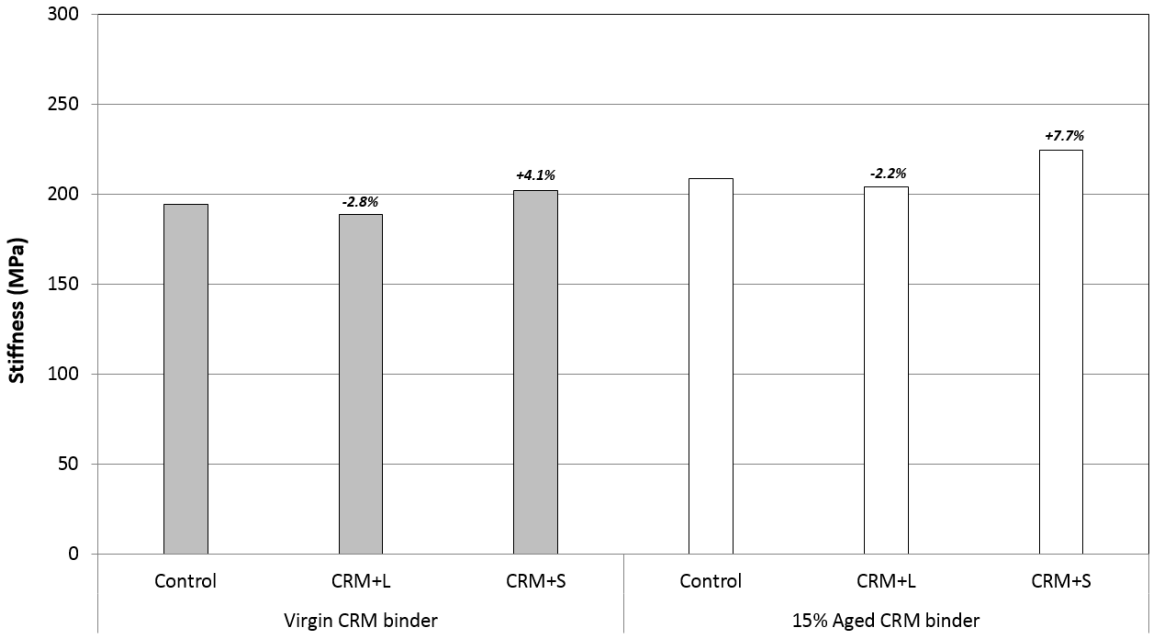


Figure 31. Stiffness of recycled CRM binders (RTFO + PAV residual) at -12°C.

Table 14. Statistical analysis results of the stiffness values as a function of recycling percentage and wax additive (α=0.05).

Stiffness		Virgin CRM binder			15% Aged CRM binder		
		Control	LEADCAP	Sasobit	Control	LEADCAP	Sasobit
Virgin CRM binder	Control	-	N	S	N	N	S
	LEADCAP		-	S	S	N	S
	Sasobit			-	N	N	N
15% Aged CRM binder	Control				-	N	N
	LEADCAP					-	S
	Sasobit						-

N: non-significant, S: significant.

Summary and Conclusions

Two types of wax additives were mixed with CRM binder to investigate the effect of the recycled aged CRM binders on performance properties. The CRM binders were produced in the laboratory using one CRM source (ambient), one CRM percentage (10% by the binder weight), and mixed with two wax additives. The CRM binders were artificially aged through accelerated aging processes. The aged CRM binders were recycled at 15% long-term aged (LTA) binder percentage by the total binder weight, and then artificially aged using the RTFO and PAV processes. A series of Superpave binder tests were carried out using the rotational viscometer, the DSR, and the BBR to evaluate the properties of the recycled CRM binders containing wax additives. The virgin CRM binders with wax additive were used to be compared with recycled CRM binders. From these test results, the following conclusions were drawn for the materials used in this study.

1) The binders containing wax additives showed significantly lower viscosity at 135°C, though the addition of LTA increased the binder viscosity. Also, the recycled CRM binder showed similar trend, indicating that the wax additives in recycled CRM binder are effective to reduce the viscosity.
2) Generally, the wax additives seemed to lead to better rutting properties for CRM binders. This same trend was observed in recycled warm CRM binders. The result shows that the wax additive has a significant influence to improve the rutting property in recycled CRM binder.
3) According to the G*sin δ and stiffness values, the addition of LTA has negative effects on cracking performance of recycled CRM binder. When using recycled

CRM binder at cold regions, the recycling content needs to be carefully decided considering cracking resistance.

4) The wax types were found to have a significant effect on the cracking properties of the warm CRM binders.
5) In general, the effects of wax additives for CRM binder were clearly observed in control CRM binders (recycling content of 0%). This trend was consistent in recycled warm CRM binders. The wax additive in recycled CRM binder plays an important role in binder properties, even though the additive was subjected to short and long-term aged.
6) The wax warm additives are expected to have significant effect in performance properties of recycled CRM binders based upon their substantial amount. Also, it is recommended to conduct a field study to generalize these findings with the field behavior.

This chapter (V) includes a part of the following publications;

Kim, H. H., Mazumder, M., Lee, S. J., "Characterization of recycled CRM binders containing wax warm additives," *International Journal of Pavement Research and Technology* (Submitted)

VII. MICROSTRUCTURAL PROPERTIES

Introduction

In order to understand the interaction between the rubber particles and the bitumen in CRM binder, it requires the understanding of the microstructural properties of such mechanical behavior. Development of modern technology has allowed to observe microstructural property using several equipment like optical microscopy, scanning electron microscopy (SEM), and atomic force microscopy (AFM).

Direct observation of the change in the binder morphology due to modified binder interaction is quite difficult. Several studies (Loeber et al., 1996; Jäger et al., 2004; Masson et al., 2006, 2007; Pauli et al., 2001; Allen et al., 2012) examine the asphalt at shorter length scales using an atomic force microscopy (AFM), which has revealed heterogeneous domains in asphalt with potentially different mechanical properties. First microstructural investigation of asphalt binder using AFM was conducted by Loeber et al. (1996). They used AFM for characterizing the natural surface of the asphalt binder without any disturbance. The research found that specifically rippled features on the microstructure which is called “bee” structure. This bee shape is considered as the change in topography on the asphalt binder surface. They described that the bee structure is related to the resin and asphaltene molecules. According to the study performed by Jäger et al. (2004), the bee shape is asphaltene and possibly resin which have a high polarity by separating experiments using the n-heptane. In addition, it was found that the bee structures are caused by the

presence of asphaltenes in the binder (Zhang et al., 2011). Oldham (2015) reported that the bee shapes increase in maximum length with aging process compared to the unaged. In general, the cracking property of asphalt binder shows different performances depending on hard part like asphaltene. Therefore, it is considered that the surface images of asphalt binder are able to illustrate the cracking properties depending on wax crystallization and aging level.

Of all the microscopic techniques, the environmental scanning electron microscope (ESEM) which has little different to conventional SEM technique is considered as one of the best techniques for oil bearing materials like asphalt. The primary advantage of the ESEM over conventional SEM is that the ESEM does not require the test sample to be under high vacuum. Thus, wet, oily, dirty, or nonconductive samples can be examined in their natural state without modification or preparation (Kimseng et al. 2001). At first Rozeveld et al. (1997) performed the ESEM techniques to understand the internal structure of unmodified and polymer-modified binders. They reported that ESEM can help to characterize the morphology of the network structure of asphaltenes and resins. Later Williams et al. (1998) used ESEM to understand the asphalt water interactions in terms of water stripping and water penetration in asphalts. They concluded that ESEM has potential applications to study the microscopic properties of asphalt binder and can be used as a practical method for studying roadway failure mechanisms. Although ESEM introduced very early, limited studies have been performed using ESEM techniques to understand the morphology of the asphalt binder with CRM (Wang and Wei, 2013; Divya et al. 2013; Yu et al. 2013).

The main objective of this study is to characterize the microstructural property of CRM binder with wax additives using AFM and to investigate the difference between the microstructural properties of unaged CRM binder and aged CRM binder using ESEM. Furthermore, to understand the chemical composition of the surface morphology obtained from ESEM, EDX was carried out on the aged CRM binder. Also, the feasibility of this two techniques to characterize the internal structure of the binder need to be investigated for future use. Figure 32 shows a flow chart of the experimental design used in this study.

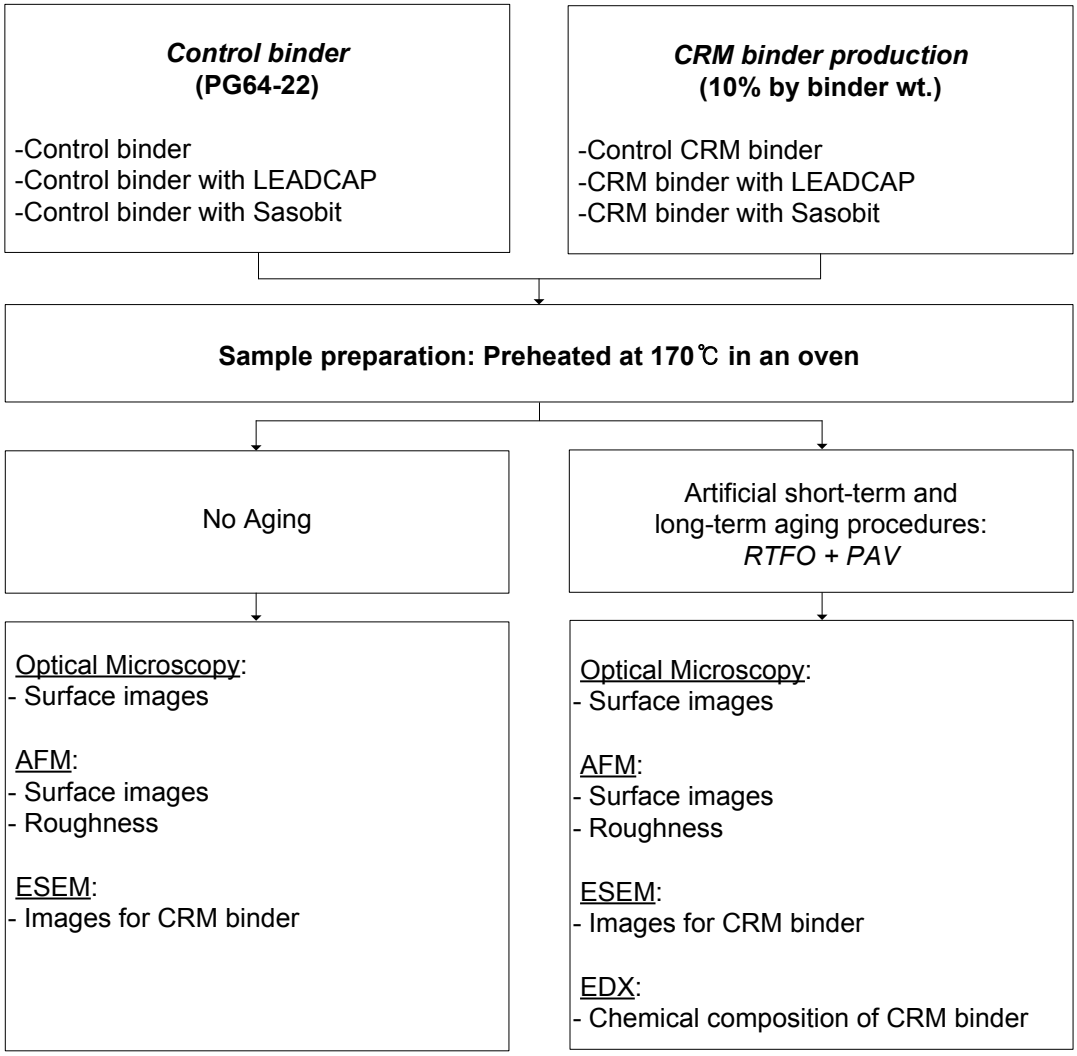


Figure 32. Flow chart of experimental design procedures of the microstructural property study.

Experimental Program

Materials

Asphalt binder

One PG 64-22 binder was used as a base binder in this study. The details of these base binders are described in Chapter 4.

Crumb rubber modifier (CRM)

The CRM (-40 mesh (0.425 mm)), produced by mechanical shredding at ambient temperature, was obtained from one source. The details of the CRM are described in Chapter 4.

Wax additives

Two commercial wax additives of LEADCAP and Sasobit were used for this research. The details of the wax additives are described in Chapter 4.

Methods

Sample preparation

In this study, the sample preparation is one of the most important task to investigate the morphology of CRM binders. A sample was prepared by pouring melted binder on the surface of a heated mold. All binders were preconditioned by controlled heating at 170°C in an oven. The binder was flowed smoothly into the circular mold which has 2 inch diameter and the mold was placed on heating plate to produce a well-dispersed sample.

The samples were examined immediately after they cooled down. At least three different regions of the surface were scanned. Figure 33 shows the sample used in this study.

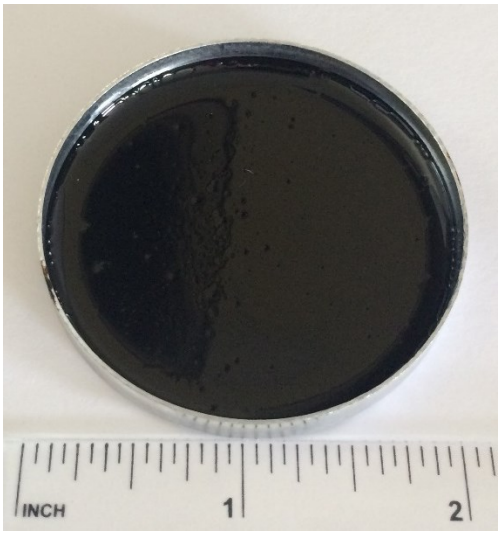


Figure 33. Asphalt binder sample for morphology investigation.

Optical Microscopy

The sample morphology was observed using an optical microscope (VHX-2000) made by KEYENCE. Binder sample with mold specimen was prepared using small amounts of the asphalt binder containing wax additives and viewed under the microscope at a magnification of 300. The optical microscopy is shown in Figure 34.

Figure 34. VHX-2000 (KEYENCE).

Atomic Force Microscopy (AFM)

A Dimension 3100 AFM (Veeco Instrument Inc.) was used to characterize the cracking properties of CRM binder with wax additives through the nanostructure images on prepared sample surfaces. Topographical images and phase images were captured. Figure 35 shows the AFM equipment used in this study.

AFMs can be used to measure the forces between the tip and the sample as a function of their mutual separation. The AFM tapping mode imaging was performed on the asphalt samples to evaluate the cracking characters on the nanostructure of the CRM binder containing wax additives. In the tapping mode, the AFM tip is oscillated at its resonance frequency by a piezoelectric element connected to the tip holder assembly. The piezo-drive is adjusted using feedback control to maintain a constant tip-up-sample distance (set point) (Bhushan and Qi 2003).

In this paper, both height and phase images were obtained with the scan rate of 0.498Hz and the scan size of 20μm × 20μm. Height images represent the topography of the surface. Phase images are displayed for the unambiguous resolution which can be hindered by surface roughness in topographic image captured by the height mode in AFM. The colors in the phase images designate different mechanical properties of the phases, as obtained from sample–tip interactions, and could be related to their different viscoelastic properties or adhesion. The colors of the phases are sometimes inverted in two different scans of the same spot upon the change of AFM parameters, and therefore color inversion does not imply inversion of material properties.

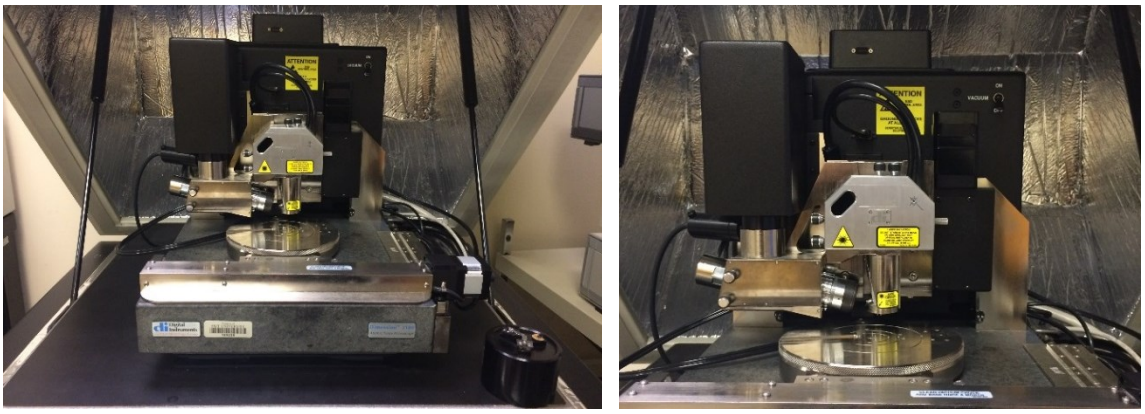


Figure 35. Dimension 3100 AFM (Veeco Instrument Inc.).

Environmental Scanning Electron Microscope (ESEM)

The ESEM used in this study is manufactured by JEOL (Model #: JSM-6010PLUS/LA) to examine the surface microstructure of unaged and aged CRM. The degree of magnification was chosen to be 200, 400, 500, 800, 1500 and 2000. The scan sizes used were 10, 20, 50, and 100 µm. The equipment settings used for scanning were as follows, 5-10kV; pressure, 40Pa. Figure 36 shows the JEOL SEM used in this study.

Figure 36. JEOL SEM.

Energy Dispersive X-ray Spectroscopy (EDX)

EDX is normally used to find the chemical composition of the materials. It can create the elemental composition maps. It used a software called EDX-Genesis to gather and analyze the energy spectra. The software works to find out the peaks based on the gathered elemental spectrum and makes it a comprehensive survey tools to do the quantitative analysis of the chemical compositions.

Results and Discussions

Surface morphology

Optical microscopy images

The surface images of control binder with wax additives at original state are shown in Figure 37 (a) ~ (c). The surface image of the control binder shows dark dot spots homogeneously spread on the surface. The addition of LEADCAP in the control binder exhibited different pattern which looks like the wax molecules elongated. The surface surrounding the elongated spots showed relatively smoother exterior compared to the control binder. Meanwhile, there is some specific pattern which has been observed on the surface image of control binder with Sasobit. Some dark spots are found to follow a specific line and this shape is surrounded by other light dot spots.

Figure 37 (d) ~ (f) illustrates the images of CRM binder with wax additives. Control CRM binder also shows many dark spots like control binder, while the size of dot is smaller and less bright. It is due to the interaction of rubber particles with the wax molecules in the base binder. These light dark spots are the rubber particles dispersed in the control binder (Zhang et al. 2015). On the other hand, the CRM binder containing LEADCAP exhibited much flat surface compared to the control binder with LEADCAP. In addition, the inclusion of Sasobit in the CRM binder showed relatively smoother surface without the specific patterns which are found in control binder containing Sasobit.

Apart from that the images captured on the artificially long-term aged binders using optical microscopy are shown in Figure 38. In Figure 38 (a), the surface morphology of the aged control binder exhibited denser dark spot compared to the unaged control binder. It

can be explained that this formation appeared due to the aging process. In contrast, the elongated shapes of the control binder with LEADCAP became light after aging (Figure 38 (b)).

The specific pattern that observed in control binder with Sasobit appeared much clear after aging in Figure 38 (c). In addition, it is worth to note that the size of the shape is increased after aging. Figure 38 (d) ~ (f) show the optical images of CRM binder with wax additives after aging. Less dense dark spots are observed on the surface of CRM and CRM+L after aging compared to the samples at original state. As expected, the aged CRM+S has the same shape which were observed in unaged CRM+S with more clear and increased dimension.

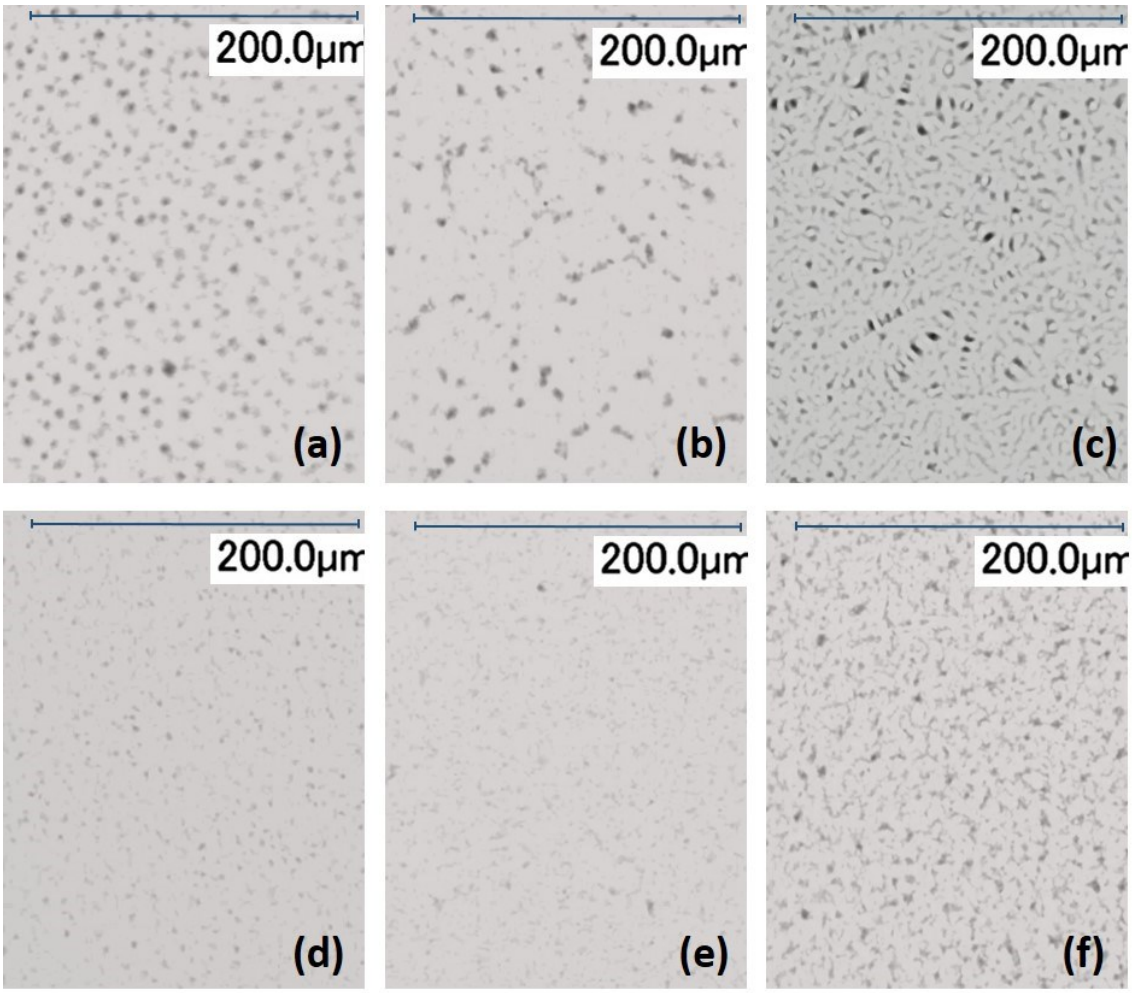


Figure 37. Optical microscopy images of control binder
with wax additives (Original state);
(a) Control (b) Control+LEADCAP (c) Control+Sasobit
(d) CRM (e) CRM+LEADCAP (f) CRM+Sasobit

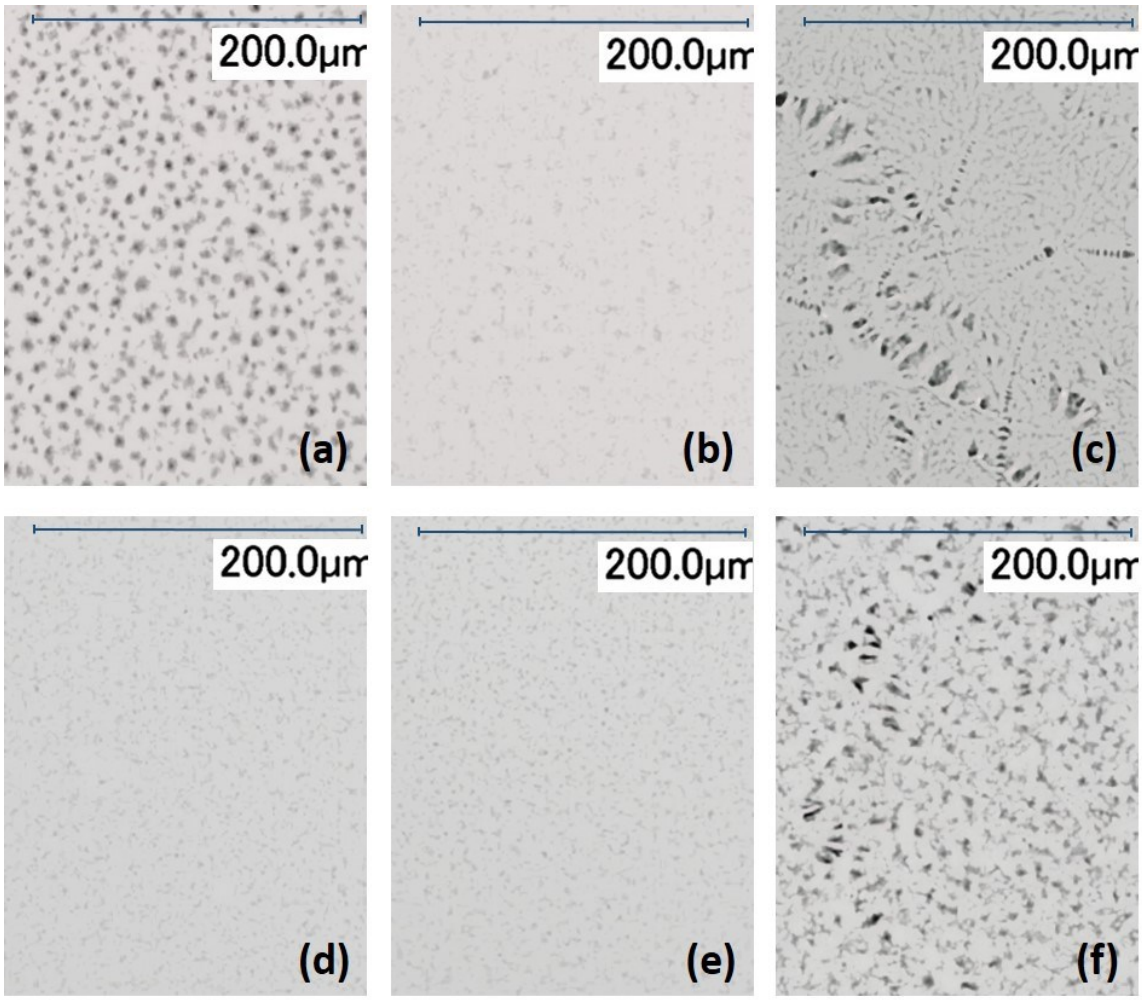


Figure 38. Optical microscopy images of CRM binder
with wax additives (after long-term aging);
(a) Control (b) Control+LEADCAP (c) Control+Sasobit
(d) CRM (e) CRM+LEADCAP (f) CRM+Sasobit

AFM images

Figure 39 shows the topographic images and corresponding phase images of unaged PG 64-22 with wax additives. It is found from the topographic image of unaged PG 64-22 that it consists of some microstructure which is a sequence of hills and valleys or a succession of pale and dark lines. In general, the phase image indicates that it consists of four different phases which are catana phase (bee-like structures), peri phase (around the bee shape), and para or perpetua phase (solvent regions). The catana and peri phase constitute the dispersed domain whereas the para phase constitutes the matrix. This 'bee-like' structures are due to the presence of microcrystalline wax and waxy molecules in the binder.

Figure 39 (c) shows the topographic feature of the control binder containing LEADCAP. In topographic image, no bee shape is found for Control+L except some brilliant points. It is due to the higher colloidal index of the binder. When LEADCAP is blended with control binder, the disperse domains or the microcrystalline waxes and waxy molecules are dissolved easily by the other components of the binder which contributes the disappearance of 'bee-like' structures. The presence of brilliant dots with two different color contrast can be clearly observed in the phase image of Control+L (Figure 39 (d)). AFM images in Figures 39 (e) and (f) show that the addition of Sasobit in the control binder significantly affect the size and width of the 'bee like' structures. The 'bee-like' structures is appeared much larger in dimension (Nazzal & Qtaish 2013; Das et al. 2013; Menapace et al. 2014; Qin et al. 2014; Menapace et al. 2015) and the width get reduced compared to the control binder (Nazzal et al. 2015). In optical microscopy, the image of control binder

containing Sasobit where some dark spots are found to follow a specific line actually resembles the ‘bee-like’ structures. It indicates that the addition of Sasobit increase the dimension of ‘bee-like’ structures which is under the resolution limit of optical microscopy.

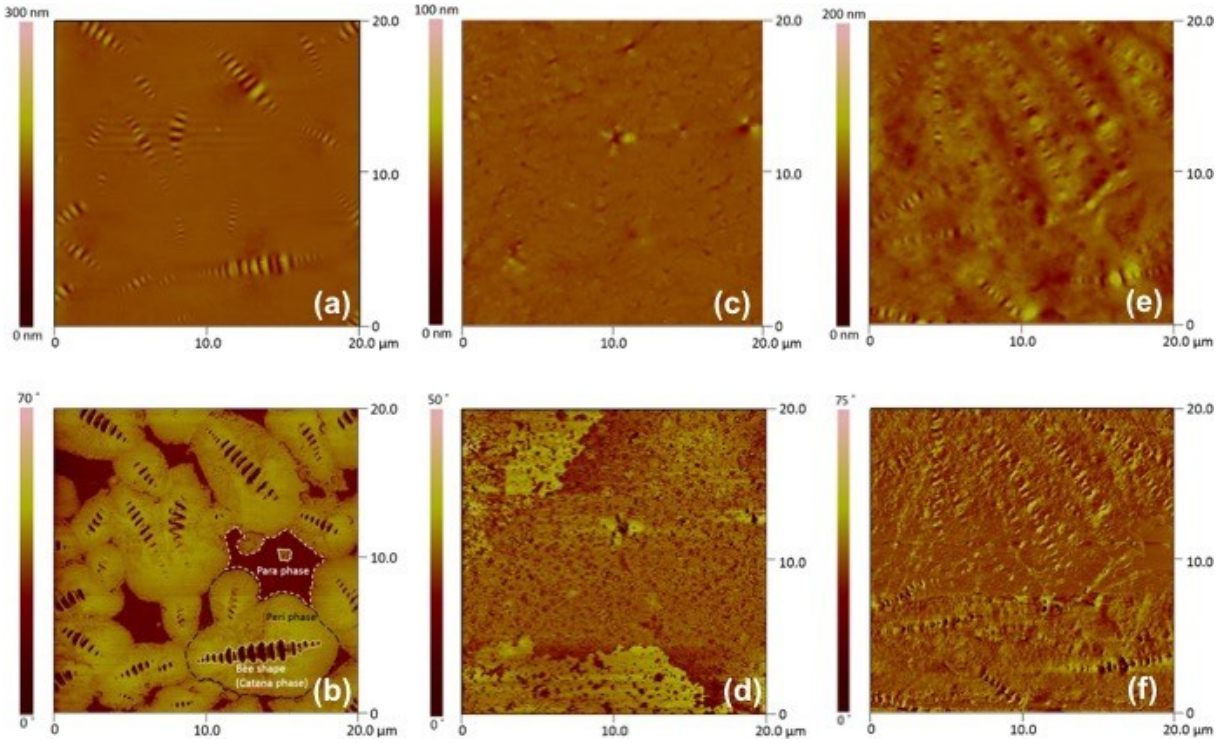


Figure 39. AFM images of control binder with wax additives at original state;
(a) Topographical image of Control (b) Phase image of Control
(c) Topographical image of Control+L (d) Phase image of Control+L
(e) Topographical image of Control+S (f) Phase image of Control+S

Figure 40 illustrates the topographic and phase images obtained on the unaged CRM binder and the CRM binder containing LEADCAP and Sasobit at scan size of 20μm after 24 hours. The topographic and phase image of CRM binder are shown in Figure 40 (a) and (b). It can be clearly observed that the number of 'bee-like' structures are increased compared to the control binder. One interesting observation is that some small spherical bright dot spot has been found in addition to the 'bee-like' structures on the topographic image of CRM binder. In previous studies, this small particulate matter was referred to as some fractions of the rubber particles which may be dissolved in the binder due to the de-vulcanization of the rubber (Huang and Pauli 2008). After the inclusion of crumb rubber in the control binder, the contrast between the dispersed domains and the matrix is decreased, meaning that the spreading of dispersed domains is less which can be seen in a comparison of Figure 39 (b) and Figure 40 (b). The dimension of the 'bee-like' structures as well as the dispersed domains get reduced compared to the control binder. It may be due to the swelling of the rubber particles. As a result, the dispersed domains can be dissolved easily in the CRM binder compared to the control binder.

Topographic and phase images captured on the CRM binder mixed with LEADCAP are presented in Figure 40 (c) and (d). Like the Control+L no bee shape has been observed on the topographic image of CRM+L. Some particulate matter has been found on the surface image of CRM+L. As mentioned earlier this particle is referred as the dissolved rubber particles in the binder. Compared to the phase image of Control+L, surface of the phase image of CRM+L is found to have more smooth. Figure 40 (e) shows the topographic image of CRM binder with Sasobit. Unlike the Control+S, the size of 'bee-

like' structures are small and few particulate matter has been found on the surface topography of CRM+S. The detail feature of the dispersed domains and the matrix has been clearly observed in the phase image of CRM+S, Figure 40 (f). Although the 'bee-like' structures are found small compared to Control+S, those are bigger than the CRM binder. The peri phase get expanded due to the addition of Sasobit in CRM binder. The contrast between the dispersed domains and the matrix is increased, meaning that the amount of dispersed domains is higher in the CRM binder with Sasobit compared to the control and CRM binder. However, the control binder containing Sasobit seems to have much larger clustering 'bee-like' structures as well as the dispersed domains compared to the CRM+S which can be observed in a comparison of Figure 39 (e) and Figure 40 (e).

Stiffness properties of the binders in terms of observed micromorphology can be described. The addition of crumb rubber in the control binder decreases the dispersed domains of the CRM binder which means it has lower stiffness compared to the control binder. The CRM binder with LEADCAP does not have any 'bee-like' structures, suggesting that it has less stiffness than CRM binder. The higher amount of dispersed domains in the phase image of CRM binder containing Sasobit indicates that the binder has higher stiffness compared to the other CRM binders.

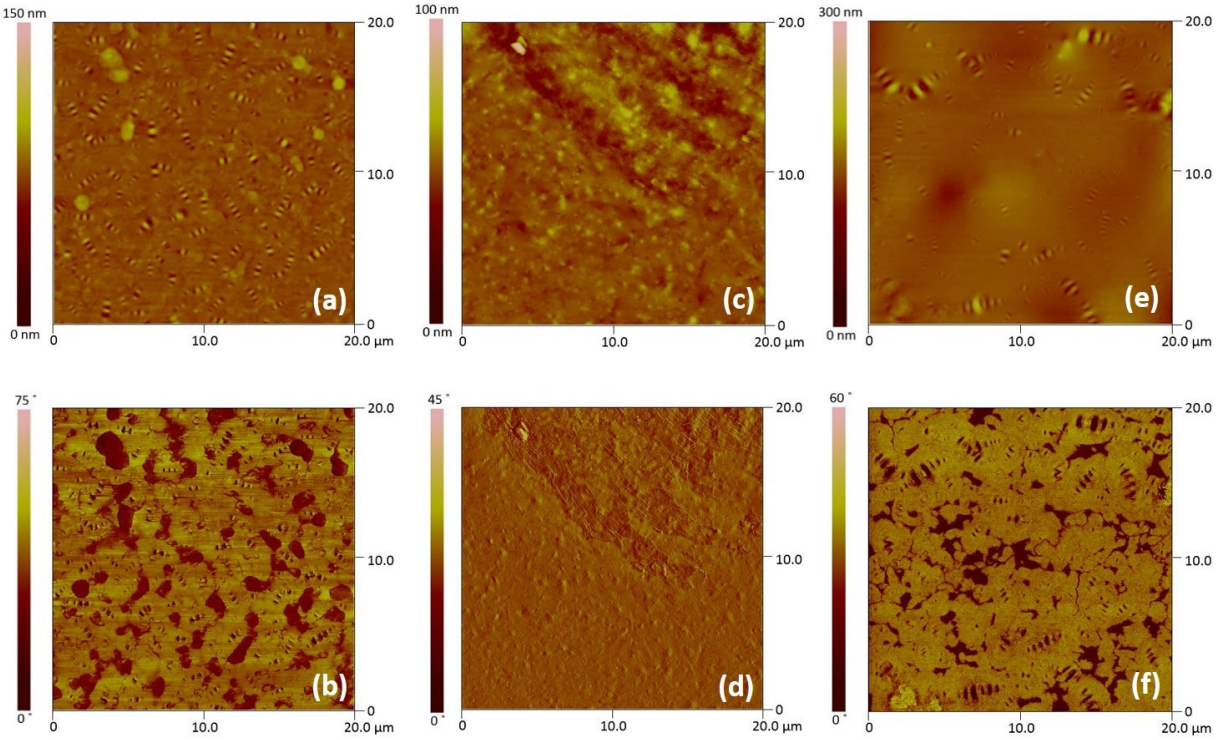


Figure 40. AFM images of CRM binder with wax additives at original state;
(a) Topographical image of CRM (b) Phase image of CRM
(c) Topographical image of CRM+L (d) Phase image of CRM+L
(e) Topographical image of CRM+S (f) Phase image of CRM+S

The binder samples were scanned with AFM after long-term aging. Topography and phase images of PG 64-22 and the same binder mixed with two wax additives are shown in Figure 41. The size of the 'bee-like' structures increased on the topography image of the aged control binder compared to the unaged binder. Compared between the phase image of unaged (Figure 39 (b)) and aged control binder (Figure 41 (b)), it can be observed that the peri phase expanded after aging and the boundary of the dispersed domains are indented. It is also worth to note that the phase color contrast of dispersed domains and the matrix is different in aged control binder compared to the unaged binder. Although the size of the 'bee-like' structures increased, the number is found to have reduced in the aged control binder. It can be explained that due to the oxidation process chemical reactions occurred and molecular structures of the binder changed.

Figure 41 (c) and (d) shows the topographic and phase image of the control binder containing LEADCAP after aging. Although no 'bee-like' structures have been observed for the aged Control+L, more smooth surface topography is found compared to the unaged Control+L. Figure 41 (e) and (f) presents the topographic and phase image of the aged control binder containing Sasobit. After aging of Control+S, the size and width of the 'bee-like' structures increased compared to the unaged Control+S. However, the number of 'bee-like' structures seems to be decreased which can be found in a comparison of Figure 39 (f) and 41 (f).

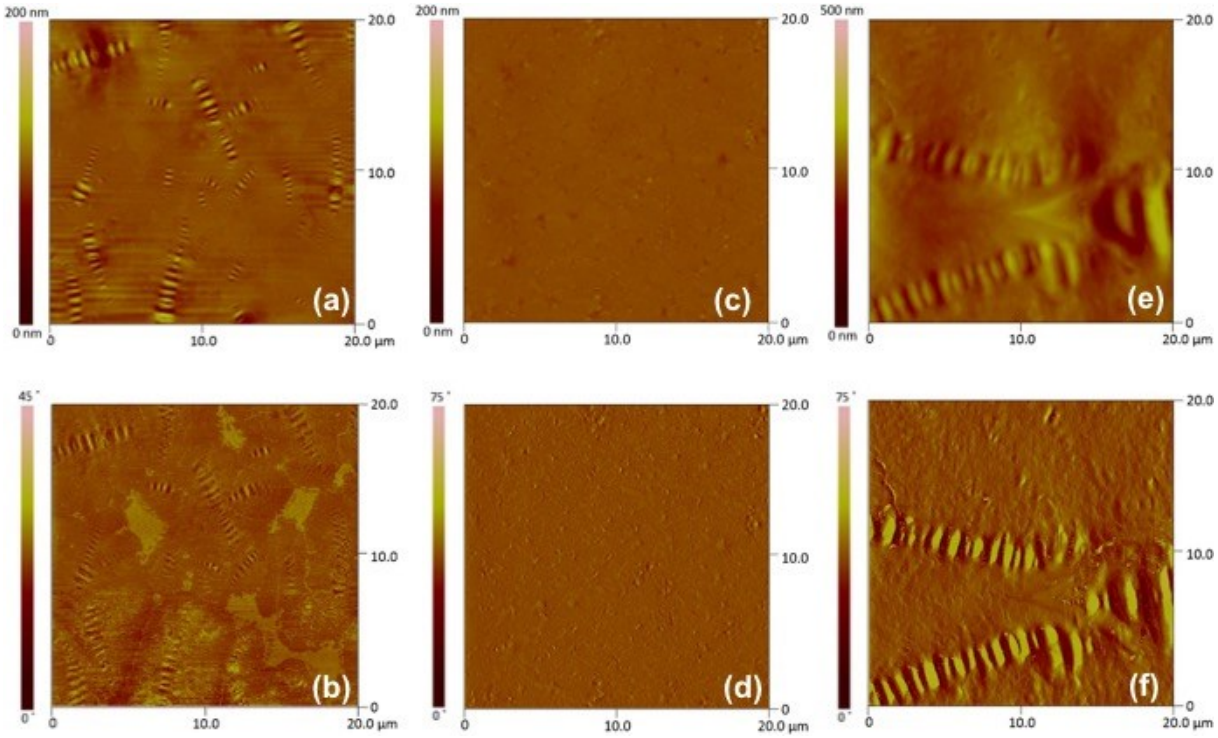


Figure 41. AFM images of PG64-22 binder after long-term aging (RTFO+PAV residuals);
(a) Topographical image of Control (b) Phase image of Control
(c) Topographical image of Control+L (d) Phase image of Control+L
(e) Topographical image of Control+S (f) Phase image of Control+S

Figure 42 represents the topography and phase images captured on the aged CRM binder and the CRM binder containing LEADCAP and Sasobit at scan size of 20μm after 24 hours. It can be clearly observed from the topography image (Figure 42 (a)) of aged CRM binder that the size of 'bee-like' structures reduced compared to the unaged CRM binder. This trend is opposite to the mechanism observed earlier between the unaged and aged control binder. However, the reduction in number of 'bee-like' structures is in agreement with the earlier observation. One interesting observation is that the small particulate matter of rubber particles still remained in the CRM binder after aging. Figure 42 (b) shows the phase image of aged CRM binder. Although the number of 'bee-like' structures decreased, peri phase expanded after aging. That means the dispersed domains occupied a bigger area compared to the para phase. It can be explained that due to aging the molecular properties of the waxes changed and the crystallization obstructed at some extent resulting the appearance of small 'bee-like' structures. A close observation on the amount of dispersed domains between the aged and unaged CRM binders in Figure 42 (b) and Figure 40 (b) indicates that the aged CRM binder has denser distribution of dispersed domains. Based on the phase image analyzed (amount of disperse domains) of two corresponding binders (aged and unaged CRM), it can be derived that the aged CRM binder has high stiffness after aging.

Topographic and phase images of aged CRM binder with LEADCAP are shown in Figure 42 (c) and (d). Similar trend has been found for the aged CRM+L compared to the aged Control+L, meaning the single phase trend without any 'bee-like' structures. The aged CRM binder with LEADCAP has the lower stiffness compared to aged CRM binder due

to the absence of ‘bee-like’ structures. Although some rubber particles are observed on the surface topography of aged CRM+L, those are not bright as unaged CRM+L. It indicates that the aging has significant effect on those small particulate matter. Topographic and phase images of aged CRM binder containing Sasobit are illustrated in Figure 42 (e) and (f). After aging the size and width of the of the ‘bee-like’ structures of the aged CRM+S increased compared to the unaged CRM+S which can be observed in a comparison of Figure 42 (f) and Figure 40 (f). It suggests that the aging process increase the dimension of ‘bee-like’ structures of the CRM binder containing Sasobit. However, it is difficult to estimate the effect of aging on the amount of peri phase between the unaged and aged CRM+S binders.

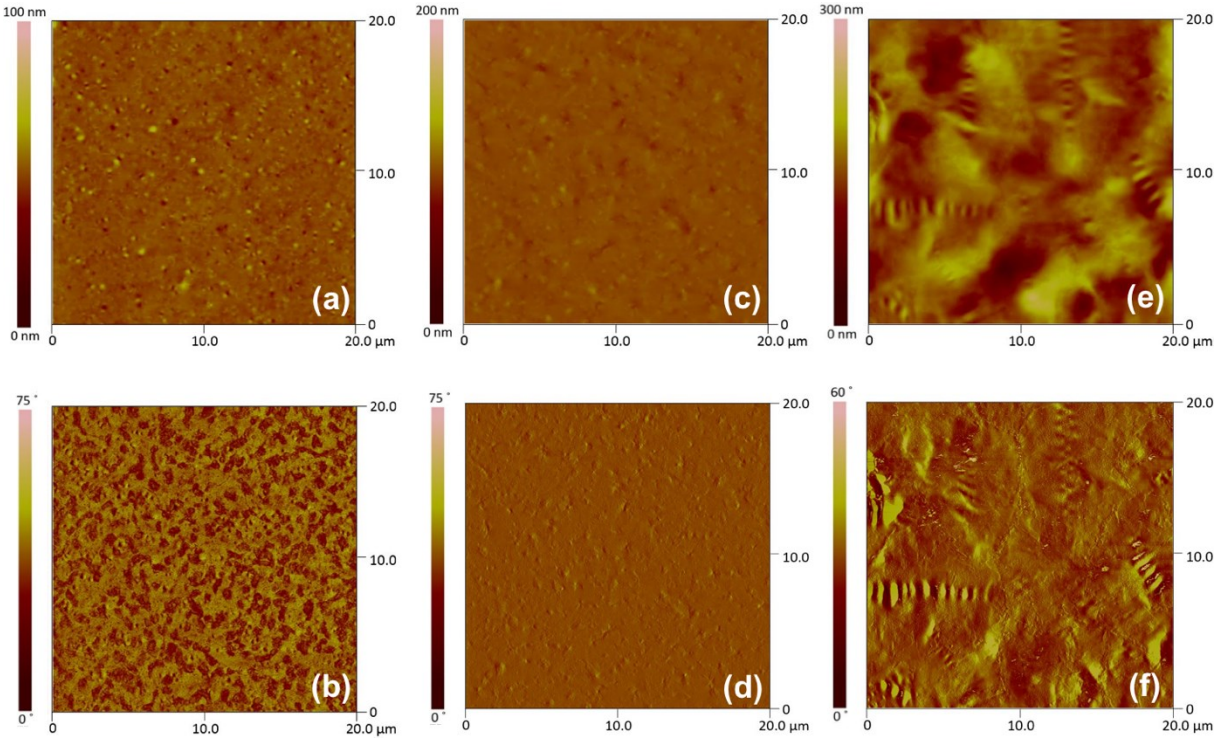


Figure 42. AFM images of CRM binder after long-term aging (RTFO+PAV residuals);
(a) Topographical image of CRM (b) Phase image of CRM
(c) Topographical image of CRM+L (d) Phase image of CRM+L
(e) Topographical image of CRM+S (f) Phase image of CRM+S

Figure 43 shows the roughness data recorded by AFM. The roughness data has been measured before and after aging of the binder. In general, the roughness values increased after long-term aging. The asphalt binder containing LEADCAP exhibited lowest roughness values except for aged CRM+L. Also, the addition of Sasobit resulted in the highest roughness values regardless of the binder types and aging levels.

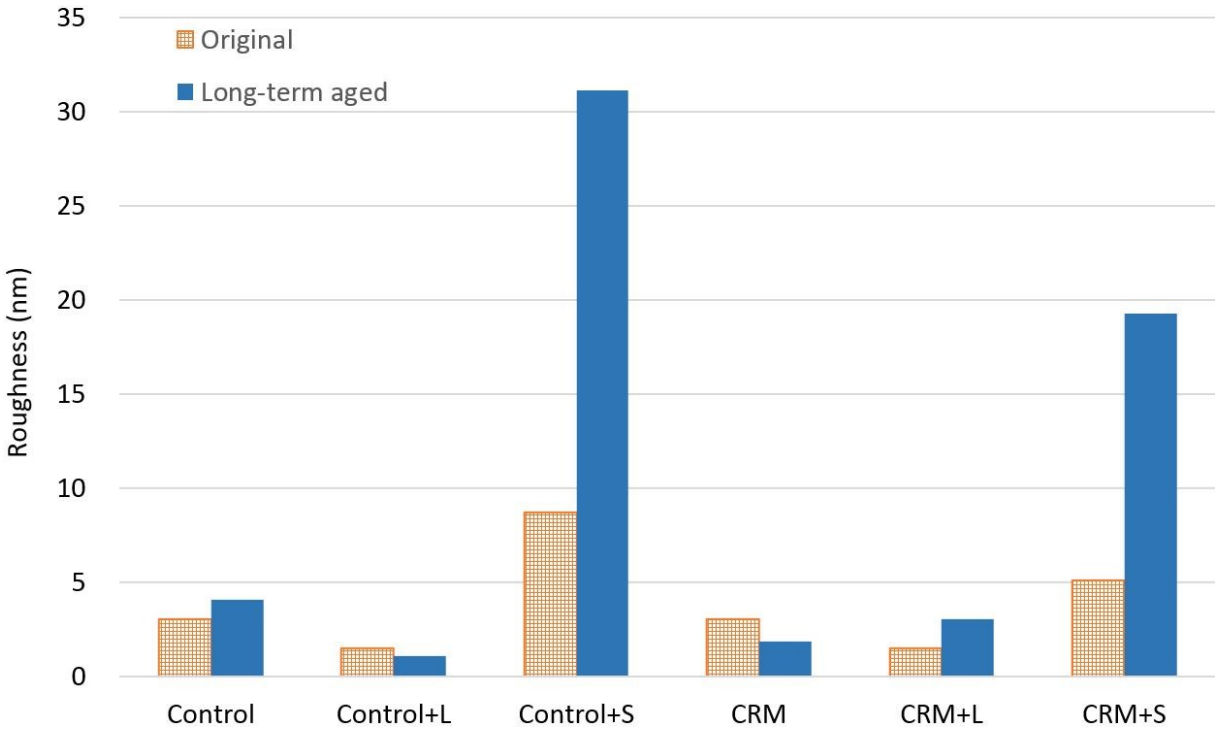


Figure 43. Roughness measured by AFM.

Statistical analysis

Using one-way analysis of variance, the statistical significance of the change in the roughness values with the binder types (control and CRM) and wax types (LEADCAP and Sasobit) was examined and the results are shown in Table 15. Interestingly, the statistical analyses of control and CRM binder with wax additives showed similar results at both aging states (original and long-term aged). The data indicated that the addition of Sasobit has a significant effect on the roughness of the binder samples regardless of base binder type. However, the difference of roughness values between the binder and binder with LEADCAP was not significant at 5% level within each base binder type.

In this study, the stiffness values of asphalt binders were correlated with the roughness data obtained by AFM scanning (Figure 44). Generally, the stiffness values are considered desirable attributes from the standpoint of resistance to cracking of the pavements. The correlation results showed that the roughness values increased as the stiffness values increased at both aging states, suggesting that the lower roughness resulted in an increase in the cracking resistance. When compared between each aging state, the roughness values at original state showed a good correlation with the stiffness values (R^2 value of 0.754). The correlation of roughness values with the stiffness values after aging was lower than that at original state.

Table 15. Statistical analysis results of the roughness value (α=0.05);
(a) Original (b) Long-term aged

(a)

Roughness		Control			CRM		
		1	2	3	1	2	3
Control	1	-	N	S	N	N	S
	2		-	S	N	N	S
	3			-	S	S	S
CRM	1				-	N	S
	2					-	S
	3						-

(b)

Roughness		Control			CRM		
		1	2	3	1	2	3
Control	1	-	N	S	N	N	S
	2		-	S	N	N	S
	3			-	S	S	S
CRM	1				-	N	S
	2					-	S
	3						-

1* CRM binder containing wax additive 1: None
2: LEADCAP
3: Sasobit

N: non-significant, S: significant.

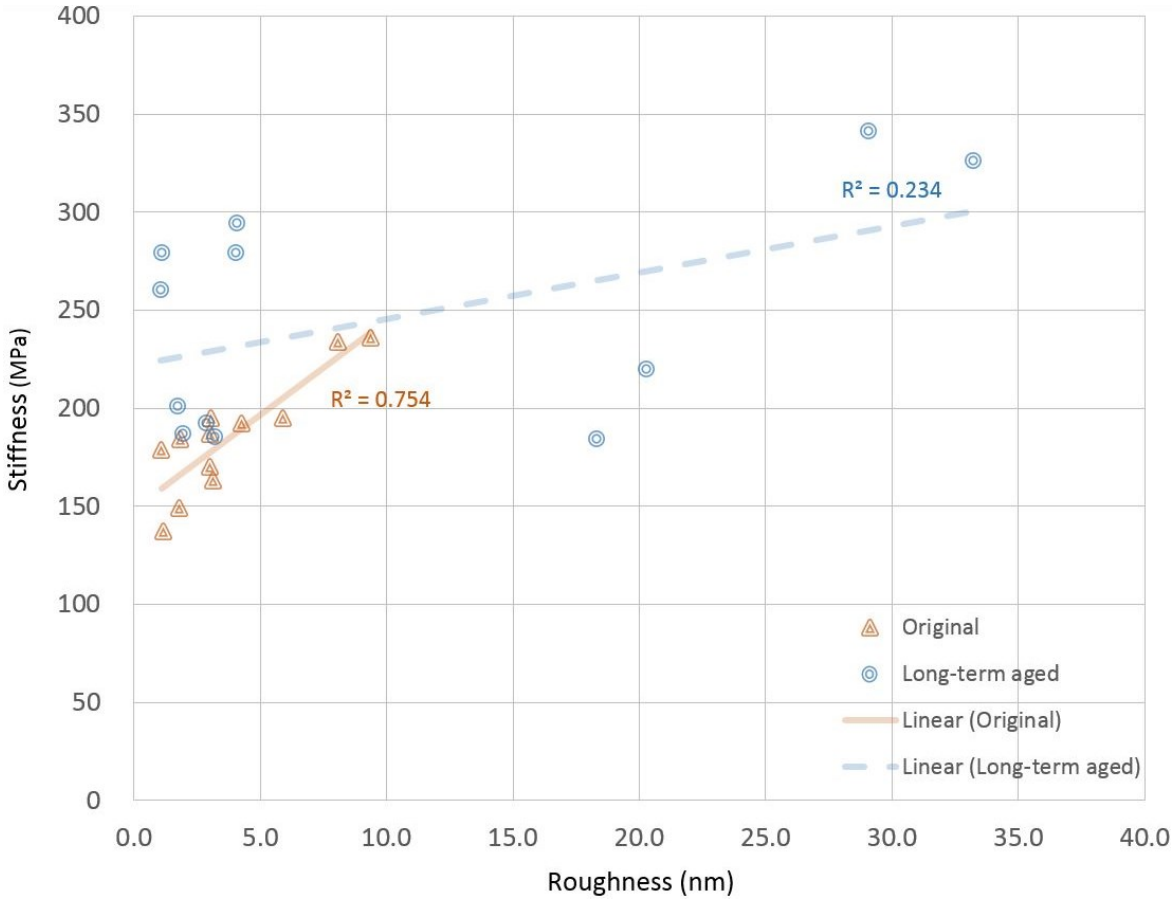
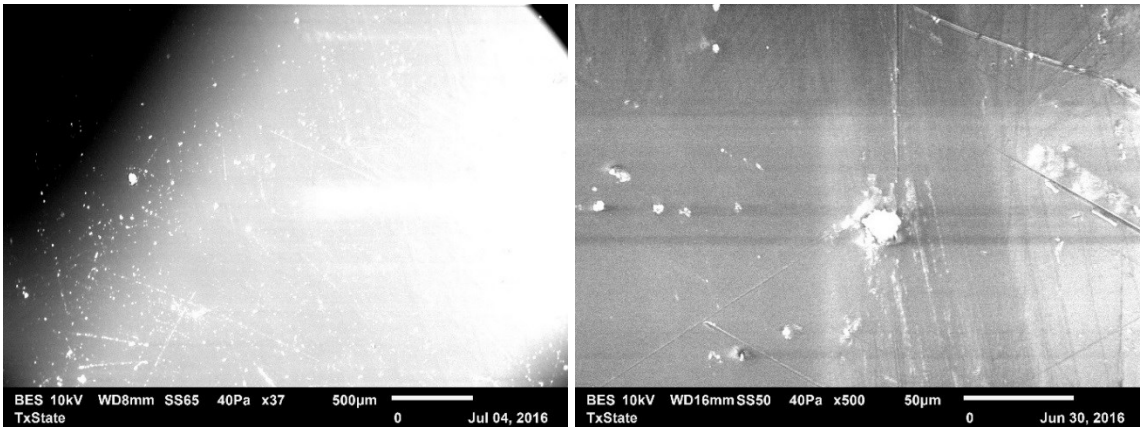


Figure 44. Relation of the roughness with stiffness.

Surface network morphology

ESEM investigations on morphology of CRM binder

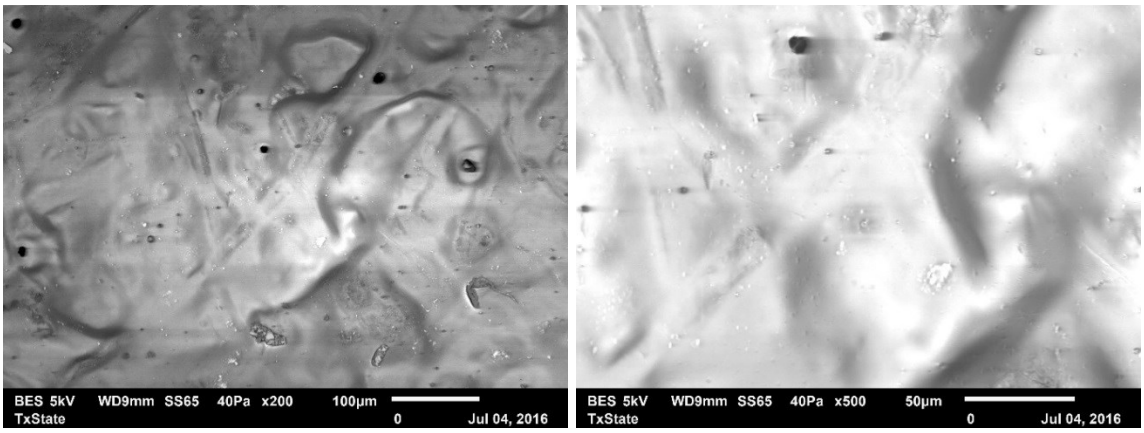
As mentioned earlier samples were preconditioned by controlled heating to 170°C in an oven. The samples were placed in the ESEM chamber. Figure 45 (a) shows the initial exposure of the sample in the ESEM chamber with relatively low magnification (37×). It seems like a plain surface. Figure 45 (b) presents the image of unaged CRM binder with a scan size of 50 µm and a magnification level of 500×. Although it shows detailed surface shape, no noticeable changes in the surface morphology have been observed. The reason might be the scanning done very adjacent to the tape surface. While making samples in this procedure one needs to be careful about the placement of the binder sample in order to avoid such situation.



(a) (b)

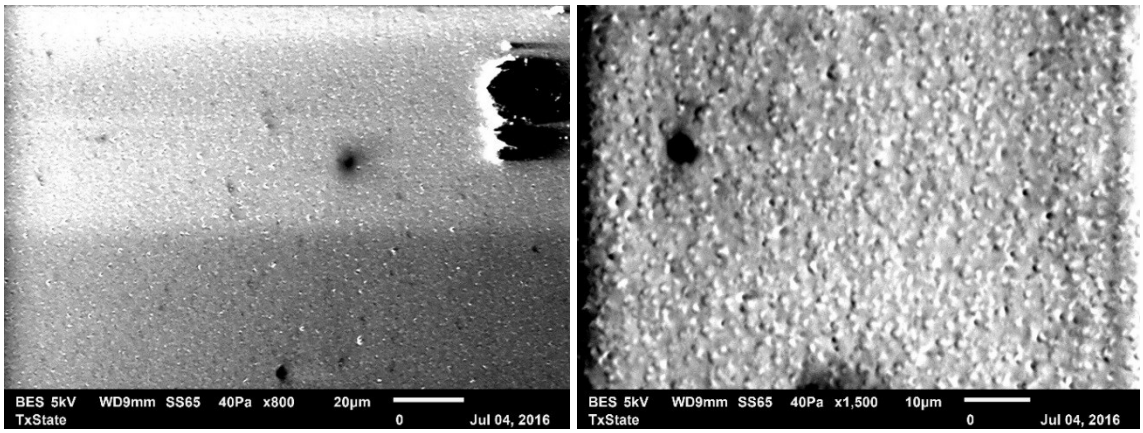
Figure 45. ESEM image of CRM asphalt binder at the border side of the sample.

Figure 46 (a) shows the liquid state of the CRM binder at 200× with the scan size of 100 μm. The image was captured as soon as the sample was exposed in the ESEM chamber. The microstructure of CRM binder appears to be a single-phase continuous non-uniform structure after adding crumb rubber particles. The rubber particles are densely packed and enveloped by the base binder. It represents the state of high viscous CRM binder after the addition of rubber particles in the base binder. This visual effect is similar to the mechanism explained in the article of Wang and Wei (2013) and Liu et al. (2013). Figure 46 (b) represents more detailed shape of the microstructural surface of the CRM binder with higher magnification of 500×. The image captured the network structure between the asphalt binder and crumb rubber. As a whole the rubber particles are not uniformly distributed in the asphalt binder. Figure 47 (a) shows a microscopic image of a uniform view in between rubber particles and asphalt binder after 24 hours with a magnification degree of 800× at a certain part of the sample. The more detailed image of this internal structure shown in Figure 47 (b).



(a) (b)

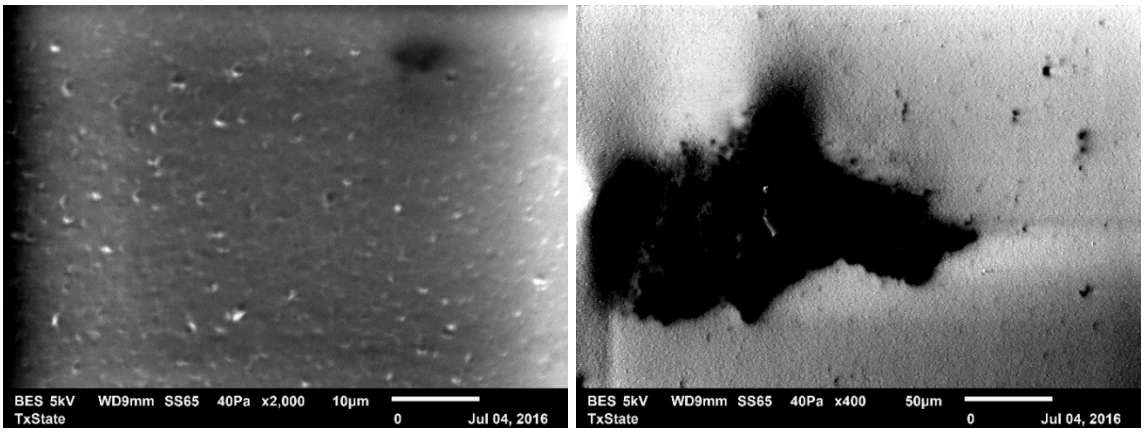
Figure 46. ESEM image of liquid state of CRM binder



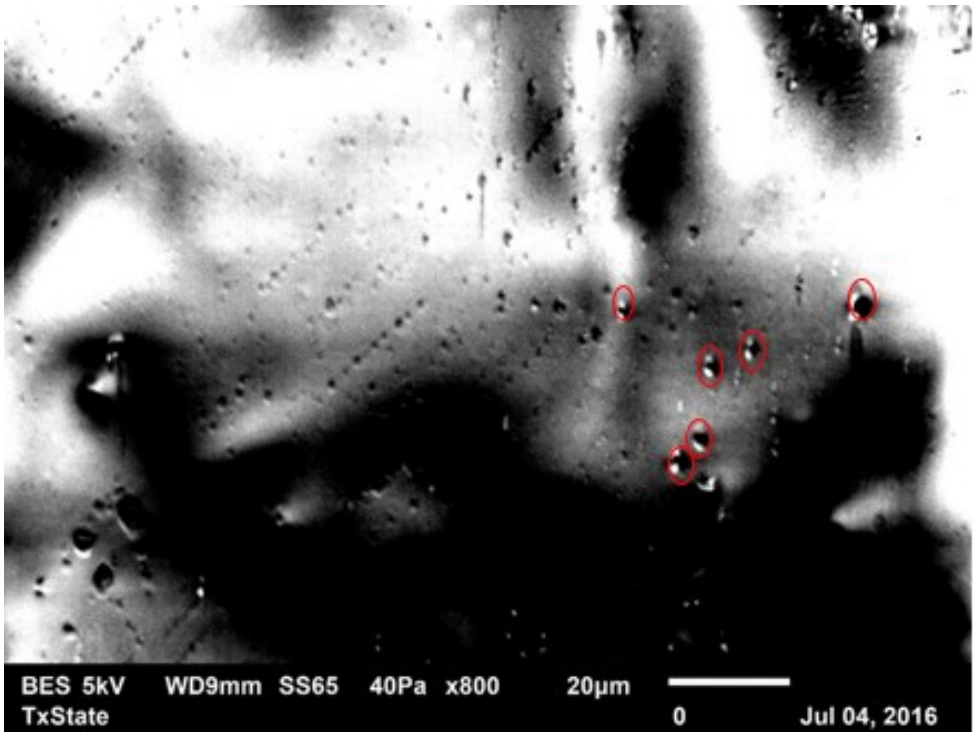
(a) (b)

Figure 47. ESEM image of interaction between rubber particles and asphalt.

ESEM images were captured for the aged CRM binder after a series of artificial aging. Figure 48 (a) presents the image of aged CRM binder with a scan size of 10 μm and a high magnification level of 2000×. The reason behind using high magnification is to capture the liquid state of the aged CRM binder as soon as it placed in the ESEM chamber after taking it from the oven. It seems like the image indicates the presence of rubber particles in the asphalt binder but due to high magnification, it is quite challenging to observe the actual morphology. This is because the density of electrons coming from the electron guns increases with the magnification of image (Danilatos, 1990). Therefore, the sample surface becomes unstable and produces a blurred image. As a result, it was decided to observe the development of microstructure of the aged CRM binder after 24 hours. Figure 48 (b) shows the morphology of the aged CRM binder after 24 hours. As it is obvious that the microstructure of the CRM binder should be changed due to the molecular mobility of the binder at room temperature and the oxidation of the surface of the binder. In order to observe this phenomenon in the aged CRM binder the same spot was observed at a high magnification degree of 800×. Figure 48 (c) illustrates the molecular changes in the aged CRM binder after experiencing the artificial aging.



(a) (b)



(c)

Figure 48. ESEM image for aged CRM binder.

The image appears to have two different phases. At the lower phase, there is a broad dark black region and the upper phase quite light compared to that bottom region. Those two phases resemble the interaction between the rubber particles and base binder at aged condition. High resolution showed more detailed shape of some small spot (red circles) which represent the property of aged asphalt binder. Masson et al. (2006) reported that this shape is formed due to the existence of asphaltene which is considered as primary material observed in aged asphalt binder.

In general, the enlarged images were taken from lower resolution to higher resolution. When decrease the resolution from highest resolution, rectangle shapes matching with visual size at each magnification were detected on sample surface. Figure 49 shows those damages on the surface of the binder. Although it is possible to take the images of asphalt binder using ESEM, the operation of ESEM need to be careful considering sample usefulness and research plan. It is recommended to use ESEM at the end of all micro-characterizing process regardless the sample damages.

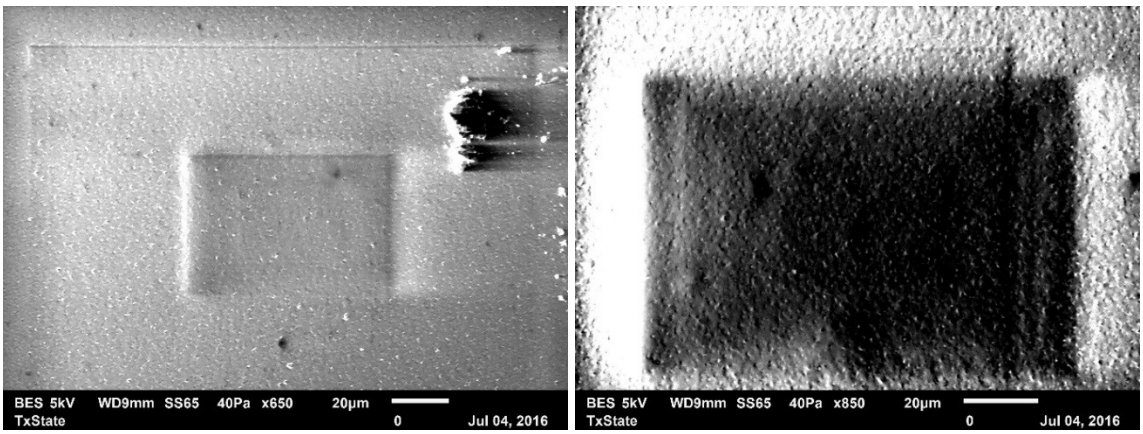


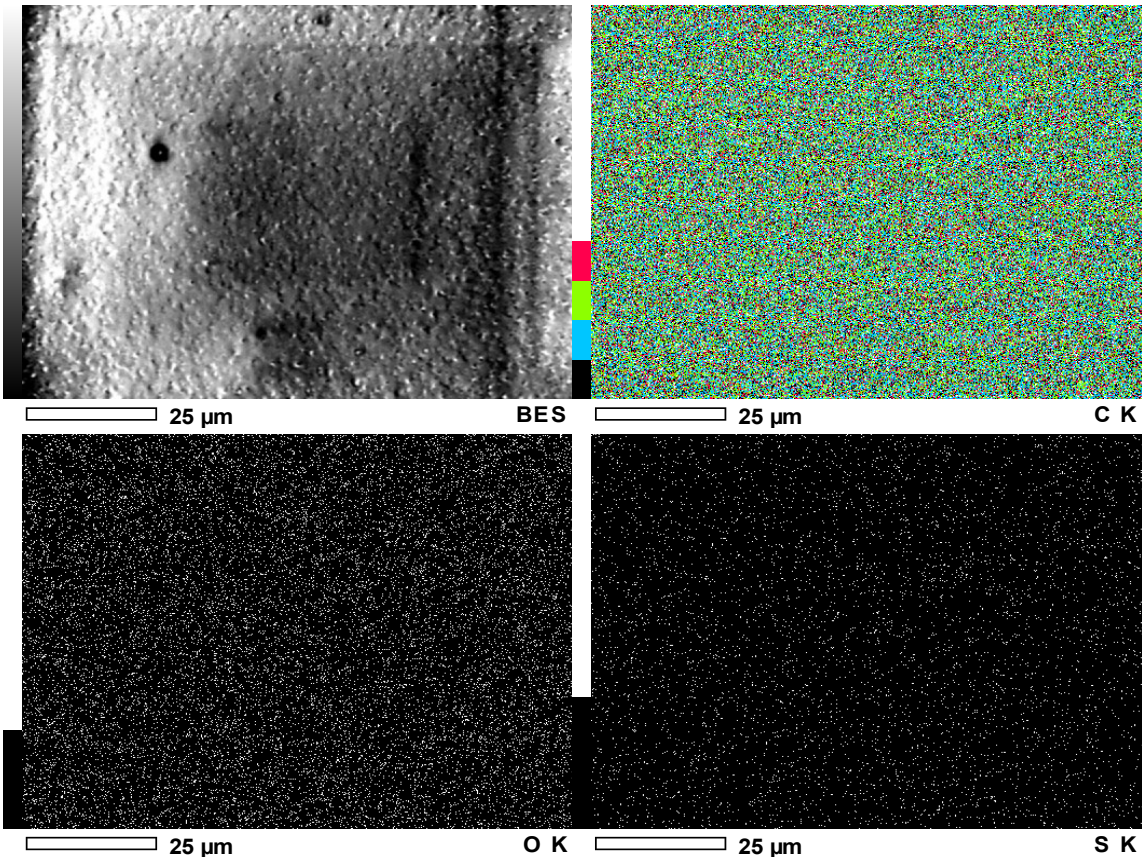
Figure 49. Sample damage by electron beam and vacuum pressure.

It is worth noting that this study also tried to take images using conventional SEM. The purpose is to observe if this sample preparation is suitable to take the image using Helios 400 SEM. It is very difficult to take the visible image of the CRM binder at liquid state just after taking the sample from the oven. Due to that reason the sample was dried at room temperature for 24 hours. Then the sample was loaded in the FEI Helios 400 SEM. Imaging was done in field free mode and with ETD detector for secondary electron. Since the sample contained organic compound, the imaging was started at lowest possible accelerating voltage and current (i.e., 2.0kV and 5.3Pa). The detector could not detect any image at this voltage. Then, the voltage was increased to 5kV keeping the current at 5.3Pa which did not produce any image. Voltage and current were further raised to 8kV and 86Pa. This time, sample was visible but the sample appeared to be melting. The detector was paused immediately and the voltage was reduced to 5kV. Another attempt was made to take images and similar phenomenon of sample damaging was observed once again. The imaging was stopped at this point. The main reason for such a behavior was found to be the chamber pressure. The chamber pressure in the FEI Helios 400 was very low because it usually works in ultra-high vacuum (<0.1torr). Based on such behavior of SEM it was confirmed that the SEM imaging of the present sample is only possible in ESEM where the pressure is not very low i.e. 1–50 Torr or 0.1–6.7 kPa.

7.3.2.1 Chemical composition analysis with EDX

One of the benefits of ESEM is elemental analysis by EDX mode. Using spot mode within a very short time the energy spectrum can be recorded and analyzed by the EDX-

Genesis software in order to get the chemical composition. The peak high ratios considered as the measurement of the chemical composition. Figure 50 shows the EDX spectrum as a plot of X-ray counts versus energy (keV) for aged CRM binder. The energy peaks corresponding to the various elements present in the aged CRM binder are also shown. The analysis helps to understand the chemical composition of the aged CRM binder. The peak high ratio of CRM binder indicates the primary composition of binder is hydro carbon. As shown in the graph, most parts of the material is consisted of hydro carbon and trace amount of oxygen and sulfur were detected. It is considered that the oxygen is generated through the aging process of CRM binder, and the sulfur is produced by the addition of crumb rubber. All expected constituents were detected through elemental analysis using ESEM. Currently, there are lots of research including different kinds of additives to produce better asphalt binder for paving roads. Therefore, it is expected that the application of elemental analysis using EDX will be beneficial for identifying the elemental property of different kinds of asphalt binder.



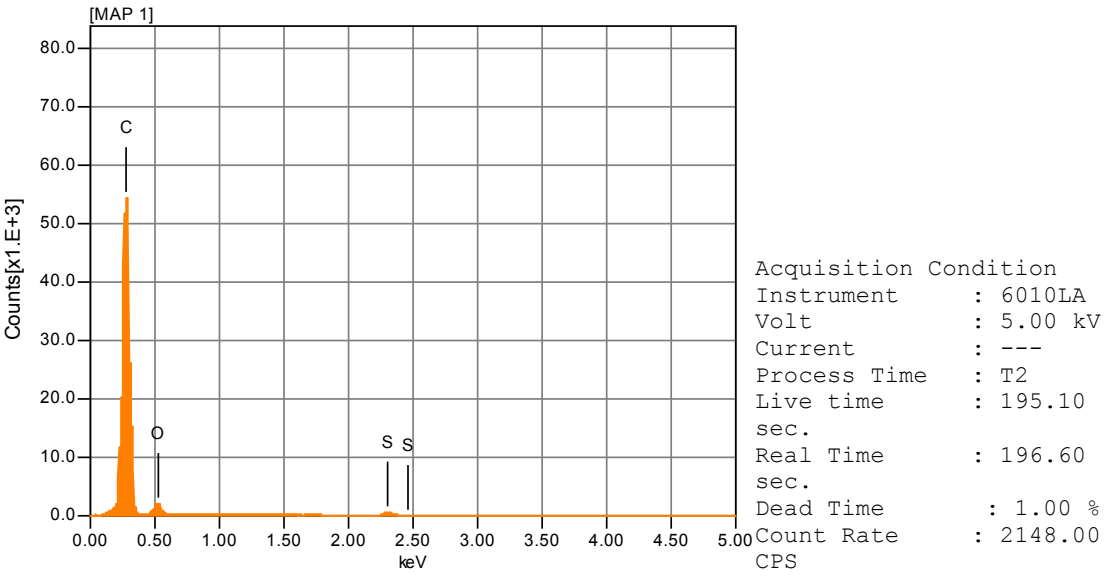


Figure 50. EDX spectrum for aged CRM binder.

Summary and Conclusions

The objective of this study is to investigate the microstructural components of CRM binder through the morphology images. The CRM binders were produced using 10% by the weight of the binder in the laboratory. Warm CRM binders were made with the wax additives of LEADCAP and Sasobit. The control and CRM binders were artificially aged through accelerated aging procedures. Optical microscopy was used to observe the surface image of the binder samples. In order to observe the microstructure on the surface of samples, the topographical and phase images were captured through AFM. In addition, microscopic morphology and chemical composition of the CRM binder was investigated by using ESEM and EDX techniques. From these test results, the following conclusions can be drawn for the materials used in this study.

1) The trends of the bee shapes shown on topographical and phase images of AFM seemed to indicate the cracking properties measured by Superpave binder tests.
2) It is found that the roughness data have the specific relationship with the cracking property of asphalt binder.
3) Unaged CRM binder appears to have a single-phase continuous non-uniform structure after adding crumb rubber particles.
4) The artificially aged CRM binder is observed to have two different phases. The lower phase consists of the broad dark region and the upper phase relatively light which resembles the interaction between the rubber and asphalt binder due to oxidation.
5) Aging effects of CRM binder were observed on the images captured by ESEM.

6) Overall, the microstructures on morphology images well reflected engineering properties depending on crumb rubber, wax additives, and aging states.

This chapter (VII) includes a part of the following publications;

Kim, H. H., Mazumder, M., Torres, A., Lee, S. J., "Characterization of CRM binders with wax additives using an atomic force microscopy (AFM) and an optical microscopy" *Advances in Civil Engineering Materials*, ASTM (Submitted)

Kim, H. H., Mazumder, M., Lee, S. J., "Identification of the microstructure components of crumb rubber modified binder (CRMB) and the feasibility of using environmental SEM (ESEM) coupled with energy dispersive X-ray spectroscopy," *International Journal of Highway Engineering* (Submitted)

VIII. SUMMARY, CONCLUSIONS, AND RECOMMENDATIONS FOR FUTURE RESEARCH

Summary

Crumb rubber modified (CRM) binder has gained an obvious reputation for several decades due to improved resistance to rutting and cracking. However, the high viscosity of CRM binder increases the mixing and compaction temperature of asphalt mixture in the plant. On the other hand, the warm mix asphalt (WMA) is being used with the specific benefit which reduces binder viscosity. The application of WMA technology is expected to reduce the managing temperatures of CRM binders. This study was initiated to characterize the properties of rubberized binder containing wax additives.

The viscosity properties of CRM binder with wax additives were investigated and the results are reported in Chapter IV. CRM binders were produced in the laboratory using four bending times and four CRM percentages. Two types of wax additives were used to produce WMA binders. The control binder of PG 64-22 was also mixed with wax additives at the same condition without adding the rubber. Basically, high temperature viscosity was measured through rotational viscometer (RV) test at 135 and 120°C. In addition, the viscosity change of CRM binders containing wax additive is evaluated using different testing temperatures (135°C and 120°C) and measuring periods (120 and 240 min) for the analysis of viscosity properties.

The rheological properties of CRM binders with wax additives were evaluated and the results are reported in Chapter V. The rutting resistance of two base binders (10% CRM and control binder) and their corresponding WMA binders were measured using dynamic shear rheometer (DSR) at original and short-term aging states. Also, four CRM percentages and two wax additives were used to investigate cracking property. DSR was used to evaluate cracking property at the thermal temperature (25°C) with long-term aged binder samples. The cracking property at low temperature (-12°C) was examined through bending beam rheometer (BBR) test. BBR test was conducted on all of the four CRM contents containing wax additives in three states: original (no aging), RTFO residual, and RTFO+PAV residual.

The performance properties of laboratory-prepared recycled CRM binders with wax additives were evaluated through Superpave binder tests in Chapter VI. Two base binders with wax additives were artificially aged using RTFO+PAV aging procedures, and the aged binders were recycled with 15% long-term aged (LTA) binders. The recycled binders were artificially aged through the same RTFO and PAV methods, and subsequently evaluated using the rotational viscosity, DSR, and BBR tests. The testing was conducted on all control and recycled binders in three states: original (no aging), RTFO residual, and RTFO+PAV residual.

The microstructural properties were studied using atomic force microscope (AFM), optical microscopy, and environmental scanning electron microscope (ESEM). In addition, the relationships between the morphology images and the cracking property depending on aging processes were evaluated and the results are discussed in Chapter VII. Two base

binders were prepared and mixed with two wax additives in the laboratory and aged using RTFO and PAV processes. Surface images of optical microscopy were captured for original and RTFO+PAV residual binder samples. Also, AFM was used to obtain the roughness data and detailed morphology images at original and long-term aging states. The difference between the microstructural properties of unaged CRM binder and aged CRM binder was investigated using ESEM.

Conclusions

Based on the results of this laboratory investigation, the following conclusions were made:

- In general, the binder viscosity increases at both testing temperature (135 and 120°C) with the increase of rubber content. Also, the increase in CRM percentage is considered to result in the steeper gradual viscosity increase. However, this study found that the addition of wax additives into asphalt binder was observed to have the significant effect on reduction of binder viscosity. On the other hand, the viscosity change of CRM binders at 120°C exhibited more stable increase curve, compared to the results at 135°C, indicating that the CRM mixes with the wax additive are advantageous to have better haul management. In addition, the blending time has little effect on the viscosity of CRM binder with wax additives.
- In the evaluation of the rheological property, it is resulted that the use of both wax additives and crumb rubber was a positive influence on increasing rutting resistance. Therefore, it is considered that the wax additives incorporated with the CRM binder

play a significant role in the resistance to permanent deformation of asphalt pavement.

- Crumb rubber in asphalt binder significantly decreased the creep stiffness and the G*sin δ values, suggesting that crumb rubber is positively effective for cracking resistance of asphalt binder. The CRM binder containing LEADCAP is found to have the lower stiffness compared to two base binders and their corresponding WMA binder with Sasobit. Also, the addition of Sasobit is resulted in negative effect on cracking resistance for all aging states.
- According to the viscosity results of recycled CRM binder with wax additives, the recycled CRM binder showed the similar trend with the results at original state. In addition, the rutting and cracking property of recycled binders exhibited quite consistent trends with original binders. These results indicate that the wax additive is still effective in recycled binders, even though the additives were subjected to short and long-term aging processes.
- Based on the surface morphology images, the trends of the bee shape which reflect aging property were consistent with the cracking properties measured by Superpave binder tests. In addition, the roughness data have the specific relation on the cracking performances of asphalt binder. It is expected that the microstructural topography and roughness data obtained through optical microscopy and AFM can be applied to evaluate the cracking property of asphalt binder.

Recommendations for Future Research

On the completion of this study, the following topics of future research are recommended to build on the findings of this investigation:

- Evaluate the mechanical properties of CRM mixtures with wax additives in order to correlate with the performance properties and compositional change of CRM binders with wax additives.
- Evaluate the properties of CRM binders recycled with reclaimed asphalt pavement (RAP) binder from the field, instead of artificially long-term aged (LTA) CRM binders produced in the laboratory.
- Evaluate the microstructural properties of CRM mixtures containing wax additives through AFM, ESEM, and other applicable equipment for further investigation.

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