Development of Long-Term Aging Factor For Asphalt Mixtures’ Cracking Resistance

Yucheng Shi
Louisiana State University and Agricultural and Mechanical College

Follow this and additional works at: https://digitalcommons.lsu.edu/gradschool_theses
Part of the Civil Engineering Commons, and the Transportation Engineering Commons

Recommended Citation
Shi, Yucheng, "Development of Long-Term Aging Factor For Asphalt Mixtures’ Cracking Resistance" (2018). LSU Master's Theses. 4827.
https://digitalcommons.lsu.edu/gradschool_theses/4827

This Thesis is brought to you for free and open access by the Graduate School at LSU Digital Commons. It has been accepted for inclusion in LSU Master's Theses by an authorized graduate school editor of LSU Digital Commons. For more information, please contact gradetd@lsu.edu.
DEVELOPMENT OF LONG-TERM AGING FACTOR FOR ASPHALT MIXTURES’ CRACKING RESISTANCE

A Thesis

Submitted to Graduate Faculty of the Louisiana State University and Agricultural and Mechanical College in partial fulfillment of the requirements for the degree of Master of Science in Civil Engineering

in

The Department of Civil and Environmental Engineering

by

Yucheng Shi
B.S., Hefei University of Technology, 2016
December 2018
# TABLE OF CONTENTS

LIST OF TABLES .................................................................................................................................................. iii

LIST OF FIGURES ................................................................................................................................................. iv

NOMENCLATURE, SYMBOLS AND ACRONYMS ............................................................................................ vi

ABSTRACT ............................................................................................................................................................ vii

Chapter 1. INTRODUCTION ................................................................................................................................. 9
  1.1. Background .................................................................................................................................................. 9
  1.2. Problem Statement ....................................................................................................................................... 10
  1.3. Research Objectives ................................................................................................................................... 11
  1.4. Research Scope ........................................................................................................................................... 11
  1.5. Research Approach ..................................................................................................................................... 13
  1.6. Reference .................................................................................................................................................... 19

Chapter 2. LITERATURE REVIEW ...................................................................................................................... 21
  2.1. Asphalt Binder ........................................................................................................................................... 21
  2.2. Oxidative Aging of Asphalt Binders ......................................................................................................... 22
  2.3. Aging Characterization of Asphalt Binder ............................................................................................... 25
  2.4. Semi-Circular Bend (SCB) Test ................................................................................................................ 30
  2.5. Reference .................................................................................................................................................... 32

Chapter 3. EXPERIMENTAL PROGRAM .......................................................................................................... 35
  3.1. Material Description .................................................................................................................................. 35
  3.2. Sample Fabrication ..................................................................................................................................... 38
  3.3. Material Characterization ......................................................................................................................... 40
  3.4. Reference ..................................................................................................................................................... 44

Chapter 4. RESULTS AND ANALYSIS ............................................................................................................ 46
  4.1. SCB Test Results ........................................................................................................................................ 46
  4.2. Rheological Characterization Results ....................................................................................................... 48
  4.3. Chemical Characterization Results .......................................................................................................... 60
  4.4. Development of Aging Factor and Cracking Resistance Prediction Model ............................................. 66
  4.6. Reference ..................................................................................................................................................... 75

Chapter 5. SUMMARY AND CONCLUSION ..................................................................................................... 77
  5.1 Future Work .................................................................................................................................................. 78

VITA ..................................................................................................................................................................... 80
LIST OF TABLES

Table 1.1. Aging Conditions and Testing Matrix ................................................................. 12
Table 3.1. Asphalt Mixture Composition ........................................................................... 38
Table 4.1. Statistical Analysis of SCB $J_c$ Results ............................................................. 48
Table 4.2. Continuous High PG Grade of Extracted Binder .............................................. 49
Table 4.3. GPC Test Results ............................................................................................... 65
Table 4.4. Mixture Characteristics for Model Validation .................................................. 74
LIST OF FIGURES

Figure 1.1. Research Approach Flow Chart ................................................................. 13
Figure 2.1. The Maxwell Model: An Elastic and Viscous Element in Series ..................... 26
Figure 2.2. A Typical Master Curve and Physical Properties .......................................... 28
Figure 3.1. Mixture Gradation ......................................................................................... 37
Figure 3.2 Typical SCB Test Displacement versus Force Curves Example ......................... 41
Figure 3.3. Example of Critical Strain Energy Release Rate (Jc) Calculation ....................... 42
Figure 4.1. Semi-Circular Bend Test Results at 25 °C ...................................................... 47
Figure 4.2. |G*| and Phase Angle Master Curves of Binder Extracted from Mix 1 at Various Aging Levels ......................................................................................................................... 51
Figure 4.3. |G*| and Phase Angle Master Curves of Binder Extracted from Mix 2 at Various Aging Levels ......................................................................................................................... 51
Figure 4.4. |G*| and Phase Angle Master Curves of Binder Extracted from Mix 3 at Various Aging Levels ......................................................................................................................... 52
Figure 4.5. |G*| and Phase Angle Master Curves of Binder Extracted from Mix 4 at Various Aging Levels ......................................................................................................................... 52
Figure 4.6. |G*| and Phase Angle Master Curves of Binder Extracted from Mix 5 at Various Aging Levels ......................................................................................................................... 53
Figure 4.7. G-R Parameter Results in Black Space Diagram for Mix 1 .............................. 55
Figure 4.8. G-R Parameter Results in Black Space Diagram for Mix 2 .............................. 55
Figure 4.9. G-R Parameter Results in Black Space Diagram for Mix 3 .............................. 56
Figure 4.10. G-R Parameter Results in Black Space Diagram for Mix 4 .............................. 56
Figure 4.11. G-R Parameter Results in Black Space Diagram for Mix 5 .............................. 57
Figure 4.12. G-R Results at Different Aging Levels ................................................................. 58
Figure 4.13. Crossover Modulus at Different Aging Levels ....................................................... 59
Figure 4.14. Determination of Maltenes and Asphaltenes Content of PG 76-22 Binder Based on the Molecular Weight Regions of The GPC Curve. ................................................................. 61
Figure 4.15. Deconvolution of the GPC elution curve of the PG 76-22 asphalt binder containing SBS polymer ................................................................................................................................................. 62
Figure 4.16. Comparison of GPC Curves ..................................................................................... 64
Figure 4.17. Correlation between G-R Parameter and SCB $J_c$.................................................. 66
Figure 4.18. G-R Aging Factors for SCB $J_c$.............................................................................. 68
Figure 4.19. Comparison of Measured and Predicted SCB $J_c$ ................................................... 68
Figure 4.20. Predicted SCB $J_c$ versus Measured SCB $J_c$ (Individual Aging Factor) ............ 69
Figure 4.21. Comparison of Measured and Predicted SCB $J_c$ ................................................... 70
Figure 4.22. Predicted SCB $J_c$ versus Measured SCB $J_c$ (Averaged Aging Factor) ............. 71
Figure 4.23. Predicted SCB $J_c$ versus Measured SCB $J_c$ (Prediction Model) ....................... 73
Figure 4.24. Predicted SCB $J_c$ versus Measured SCB $J_c$ (Validation) ................................. 74
**NOMENCLATURE, SYMBOLS AND ACRONYMS**

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>AASHTO</td>
<td>American Association of State Highway and Transportation Officials</td>
</tr>
<tr>
<td>ANOVA</td>
<td>Analysis of Variance</td>
</tr>
<tr>
<td>ASTM</td>
<td>American Society for Testing Materials</td>
</tr>
<tr>
<td>BBR</td>
<td>Bending Beam Rheometer</td>
</tr>
<tr>
<td>C-A Model</td>
<td>Christensen-Anderson Model</td>
</tr>
<tr>
<td>COV</td>
<td>Coefficient of Variation</td>
</tr>
<tr>
<td>DSR</td>
<td>Dynamic Shear Rheometer FHWA Federal Highway Administration</td>
</tr>
<tr>
<td>FTIR</td>
<td>Fourier-transform Infrared Spectroscopy</td>
</tr>
<tr>
<td>G-R Parameter</td>
<td>Glover-Rowe Parameter</td>
</tr>
<tr>
<td>G(_{\text{mm}})</td>
<td>Maximum Theoretical Specific Gravity</td>
</tr>
<tr>
<td>HMA</td>
<td>Hot-Mix Asphalt</td>
</tr>
<tr>
<td>J(_{c})-value</td>
<td>Critical Strain Energy Release Rate</td>
</tr>
<tr>
<td>LADOTD</td>
<td>Louisiana Department of Transportation and Development</td>
</tr>
<tr>
<td>LTRC</td>
<td>Louisiana Transportation Research Center</td>
</tr>
<tr>
<td>NAPA</td>
<td>National Asphalt Pavement Association</td>
</tr>
<tr>
<td>NMAS</td>
<td>Nominal Maximum Aggregate Size</td>
</tr>
<tr>
<td>QC/QA</td>
<td>Quality control /Quality assurance</td>
</tr>
<tr>
<td>RAP</td>
<td>Recycled Asphalt Pavement</td>
</tr>
<tr>
<td>RAS</td>
<td>Recycled Asphalt Shingles</td>
</tr>
<tr>
<td>REOB</td>
<td>Re-refined Engine Oil Bottom</td>
</tr>
<tr>
<td>SBS</td>
<td>Styrene Butadiene Styrene</td>
</tr>
<tr>
<td>SCB</td>
<td>Semi-Circular Bending</td>
</tr>
</tbody>
</table>
ABSTRACT

To characterize the cracking resistance of asphalt mixtures, a semi-circular bend (SCB) test has been developed and subsequently accepted as a routine test in mixture design in Louisiana. However, a SCB test requires specimens to be aged for five days in the laboratory, which makes it not implementable in quality control (QC)/quality assurance (QA).

The primary project objective was to develop an aging factor to transfer the unaged SCB $J_c$ to the aged SCB $J_c$. Specific objectives were to: (1) investigate the effect of laboratory aging on asphalt mixtures’ cracking resistance and asphalt binders’ rheological and chemical properties, (2) develop an aging factor to represent the laboratory aging effect on asphalt binder property; and (3) develop a cracking resistance prediction model based aging index.

To achieve the objectives, a SCB test was performed on five plant-produced mixtures that were laboratory-aged for designated durations (0, 2, 5, and 7 days) to obtain the $J_c$ values at various aging levels. Then, the asphalt binder was extracted and recovered from each mixture. After that, the rheological and chemical properties of extracted binders were evaluated by using a dynamic shear rheometer (DSR) and Gel permeation chromatography (GPC) test.

SCB test results indicated that the cracking resistance of asphalt mixtures decreased with aging, and 5 day aging SCB $J_c$ values were found to be comparable to that of 7 days aged mixtures. The aging index of Glover-Rowe (G-R) parameter obtained from the DSR test increased with aging, and the G-R parameter values at 5 days aging level were comparable to that of 7 days aged mixtures. GPC test indicated that asphaltene fraction increased while maltene fraction decreased with the aging. By considering the remarkable correlation between of G-R parameter with SCB $J_c$, an aging factor of 1.62 based on G-R parameter was proposed for mixture containing polymer modified binder. The aged SCB $J_c$ values were transferred from
unaged SCB $J_c$ by utilizing proposed aging factor, and the transferred SCB aged $J_c$ were found in good agreement with the measurement. Furthermore, a preliminary cracking resistance prediction was explored and validated by considering other mixture characteristics.
CHAPTER 1. INTRODUCTION

1.1. Background

Cracking is considered as one of major distresses in asphalt pavements that reduces the service life and drives rehabilitation need. In Louisiana, the Superpave method has been commonly used to design asphalt mixtures by mainly controlling of asphalt mixtures’ volumetric and physical properties. However, it has been realized that the volumetric properties are not directly related to the mechanical response of the mixtures. Moreover, the increasing uses of recycled materials including reclaimed asphalt pavement (RAP) and recycled asphalt shingles (RAS), rejuvenators such as re-refined engine oil bottom (REOB), and a number of warm-mix technologies make the asphalt mixture design more complicated. The uses recycled materials have been reported to potentially result in negative effect on pavement performance. Particularly, the incorporation of recycled asphalt binder from RAP and RAS would tend to deteriorate the stress relaxation capability of the mixture, thereby making asphalt mixture more prone to cracking [1]. The application of REOB has been reported to have a negative effect on cracking resistance at intermediate temperature [2].

For giving facts, the volumetric-based Superpave mixture design method is recommended to be implemented with mechanistic tests in respect of crack resistance [3]. In order to evaluate the cracking resistance of asphalt mixture, Semi-circular bend (SCB) test has been developed based on fracture mechanism concept. SCB test has been validated with field cracking performance in Louisiana and subsequently accepted by Louisiana Department of Transportation and Development (LADOTD) as a routine test in mixture design in the 2016 Specifications for Roads and Bridges [4-6].
1.2. Problem Statement

The quality control (QC) and quality assurance (QA) is also essential to accomplish construction of asphalt pavements with satisfactory performance in addition to adopting appropriate asphalt mixture design. The use of QC/QA specifications has been reported to be able to achieve the reduction in variability and decrease the cost of the pavement over time through better performance and longer service life [7]. In QC/QA, Hot Mix Asphalt (HMA) is produced with constant control throughout production and placement operations by monitoring the required characteristics to guarantee the mixtures are produced as designed.

Currently, the QC/QA specification in Louisiana mainly controls asphalt mixtures volumetric and physical properties as key characteristics, while no fundamental properties obtained from mechanistic tests are incorporated in regarding of cracking resistance. By implementing SCB test in QC/QA, the mixture quality in terms of cracking resistance will be monitored during the whole production and construction. However, the challenge is that SCB test requires a laboratory aging process for 120 hours as per AASHTO R30 to simulate long-term aging in the field. Obviously, five days aging duration is not feasible for the implementation of SCB test in QC/QA. Although a number of research studies have been conducted to develop expedite laboratory aging method, none of proposed method is reliable and practical with consensus.
1.3. Research Objectives

The primary objective of the proposed research is to investigate the mechanism of laboratory aging and develop an expedite test protocol to implement SCB test for QC/QA. Specific objectives of the project include:

a) to investigate the effect of aging on asphalt binder properties in regard to aging and asphalt mixtures’ cracking resistance;

b) to develop an aging factor that represent the laboratory aging effect on asphalt binder’s property;

c) to develop a cracking resistance prediction model based aging index.

1.4. Research Scope

To achieve the objective of this study, a number of plant-produced asphalt mixtures with a good plant record of consistency were identified. The selected projects are expected to incorporate a range of material types (e.g., virgin and recycled) and volumetric. The laboratory compacted samples were conditioned to obtain a series of progressive aging intensities and then evaluated using SCB test; see Table 1.1 For each mixture with each aging condition, the asphalt binder were extracted and evaluated with respect to rheological and chemical characteristics. The rheological binder testing under consideration includes a frequency sweep test at multiple temperatures and Superpave performance grading. The chemical evaluation is aimed to examine the shift of components using gel permeation chromatography (GPC) during progressive aging, respectively. Table 1.1 presents the testing matrix for each mixture and extracted binder. Based on the evaluation of measured rheological and chemical characteristics, appropriate aging indexes were selected based on the screening of sensitivity and reliability. After that, selected aging indexes were correlated with $J_c$ of asphalt mixtures to establish the relationship between
asphalt binder aging index and asphalt mixtures cracking resistance. Eventually, an aging factor is expected to be develop and proposed to transfer the $J_c$ of unconditioned asphalt mixture to that of the long-term aged mixtures.

Table 1.1. Aging Conditions and Testing Matrix

<table>
<thead>
<tr>
<th>Aging Duration (85°C)</th>
<th>Mixture Test</th>
<th>Binder Tests</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Rheological</td>
</tr>
<tr>
<td>0, 2, 5, 7 days</td>
<td>SCB</td>
<td>Performance grading</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Frequency sweep</td>
</tr>
</tbody>
</table>

Note: SCB: semi-circular bend; GPC: gel permeation chromatography.
1.5. Research Approach

Figure 1.1 illustrates the overall research approach of this study. In this study, asphalt binder and mixture laboratory experiments were conducted. First, the SCB test was conducted to characterize the cracking resistance of mixtures with various aging levels, including plant mixtures (considered as short-term aged mixtures) and laboratory long-term aged mixtures for designated durations (2, 5, and 7 days). After that, asphalt binders were extracted and recovered.
from the tested SCB specimens. The DSR test was conducted to obtain the rheological properties of extracted binder; while the GPC tests were conducted to obtain the chemical properties of binders. This task was performed as described in the following subtasks.

Task 1: Literature Review

During this task, the data from a variety of sources regarding completed and on-going laboratory studies was reviewed with respect to aging of asphalt binders and mixtures. This task should include the most up-to-date testing technology, theories, and analysis approaches that have been developed and used to characterize the effects of oxidative aging on the rheological and chemical properties of asphalt binders and on the mechanical responses of mixtures in terms of cracking resistance. The literature review included, but not be limited to, standard methods such as Transportation Research Information Database (TRID), Computerized Engineering Index (COMPENDEX), National Technical Information Services (NTIS), as well as consulting with domestic and international experts in the field.

Task 2: Identify Field Projects and Collect Plant Loose Mixtures

During this task, five plant-produced loose asphalt mixtures were selected through consultation with the Louisiana DOTD construction and research personnel. Fortunately, this study coordinates with FHWA Demonstration Project in Louisiana titled Enhanced Durability of Asphalt Pavements through Increased In-Place Pavement Density.

During mixture production, loose mixtures were sampled and transported to the asphalt laboratory of the Louisiana Transportation Research Center (LTRC) for experimental characterization. During the design, production, and construction phases, all relevant records were be collected, including job mix formulas (JMF), typical QC/QA reports, and other observations.
Task 3: Conduct Laboratory Experiments

The objective of this task is to perform a comprehensive experimental characterization of the asphalt mixtures with various aging levels. The loose mixtures were compacted in the laboratory using a Superpave gyratory compactor. Prior to testing, all samples were conditioned for the designated aging periods as listed in Table 1.1. SCB test was conducted to ascertain the crack resistance of mixtures in each case, while a suite of rheological and chemical tests were performed on the extracted binders.

a) Subtask 3.1: Laboratory aging on compacted specimen

According to AASHTO R30 [8], Standard Practice for Mixture Conditioning of Hot Mix Asphalt (HMA), the specimens are required to be aged in a compacted state in an air force oven for 120 hours (5 days). In this study, all the specimens were compacted to 7±0.5 %. Then the cylindrical specimens were cut into two semi-circular specimens. After that, the specimens were notched in the center of specimens with notch depth of 25.4 and 38.1 mm. The middle notch is used to inspect the linearity relationship between strain energy and notch depth and was reported to not affect the calculation of $J_c$ [9]. When the completion of fabrication, the specimens were put on flat, clear aging plates and then put into the ovens at a temperature of 85°C, which will be monitored continuously. The aging durations were designated as 0, 2, 5 and 7 days to incorporate a reasonable range of aging levels.
b) Subtask 3.2: Asphalt mixture testing

The SCB test was conducted using an AMPT equipment in accordance to ASTM D8044 [10], Standard Test Method for Evaluation of Asphalt Mixture Cracking Resistance using the Semi-Circular Bend Test (SCB) at Intermediate Temperatures. A minimum of four semi-circular specimens should be prepared with each of the three notch depths: 25.4 and 38.1 mm. During testing, each specimen is loaded in a three-point bending configuration until fracture at a constant displacement rate of 0.5 mm/min. and a testing temperature of 25°C. The collected load and displacement data can be processed to yield the crack resistance parameter, $J_c$. As already mentioned, the SCB test has been implemented by Louisiana DOTD as part of its balanced mix design practice in the Specification. The Louisiana DOTD specifications require a minimum SCB $J_c$ values of 0.5- and 0.6 kJ/m$^2$ for low (≤ 3 million ESALs) and high traffic volume (> 3 million ESALs), respectively.

c) Subtask 3.3: Extract Binders from Mixtures

For rheological and chemical analysis, short-term aging mixtures and long-term aging mixtures aged for designated durations will be extracted in accordance with AASHTO T 164–Method A and AASHTO R 59. Trichloroethylene (TCE) was used for extraction and the solutions of TCE and asphalt binder were then heated and distilled within the specific temperature range of 160° to 166°C under the protective gas environment of carbon dioxide to remove all traces of TCE. It should be noted that recovery may potentially alter asphalt binder properties due to the inability to remove all asphalt from aggregate and the residual solvent left in the binder [11, 12]. However, previous studies showed procedures used by the research team of extraction and recovery process had minimal effect on asphalt binder properties.
d) Subtask 3.4: Measurement of Rheological Properties

Rheological properties were indicated to be related directly to the mechanical properties of an asphalt mixture and are readily relatable to pavement distresses [9]. In preliminary plan, Superpave performance grading and frequency sweep at multiple temperatures (also referred as dynamic shear modulus test, or G* test) were conducted. By conducting dynamic shear modulus test, the master curves were established for each binder, and the rheological information needed for calculating rheological aging index was extracted from the built master curves.

- Superpave performance grading

The extracted asphalt binders will be graded using the Superpave method in accordance with AASHTO R 29, Standard Practice for Grading or Verifying the Performance Grade (PG) of an Asphalt Binder. A DSR will be employed to determine the high PG. The high PG grading will be used as a quick-check test to track the aging process of asphalt materials.

- Frequency sweep at multiple temperatures

The frequency sweep test was performed from 0.1 to 100 rad/s at multiple temperatures of 45°, 35°, and 15°C using a DSR with parallel plates of 8-mm diameter and 2-mm gap. During the test, the strain level was controlled at 1% to ensure the asphalt binder was in the linear viscoelastic range. The obtained test data from the frequency sweep test was used to construct the master curves for dynamic shear modulus and phase angle, from which the effects of various aging intensities on the stiffness and elasticity properties were examined and quantified.
e) Subtask 3.5: Measurement of Chemical Properties

- Gel permeation chromatography (GPC)

Asphalt chemistry is extremely complicated. Researchers have been using the high-pressure gel permeation chromatography (HP-GPC) to study the molecular size distribution in asphalt. GPC accomplishes the separation of molecules in a sample based on the sizes, or more specifically, the hydrodynamic volumes of the molecules, a technique analogous to the aggregate sieving process in which the largest molecules elute first followed successively by smaller ones. Use of GPC opens an opportunity to examine the material from a microscopic perspective and makes it possible to correlate the macroscopic behaviors of asphalt cement and mixtures with molecular composition of asphalt binder. Aging is able to degrade large molecules of polymer modifier into smaller molecular sizes whereas on the other side in the base asphalt cement aging significantly increases the amount of large molecular size species and decreases those of medium and small molecular sizes.

Task 4: Perform data analysis

During this task, the measured mechanistic test data of asphalt binders and mixtures from laboratory experiments were compiled into an integrated database, which was used for further data reduction and analysis. The evaluation parameters from mechanical testing were obtained by following corresponding test standard and the most recent literature. A series of plots were created for qualitative comparisons and visual inspections of the test data. Focus was placed on the development of correlation between $J_c$ of the mixtures and the rheological/chemical properties of asphalt binders at different aging intensities. Statistical techniques such as analysis of variance (ANOVA) and regression analysis was also under consideration and employed.
Task 5: Prepare thesis report

A final thesis was prepared that summarize and document all the findings, experiments, results, conclusions, and problems encountered during the project period.

1.6. Reference


CHAPTER 2. LITERATURE REVIEW

2.1. Asphalt Binder

Asphalt binder (or asphalt cement) is known as a dark brown to black cementitious material that is either naturally occurring or is produced by petroleum distillation [1]. Asphalt has been used as a waterproof material in shipbuilding and hydraulics at early time, but it is primarily used as a “gluing” material in pavement construction nowadays. In pavement construction, asphalt is commonly heated up and then mixed with aggregates to produce asphalt mixture, which is also known as asphalt concrete. Asphalt is the most used material in pavement application in the United States, as evidenced by more than 2.7 million miles of paved roads, of which 94 percent are surfaced with asphalt materials per a survey of National Asphalt Pavement Association (NAPA).

Along with the mass development of petroleum industry, the refineries developed the air blowing technology to produce paving grade asphalt from crude oil [2]. Nowadays, the main source of asphalt for paving applications is crude oil with a high specific gravity. Asphalt is produced from the residues of crude oil distillation process, and the asphalt properties are highly dependent on the source of crude oil and the refining process.

Generally, the neat asphalt binders without polymer modification are comprised of asphaltenes and maltenes. These components are expected to form a balanced, compatible system to show good durability for asphalt binders. Asphaltenes are mixed of paraffin-naphthene-aromatics with polycyclic structures, which comprise the most complex fraction. Polycyclic structures include sulfur, oxygen, and nitrogen and contain heterocyclic compounds with some complex metals such as nickel and vanadium. Maltenes are the hydrocarbon soluble
components that contain both neutral oils and aromatic resins. The asphaltenes solve into the maltene oils with the aromatic resins, making asphalt analogous to a colloidal system [3].

The asphalt binder is a time-temperature dependent material, which means that it exhibits different properties at different temperatures and frequencies. At high temperature and/or low frequency, the asphalt has low viscosity and susceptible to rutting. While it becomes stiff and brittle at low temperature and/or high frequency. Due to this special behavior of asphalt, the states with cold climates tend to apply relatively softer asphalt binder compared to that of the southern states to reduce the cracking potential during the winter. On the other hand, the southern state commonly utilize stiffer asphalt binder to address rutting issues. However, the asphalt refined from crude oil cannot meet the need with the increasing traffic volume and load.

To improve the quality of asphalt binder used in pavement construction, the polymer modified asphalt binder has been developed. Polymer additives basically make the asphalt binder more elastic, which means more ductile/flexible at low temperatures and stiffer at higher temperatures. Polymers additives typically used in polymer modified asphalt binder are plastomers, elastomers, and fibers. Examples are natural rubber latexes, synthetic rubber latexes (SBR), block copolymers of styrene with butadiene or isoprene (SBS), polyethylene, polypropylene and other polyolefins, and ethylene-vinyl acetate copolymers [3].

2.2. Oxidative Aging of Asphalt Binders
Aging of asphalt materials occurs since the mixture production process and lasts until the end of pavement service life. Generally, aging increases the stiffness and influences the flexibility, and brittleness properties of asphalt materials, which subsequently leads to the deterioration in cracking resistance. Considering the significant effect of aging on the long-term pavement
performance, numerous research studies have been conducted to investigate the aging of asphalt binders.

Aging of asphalt binder can be caused by many mechanisms including oxidation, polymerization, volatilization, condensation, and structural morphological changes. However, the oxidative aging has been shown to be the principal reaction responsible for hardening of asphalt in the road [4]. Standard laboratory aging protocol developed under the Strategic Highway Research Program (SHRP) was also focused on the simulation of oxidative aging in the laboratory [5, 6] by aging the asphalt materials at elevated temperatures.

It is commonly accepted that the short-term aging of asphalt mixtures occurs during production and construction due to the high temperatures involved, while the long-term aging continues throughout the pavement service life under the combined traffic and environmental loading. Petersen et al [4] investigated the relationship between viscosity and chemistry during the oxidative aging process on a group of asphalt binders from the SHRP materials library. Results indicated that the studied asphalt binders showed similar aging kinetics, with an initial rapid reaction “spurt” followed by a slower, constant rate reaction. The slow and constant rate reaction was found to be the dominant aging reaction in the field. The formation of ketones and sulfoxides was reported to be the major reason that contributed to the viscosity increase. Additionally, Petersen et al. [7] also observed that asphalt binder aging reaction “quenched” at a limiting viscosity after certain field service duration. It was indicated that asphalt aging slowed down or ceased after a certain aging level.

To simulate the field aging in laboratory, Bell et al. [8] explored that the elevated temperature and pressure of oxygen were able to accelerate the oxidative aging process of asphalt binder during SHRP study. It was also reported that the oxidative aging progression was
affected by mixture characteristics inducing aggregates absorption properties, mixture densities, and asphalt film thickness. Thus, it is still necessary to develop a laboratory aging protocol for asphalt mixtures to account these factors. The standard laboratory asphalt mixture aging procedure, AASHTO R30 [6] that developed under SHRP project, requires conditioning compacted specimens in a forced oven at 85°C for 120 hours in a laboratory to simulate the long-term field aging for performance test.

Further, Kim et al [9] completed the National Cooperative Highway Research Program (NCHRP) Project 9-54, Long-Term Aging of Asphalt Mixtures for Performance Testing and Prediction. The objective of study was aimed to develop a practical and efficient laboratory long-term aging method for asphalt mixture performance testing. This study investigated the conditioning of loose mixtures and compacted samples using the conventional forced draft oven and pressure aging vessel (PAV). Loose mixture aging in the oven at 95°C was proposed as the optimum long-term aging procedure for the performance testing by this study, which exhibited highest aging efficiency without changing chemistry of asphalt binders. Besides, the field aging levels obtained from field cores were compared against loose mix aging rates at 95°C to determine the laboratory loose mixture aging duration. Additionally, a series of laboratory aging duration maps to match 4, 8, and 16 years of field aging at depths of 6 mm, 20 mm, and 50 mm below the pavement surface under different climate conditions in the United States were developed. However, for southern locations of Louisiana, it was recommended to age the loose mixture for 27 days to simulate 16 years field aging at 6 mm below pavement surface, which is not practical for industry exercise.

Although it has been criticized to be not severe enough to simulate long-term field aging [9, 10], this standard AASHTO protocol is still the only practical standard protocol and used by
many states including Louisiana. Meanwhile, 5 days aging is considered as a simulation of certain level of aging that can be used to differentiate the quality of mixtures. Further, correlation between the cracking resistance at this aging level and field cracking performance has been validated in Louisiana [11], which means the 5 days aging is representative to Louisiana pavement aging in terms of cracking performance.

2.3. Aging Characterization of Asphalt Binder

Among these studies regarding oxidative aging, a number of methodologies have been developed to characterize aging intensities of asphalt materials.

2.3.1 Rheological Aging Characterization of Asphalt Binder

The advantages of rheological aging indexes are: (1) the rheological experiment is more efficient and accessible, which can be conducted in state or industry materials laboratory; (2) the rheological aging indexes are more directly related to the mechanistic response of asphalt materials.

In early studies [12, 13], ductility measured at reduced temperature (near 15°C) and elongation rate of 1 cm/min was reported to be a good indicator of cracking susceptibility of asphalt binders. It was generally concluded that serious cracking would occur when the ductility of asphalt binders at near 15°C, 1 c/min decreased to 3 cm or lower. However, ductility measurement is a time- and material-consuming process.

Glover [14] studied the effect of asphalt binders aging on long-term pavement cracking performance by tracking asphalt binder ductility and rheological properties during aging. This study explored the alternate measurement of unmodified asphalt binders’ ductility using rheological properties measured from DSR. A Maxwell model consisted of a spring (linear
elastic element) and a dashpot (viscous element) was used to describe the extensional behavior of asphalt binders using rheological parameters as shown in Figure 2.1.

![MAXWELL MODEL](image)

\[ \tau + \frac{\eta_e}{E} \frac{d\tau}{dt} = \eta_e \frac{d\varepsilon}{dt} \]

**Figure 2.1. The Maxwell Model: An Elastic and Viscous Element in Series[14]**

The Maxwell model was derived to be expressed in term of shear modulus \( G \) and viscosity \( \eta \). Within this Maxwell model, the elongation rate of 1 cm/min is equivalent to the strain rate of approximately 0.005 s\(^{-1}\). Moreover, it was found that the ratio of dynamic viscosity to the storage modulus (\( \eta' / G' \)) and the value of the storage modulus \( G' \) were two parameters that represent the extensional behavior. The plotted map of \( G' \) versus \( \eta' / G' \) (measured at 15°C and 0.005 rad/s) was able to identify the different aging level. Additionally, it was found that the ductility obtained from ductility test (15°C, 1 cm/min) correlated remarkably well with DSR function of \( G'/ (\eta' / G') \) (15°C, 0.005 rad/s). Based on this finding, the DSR function of \( G'/ (\eta' / G') \) was proposed to use as a surrogate for ductility of asphalt binders as the former is easier to measure and requires less material. Further, the DSR function of \( G'/ (\eta' / G') \) determined at 15°C and 0.005 rad/s was also recommended to be used to represent the aging intensities induced in asphalt binders due to its sensitivity to asphalt aging levels.
Anderson et al. [15] showed that a parameter $\Delta T_c$ (difference between $T_{critical}$ from S and m values for BBR test) essentially describes the same behavior with DSR function. A limiting value of 0.0009 MPa/sec was suggested to be an onset cracking criterial for DSR function.

It is important to note that later on Rowe [16] demonstrated that $G'/\left(\eta'/G'\right)$ was equivalent to $|G^*|\cos^2\delta/\sin\delta$ which has been referred to as the Glover-Rowe (G-R) parameter, as shown as following.

\[
\frac{G'}{\eta'} = \frac{G'}{G''} = \frac{G'}{\tan\delta} = \frac{G^*(\cos\delta)^2}{\sin\delta} \left(\frac{\omega}{\omega_g}\right) \tag{1}
\]

Note the frequency is a constant (0.005 rad/s), thus the only variables are $G^*$ and $\delta$. Besides, the typical values of phase angle $\delta$ range from 50-70°C, which resulting in 0.1-0.5 for the values for $\cos^2\delta/\sin\delta$. The weighting of $G^*$ is typically multiplied by $\cos^2\delta/\sin\delta$ ranging from 0.1-0.5. Correspondingly, the limit value becomes 180 kPa, and another limit value of 600 kPa for significant cracking was also suggested in this study.

In addition to G-R parameter, other rheological aging index properties are also under consideration. The master curve characterizes the stiffness of asphalt binders over a wide range of loading times and temperatures. A typical master curve that utilizes the complex shear modulus, $G^*$, of asphalt binder as the stiffness measurement is shown in Figure 2.2. Moreover, a mathematical model was developed that can characterize the viscoelastic properties of asphalt binder, as show in Equation (1) [17].

\[
G^* = G_g \left[1 + \left(\frac{\omega}{\omega_g}\right)^{1\log 2/R}\right]^{-R/\log 2} \tag{1}
\]
Figure 2.2. A Typical Master Curve and Physical Properties [18]

Where, $G^\ast(\omega) = \text{complex shear modulus}$; $G_g = \text{glass modulus}$ (assumed equal to 1 GPa); $\omega_r = \text{reduced frequency at the defining temperature (rad/s)}$; $\omega_c = \text{crossover frequency at the defining temperature (rad/s)}$; $\omega = \text{frequency (rad/s)}$; and $R = \text{rheological index}$. The master curve parameter ($R$ and $\omega_c$) have specific physical significance. The rheological index, $R$, is an indicator of the rheologic type. It is defined as the difference between the log of the glassy modulus and the log of the dynamic modulus at the crossover frequency. The $R$ reduces with the stuffiness. The crossover frequency, $\omega_c$, is the frequency at which the storage modulus $G'$ equal to loss modulus $G''$, or where the phase angle equal to 45°C. The modulus at crossover frequency is defined as crossover modulus. As the hardness of asphalt binder increases, crossover frequency increases. These physical parameters are used to describe the aging intensities.

2.3.2 Chemical Aging Characterization of Asphalt Binder

Several chemical analysis technologies that can be used to investigate the components and molecules transformation of asphalt binder during the oxidation aging have been identified in
this literature review. Researchers have been using the high-pressure gel permeation chromatography (HP-GPC) to study the molecular size distribution in asphalt. GPC accomplishes the separation of molecules in a sample based on the sizes, or more specifically, the hydrodynamic volumes of the molecules, a technique analogous to the aggregate sieving process in which the largest molecules elute first followed successively by smaller ones. Use of GPC opens an opportunity to examine the material from a microscopic perspective and makes it possible to correlate the macroscopic behaviors of asphalt cement and mixtures with molecular composition of asphalt binder. Aging can degrade large molecules of polymer modifier into smaller molecular sizes whereas on the other side in the base asphalt cement aging significantly increases the amount of large molecular size species and decreases those of medium and small molecular sizes [19]. The transformation of asphalt components due to the oxidation aging will provide the basis of explaining the mechanistic/physical properties changes.

Petersen [7] conducted a thorough study to understand the role of sulfoxide formation on physical properties during oxidative age hardening in asphalt binders. By evaluating the relationship between the sum of the absorbance of the ketones and sulfoxides and the logarithm of viscosity, the sulfoxides that are produced during oxidative aging was indicated to have a significant impact on viscosity, especially for asphalt with high sulfur content. Newcomb et al. [20] used the continuous performance grades (high and low temperatures) and the FTIR carbonyl area obtained from extracted binders to assess the aging equivalence between field aging due to production and/or construction and the laboratory short-term aging protocol used. It is interesting to note that the results indicated that most short-term aging of asphalt binders and mixtures occurs during plant production, while the aging induced by the construction process (i.e., transportation, laydown, and compaction) may be insignificant.
2.4. Semi-Circular Bend (SCB) Test

Cracking is considered as one major distress that reduces the service life and drives the need for rehabilitation of flexible pavements. Sufficient cracking resistance of asphalt mixture is essential and critical to minimize the cracking potential of pavement. SCB test has been developed based on fracture mechanics to evaluate the cracking resistance of asphalt mixtures.

In 1963, Paris and Erdogan et al. [21] proposed the Paris law, which indicated the relationship between the crack growth rate and the stress intensity factor in elastic materials. Based on Paris law, Rice et al. [22] developed the to represent the fracture resistance of elastic-plastic materials, such as asphalt mixture. The critical strain energy release rate is determined by the following Equation (2):

\[
J_c = -\left(\frac{1}{b}\right) \frac{dU}{da}
\]  

(2)

Where,

- \(J_c\) = critical strain energy release rate,
- \(a\) = notch depth,
- \(b\) = specimen thickness,
- \(U\) = total strain energy up to failure.

Chong and Kuruppu et al. [23] developed the semi-circular bend (SCB) test configuration to obtain the fracture toughness for rock, concrete, and ceramic materials. Mull et al. [24] introduced SCB configuration and \(J\)-integral approach into the characterization of asphalt mixture, and the \(J_c\) was explored as a fracture resistance indicator. In this study, the SCB tests were conducted on three mixtures, including chemically modified crumb rubber (CMCR) asphalt mixture, crumb rubber modified (CRM) asphalt mixture, and a control conventional mixture. For each mixture, specimens with three different notch depths (25.4 mm, 31.8 mm, 38.0 mm) were
tested with three replicates for each notch depth. It was found that the $Jc$ value of the CMCR mixture was significantly higher than that of the CRM mixture, and the CRM mixture showed a slightly higher fracture resistance than the control mixture. This study indicated the SCB test has the potential to distinguish fracture resistance of asphalt mixtures containing different materials, such as crumb rubbers.

To validate and standardize the SCB test in characterization of asphalt mixture fracture resistance, Wu et al. [25] further specified the specimen dimensions (150 mm diameter and 57 mm thickness) and testing temperature (25 °C). In this study, thirteen Superpave mixtures with various asphalt binder types, nominal maximum aggregate sizes (NMAS), and compaction levels were evaluated. It was found that the mixtures with larger NMAS and harder binders tend to have higher $Jc$ values or better fracture resistance. Additionally, a two-way ANOVA statistical analysis was conducted on test results, and it showed the $Jc$ values obtained from SCB tests were fairly sensitive to all selected variables, including binder types, NMAS and compaction level.

Elseifi et al. [26] utilized the SCB test to evaluate fracture resistance of asphalt mixtures, including mixtures contained unmodified binder (PG 64-22) and polymer-modified binder (PG70-22 M and PG 76-22M). It was found that the SCB test results indicated the mixtures contained polymer-modified binder had higher fracture resistance than mixture with unmodified binder, and mixtures with RAP are more brittle than mixtures contained no RAP. This SCB test results of this study showed the SCB test was sensitive to the content of recycled materials and binder types. Results of the SCB tests were validated by a three-dimensional finite element model to interpret and analyze the mechanisms during the SCB test.

Mohammad et al. [27] compared the $Jc$ results obtained from laboratory SCB tests with the field cracking rates retrieved from the Louisiana Pavement Management System (PMS). The
SCB test was conducted on thirteen plant mixed-laboratory compacted (PL) asphalt mixtures, which were collected as the time of construction, to obtain $J_c$ values. The field cracking rate was defined as the crack length per mile of pavement per million applications of equivalent single axle load (ESAL). It was found that approximately 58% of the field cracking rate can be explained by the factor of $J_c$, which indicated the $J_c$ has potential to correlate the field cracking performance. Further, by using the cracking index system from Louisiana PMS, Mohammad et al. [11] compared the random cracking of existing pavement sections with the $J_c$ values obtained from the corresponding field cores. Despite there are many other structural and environmental factors affecting the field performance of pavement, it was still found that the field random cracking was well correlated with the laboratory SCB results with an R$^2$-value of 0.73. This study concluded that the promising potential of using the SCB test to assess the mixtures’ cracking performance in field pavements. Additionally, this study proposed and validated that only two notch depths approach can be used to determine the critical strain energy release rate of asphalt mixtures.

2.5. Reference


CHAPTER 3. EXPERIMENTAL PROGRAM

3.1. Material Description

Five typical Louisiana Superpave asphalt mixtures were used in this study. All studied mixtures were produced by one local asphalt materials manufacturer but at two different plants. One of the mixtures was produced and acquired from the plant located in Lafayette, Louisiana and was referred as Mix 1 in this study. The field project that applied Mix 1 is located at Lafayette, Louisiana. The remaining four asphalt mixtures employed in this study (referred as Mix 2-5 accordingly, described later) were produced from the plant located in Port Allen as part of the FHWA Demonstration Project in Louisiana titled Enhanced Durability of Asphalt Pavements through Increased In-Place Pavement Density. The field project is located at US Route 190, Livingston Parish, Louisiana.

As shown in Figure 3.1, two gradations with different nominal maximum aggregate size (NMAS, 19- and 12.5-mm) were applied in this study. The top gradation showed higher passing percentage suggesting that it is finer than lower one. The mixtures with finer gradation were used for wearing course (WC) layer, while the coarser mixtures were used for binder course (BC) layer (underneath the wearing layer) in this study.

The Mix 1 was applied an unmodified binder and contained the same 12.0 mm NMAS gradation with the optimum AC content of 5.0%. For FHWA mixtures, two asphalt contents for each gradation were used in this study. For each gradation, a typical Louisiana conventional asphalt mixture was designed with the optimum asphalt content. For instance, the Mix 2 contained the WC gradation and an optimum AC content of 5.0 %, while the Mix 4 contained the 19 mm gradation and an optimum AC content of 4.8%. Two plus asphalt content (AC) mixture was designed with same gradation but 0.2% extra asphalt content than optimum AC which was
designed as a part of the scope of FHWA project [1]. Mix 3 was applied 5.2 % AC content with WC gradation, while and Mix 5 was applied 5.0 % with BC gradation. The mixtures containing extra 0.2% asphalt content were also referred to as Plus AC mixtures in this study. A styrene-butadiene-styrene (SBS) polymer-modified asphalt binder meeting Louisiana specifications for PG 76-22M was used in the FHWA mixtures [2]. Reclaimed asphalt pavement (RAP) was incorporated in the studied mixtures, and the RAP content was expressed in terms of recycled binder ratio (RBR), which is defined as the percentage of binder contributed from recycled materials in the total asphalt content. The asphalt film thickness of each mixture was calculated using surface area (based on mixture gradation) and effective asphalt content [3], as shown in Table 3.1. The wearing course (WC) mixtures were found to have a lower film thickness than that of binder course mixtures (BC), which may be contributed by the higher surface area of finer WC mixture gradation. Additionally, Mix 1 employed the same WC gradation and applied same optimum asphalt content with Mix 2 but was produced at the plant located at Lafayette with different RBR ratio, the details are showed in Table3.1.
Figure 3.1. Mixture Gradation
Table 3.1. Asphalt Mixture Composition

<table>
<thead>
<tr>
<th>Mixtures</th>
<th>Mix 1</th>
<th>Mix 2</th>
<th>Mix 3</th>
<th>Mix 4</th>
<th>Mix 5</th>
</tr>
</thead>
<tbody>
<tr>
<td>NMAS (mm)</td>
<td>12.5</td>
<td>12.5</td>
<td>12.5</td>
<td>19</td>
<td>19</td>
</tr>
<tr>
<td>$G_{mm}$</td>
<td>2.448</td>
<td>2.448</td>
<td>2.441</td>
<td>2.468</td>
<td>2.48</td>
</tr>
<tr>
<td>Total % AC</td>
<td>5.0</td>
<td>5.0</td>
<td>5.2</td>
<td>4.8</td>
<td>5.0</td>
</tr>
<tr>
<td>RAP RBR, %</td>
<td>16</td>
<td>18.0</td>
<td>17.3</td>
<td>25.0</td>
<td>24.0</td>
</tr>
<tr>
<td>% $G_{mm}$ at $N_{ini}$</td>
<td>88.5</td>
<td>88.3</td>
<td>88.3</td>
<td>88.6</td>
<td>88.8</td>
</tr>
<tr>
<td>% $G_{mm}$ at $N_{max}$</td>
<td>97.7</td>
<td>97.5</td>
<td>97.8</td>
<td>97.2</td>
<td>97.6</td>
</tr>
<tr>
<td>Air Voids (%)</td>
<td>3.5</td>
<td>3.5</td>
<td>3.5</td>
<td>3.5</td>
<td>3.5</td>
</tr>
<tr>
<td>VMA (%)</td>
<td>14.8</td>
<td>14.6</td>
<td>14.9</td>
<td>14.3</td>
<td>14.7</td>
</tr>
<tr>
<td>VFA (%)</td>
<td>76</td>
<td>76</td>
<td>77</td>
<td>76</td>
<td>76</td>
</tr>
<tr>
<td>19.0 mm (3/4 in.)</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>97</td>
<td>97</td>
</tr>
<tr>
<td>12.5 mm (1/2 in.)</td>
<td>93</td>
<td>93</td>
<td>93</td>
<td>86</td>
<td>86</td>
</tr>
<tr>
<td>9.5 mm (3/8 in.)</td>
<td>80</td>
<td>80</td>
<td>80</td>
<td>72</td>
<td>72</td>
</tr>
<tr>
<td>4.75 mm (No. 4)</td>
<td>45</td>
<td>45</td>
<td>45</td>
<td>42</td>
<td>42</td>
</tr>
<tr>
<td>2.36 mm (No. 8)</td>
<td>35</td>
<td>35</td>
<td>35</td>
<td>32</td>
<td>32</td>
</tr>
<tr>
<td>1.18 mm (No. 16)</td>
<td>27</td>
<td>27</td>
<td>27</td>
<td>23</td>
<td>23</td>
</tr>
<tr>
<td>0.600 mm (No. 30)</td>
<td>22</td>
<td>22</td>
<td>22</td>
<td>18</td>
<td>18</td>
</tr>
<tr>
<td>0.300 mm (No. 50)</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>0.150 mm (No. 100)</td>
<td>7</td>
<td>7</td>
<td>7</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>0.075 mm (No. 200)</td>
<td>5</td>
<td>5</td>
<td>5</td>
<td>4.1</td>
<td>4.1</td>
</tr>
<tr>
<td>Dust Ratio</td>
<td>0.98</td>
<td>1.02</td>
<td>0.98</td>
<td>0.87</td>
<td>0.84</td>
</tr>
<tr>
<td>$P_{be}$ (%)</td>
<td>5.0</td>
<td>4.9</td>
<td>5.1</td>
<td>4.7</td>
<td>4.9</td>
</tr>
<tr>
<td>Film Thickness ($\mu m$)</td>
<td>10.1</td>
<td>9.9</td>
<td>10.3</td>
<td>11.1</td>
<td>11.6</td>
</tr>
<tr>
<td>Surface Area ($A_s$)</td>
<td>25.34</td>
<td>25.34</td>
<td>25.34</td>
<td>21.64</td>
<td>21.64</td>
</tr>
</tbody>
</table>

Note: NMAS = Nominal Maximum Aggregate Size; $G_{mm}$ = maximum Theoretical Specific Gravity; RBR = recycled binder ratio; VMA = Voids in the Mineral Aggregate; VFA = Voids Filled with Asphalt; $P_{be}$ = effective binder percentage.

### 3.2. Sample Fabrication

#### 3.2.1. Aging Condition and Notching

Kim et al. [4] proposed loose mixture aging protocol that requires different aging durations to reflect different field aging levels, climates, and pavement depths for a given pavement location. For southern locations of Louisiana, it was recommended to age the loose mixture for 27 days to simulate 16 years field aging at 6 mm below pavement surface. The technical panel for this project recommend the use AASHTO R30 protocol for long-term asphalt mixture aging.

According to AASHTO R30 [5], laboratory aging was conducted on compacted specimens in a forced draft convection oven at 85°C. Specimens were compacted to 57-mm in
height and 150-mm in diameter with air void within 7 ± 0.5% using a Superpave gyratory compactor for Semi-circular bend (SCB) testing. After compaction, the collected plant-produced loose mixtures (PLM) were not subjected to additional aging treatment after compaction and were considered as short-term aged materials in this study. For other aging levels, the compacted specimens were subjected long-term oven aged (LTOA) for designed durations of 2, 5, and 7 days, hereafter designated as LTOA 2D, LTOA 5D, and LTOA 7D, respectively. Among these durations, the LTOA 5D was used for AASHTO R30, while other two aging durations were selected to explore and establish aging mechanism for a reasonable laboratory aging duration range.

To apply aging condition properly, some treatments were conducted on the compacted specimens prior to aging. First, each compacted specimen was cut into two semi-circular samples to improve the homogeneity for later aging conditioning. Then the cut specimens were carefully placed on flat, smooth and tough ceramic aging plates evenly to eliminate potential deformation during aging conditioning. Since the specimens were ready, the forced oven was set at 85°C at least for 1-hour prior to putting specimens into it to prevent the effect of temperature fluctuation. After the aging conditioning achieved the designed durations, turn off the oven and open the door and allow the specimens to cool down at room temperature for at least 16 hours. Note, the notching was conducted on the specimens after aging conditioning for the following reasons: (1) to avoid the deformation around notch during the aging conditioning caused by the loss of surrounding support; (2) to avoid extreme oxidation concentrated around notch that would contribute significantly to initiation of micro cracking.

Once the aging treatments were completed, semi-circular specimens were required to be notched to complete the fabrication. The test standard ASTM D 8044 [6] requires the use of
semi-circular specimens with three notch depths of 25.4, 31.8, and 38.1 mm. The middle notch is used to inspect the linearity relationship between strain energy and notch depth and was reported to not affect the calculation of $J_c$ [7]. Given the limited material availability in this study, only two notch depths of 25.4 and 38.1 mm were employed. Four test specimens were prepared and tested for each notch depth.

After notching, a quality control procedure was conducted in this study to ensure the sample fabrication quality. Specific requirement for each dimension of fabricated specimen was checked, including the notch depth, thickness, center of notch, and the height of specimen. The specimens with undesired fabrication dimension were discarded in this study.

3.3. Material Characterization

3.3.1. Intermediate-Temperature Cracking resistance characterization

The intermediate-temperature cracking resistance of asphalt mixture was evaluated using the SCB test according to ASTM D8044 [6]. Once the specimen’s fabrication was completed, SCB test was conducted on specimens with a monotonic loading rate of 0.5 mm/min at 25°C using an Asphalt Mixture Performance Tester (AMPT). The deformation and force data was recorded with 100 Hz data acquisition frequency. After testing, the curves of deformation versus force were plotted to eliminate the potential outlier specimen as shown in Figure 3.2. Then the selected curves were fitted with polynomial model to minimize the testing variation. The area under the deformation-force curves was calculated up to the peak force point as the critical strain energy.
The averaged critical strain energy values for two notch depths were used to calculate $J_c$, as shown in Figure 3.3. Eventually, the critical strain energy release rate, also known as the critical value of the J-integral ($J_c$), was calculated to represent mixtures’ cracking resistance using Equation (2) mention in Chapter 2. It is noted that the Louisiana DOTD specifications [2] require a minimum SCB $J_c$ values of 0.5- and 0.6 kJ/m$^2$ for low ($\leq$ 3 million ESALs) and high traffic volume (> 3 million ESALs), respectively.
3.3.2. Asphalt binder extraction

Following the aging treatment, asphalt binders were extracted and recovered from the mixtures with different aging levels in accordance with AASHTO T 164 – Method A and AASHTO R 59 [8]. Trichloroethylene (TCE) was used for extraction and the solutions of TCE and asphalt binder were then heated and distilled within the specific temperature range of 160° to 166°C under the protective gas environment of carbon dioxide to remove all traces of TCE. It should be noted that recovery may potentially alter asphalt binder properties due to the inability to remove all asphalt from aggregate and the residual solvent left in the binder [9, 10]. However, previous studies showed procedures used by the research team of extraction and recovery process had minimal effect on asphalt binder properties [11].

Figure 3.3. Example of Critical Strain Energy Release Rate ($J_c$) Calculation
3.3.3. Rheological characterization of extracted binders

Superpave performance grading

The extracted asphalt binders were graded using the Superpave method in accordance to AASHTO R 29 [12]. A dynamic shear rheometer (DSR) with parallel plates of 25-mm diameter and 1-mm gap was employed to determine the continuous high-temperature PG grades of the extracted asphalt binders. Two replicates were tested for each binder, and the averaged value was considered as the results.

Frequency sweep at multiple temperatures

The frequency sweep test was performed from 0.1 to 100 rad/s at multiple temperatures of 45°, 35°, and 15°C using a DSR with parallel plates of 8-mm diameter and 2-mm gap. During the test, the strain level was controlled at 1% to ensure the asphalt binder was in the linear viscoelastic range. The obtained test data from the frequency sweep test was used to construct the master curves for dynamic shear modulus and phase angle, from which the effects of various aging intensities on the stiffness and elasticity properties were examined and quantified.

3.3.3. Chemical characterization of extracted binders

Gel Permeation Chromatography (GPC)

Asphalt chemistry is extremely complicated. Researchers have been using the high-pressure gel permeation chromatography (HP-GPC) to study the molecular size distribution in asphalt. GPC accomplishes the separation of molecules in a sample based on the sizes, or more specifically, the hydrodynamic volumes of the molecules, a technique analogous to the aggregate sieving process in which the largest molecules elute first followed successively by smaller ones. Use of GPC opens an opportunity to examine the material from a microscopic perspective and makes it
possible to correlate the macroscopic behaviors of asphalt binders and mixtures with molecular composition of asphalt binder.

In this study, GPC was conducted using an EcoSEC high-performance GPC system (HLC-8320GPC) of Tosoh Corporation with a differential refractive index detector (RI) and UV detector. The separation of asphalt components was performed with a series of four microstyragel columns of pore sizes 200 angstrom (Å), 75 Å (2 columns), and 30 Å from Tosoh Bio-science., Tetrahydrofuran (THF) was used as the solvent using a flow rate of 0.35 mL/min. The apparent molecular weight distribution based upon polystyrene standards was divided into four fractions: high molecular weight polymeric species (HMW, 45K to greater than 300K Daltons); associated asphaltenes (MMW, 45K to 19K Daltons), asphaltenes (19K to 3K Daltons), and low molecular weight maltenes (LMW, less than 3K Daltons).

3.4. References


12. AASHTO R 29, Standard Practice for Grading or Verifying the Performance Grade (PG) of an Asphalt BinderAmerican Association of State Highway and Transportation OfficialsWashington, DC 2015
CHAPTER 4. RESULTS AND ANALYSIS

4.1. SCB Test Results

The SCB test results are shown in Figure 4.1. The coefficient of variation (COV) ranged from 8.1% to 18.9% with an overall average of 13.6%, which indicated a good test repeatability. Tougher asphalt mixtures with higher cracking resistances are expected to provide higher \( J_c \) values. It was found that the \( J_c \) values of all mixtures generally decreased with the aging duration, indicating the progressive deterioration in cracking resistance. Similar findings regarding the effect of aging on cracking resistance have been reported elsewhere [1, 2].

In order to discriminate the cracking resistance of evaluated mixtures, the one-way ANOVA statistical analysis using Tukey comparison was performed on SCB \( J_c \) results. The letters A, B, and C were used to show statistically distinct crack resistance from best to worst. Letter A was assigned to the highest mean followed by the other letters in appropriate order. A double letter designation, such as A/B indicates that the difference in the means is not clear-cut, and that the mean is close to either group in the analysis. As shown in Table 4.1 (b), \( J_c \) values of mixtures with same designed gradation were compared at different aging levels. For instance, \( J_c \) values of Mix 1, 2, and 3 containing WC gradation were compared at each aging level. Correspondingly, \( J_c \) values of Mix 4 and Mix 5 were compared at each aging level for BC gradation mixtures.

For WC gradation mixtures, it was observed that the Mix 3 exhibited significant higher \( J_c \) value compared to that of Mix 1 and Mix 2 at all aging levels. Additionally, although Mix 1 exhibited comparable \( J_c \) value at PLM aging level, it showed significant lower \( J_c \) value than that of Mix 2. The results indicated that the 0.2% extra AC content significantly improved the cracking resistance and aging susceptibility for WC gradation mixtures. For BC gradation
mixtures, the Mix 5 generally showed comparable $J_c$ value with Mix 4 except at LTOA 2D aging level. It may indicate that the extra AC content was beneficial for cracking resistance improvement, while the improvement was also affected by the gradation of the mixture.

Besides, the $J_c$ values at various aging levels of each mixture were statistically analyzed as shown in Table 4 (a). In general, the SCB $J_c$ values were significantly higher for PLM mixtures (short term aging) as compared to other adding levels, and it was also observed that the $J_c$ results of LTOA 5D were comparable to those of LTOA 7D for all four mixtures, suggesting that the aging effect on cracking resistance started to stabilize after 5 days aging. This observation is similar to the ones presented in Figure 4.1. In addition, the $J_c$ values of plus AC mixtures at LTOA 2D aging were found were comparable with PLM aging mixtures, while the conventional mixtures with optimum AC contents showed significant reduction. This may be attributed to the oxidative aging process requiring longer time to penetrate the thicker asphalt film as a result of the 0.2% extra AC content. Comparatively, the aging effect on the conventional mixtures with thinner asphalt film was faster to reach the steady state.

![Figure 4.1. Semi-Circular Bend Test Results at 25 °C.](image-url)
Table 4.1. Statistical Analysis of SCB \( J_c \) Results

(a) SCB \( J_c \) statistical comparison at various aging levels for each mixture

<table>
<thead>
<tr>
<th>Mixture Type</th>
<th>PLM</th>
<th>LTOA 2D</th>
<th>LTOA 5D</th>
<th>LTOA 7D</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mix 1</td>
<td>A</td>
<td>B</td>
<td>B</td>
<td>B</td>
</tr>
<tr>
<td>Mix 2</td>
<td>A</td>
<td>B</td>
<td>B</td>
<td>B</td>
</tr>
<tr>
<td>Mix 3</td>
<td>A/B</td>
<td>B</td>
<td>B</td>
<td>B</td>
</tr>
<tr>
<td>Mix 4</td>
<td>A</td>
<td>B</td>
<td>B</td>
<td>B</td>
</tr>
<tr>
<td>Mix 5</td>
<td>A</td>
<td>A</td>
<td>B</td>
<td>B</td>
</tr>
</tbody>
</table>

Note: Statistical grouping results are indicated by letters A, B, and C, representing significantly distinct crack resistance from best to worst.

(b) SCB \( J_c \) comparison of conventional and plus AC mixtures at each aging level

<table>
<thead>
<tr>
<th>Aging Level</th>
<th>Mix 1</th>
<th>Mix 2</th>
<th>Mix 3</th>
<th>Mix 4</th>
<th>Mix 5</th>
</tr>
</thead>
<tbody>
<tr>
<td>PLM</td>
<td>B</td>
<td>B</td>
<td>A</td>
<td>A</td>
<td>A</td>
</tr>
<tr>
<td>LTOA 2D</td>
<td>C</td>
<td>B</td>
<td>A</td>
<td>B</td>
<td>A</td>
</tr>
<tr>
<td>LTOA 5D</td>
<td>C</td>
<td>B</td>
<td>A</td>
<td>A</td>
<td>A</td>
</tr>
<tr>
<td>LTOA 7D</td>
<td>C</td>
<td>B</td>
<td>A</td>
<td>A</td>
<td>A</td>
</tr>
</tbody>
</table>

Note: The comparison was between mixtures with same gradation (Mix 1-3 for WC mixes; Mix 4 and 5 for BC Mixes).

4.2. Rheological Characterization Results

4.2.1. Continuous high-temperature PG results

The continuous high temperature PG grade of each binder was determined by averaging results of two 25-mm samples. The difference between two replicates was found within the range of 0-0.4°C indicating consistence in terms of repeatability. It needs to be noted that the extracted binders were incorporated with recycled binder from RAP with virgin PG 76-22m binder. Table 4.2 presents the continuous high PG grading results of the asphalt binders extracted from plant-produced loose mixture and laboratory aged compacted specimens. As shown in Table 4.2, asphalt binders showed a consistent increasing trend when aging duration increased for mixtures
evaluated, except that the Mix 2 mixture at LTOA 7D aging level was found slightly lower than that of LTOA 5D. However, the difference between LTOA 5D and 7D aged mixtures (0.1°C) was negligible, which may be due to testing variability.

The binders extracted from Mix 1 showed the lowest PG grade at all aging levels in comparison with other mixtures. It might be attributed to the resource of RAP and asphalt binders. Further investigation was conducted by using following chemical analysis.

On the other hand, the other four mixtures were produced at same plant at adjacent time using the same source of RAP and asphalt binder. Thus, the comparison between the four FHWA mixtures could eliminate the effect of production. It is noted that asphalt binders extracted from WC mixtures showed higher high PG grade than that of BC mixtures at all aging durations. The asphalt film thickness was used to account for this observation. As shown in Table 3.1, the film thicknesses of WC mixtures were found to be lower than those of BC mixtures, which may indicate the WC mixtures were more susceptible to aging and hence the observed higher high PG results. However, it is found that the increasing trend of high PG grade generally stabilized after LTOA 5D aging.

Table 4.2. Continuous High PG Grade of Extracted Binder

<table>
<thead>
<tr>
<th>Mixture Type</th>
<th>PLM</th>
<th>LTOA 2D</th>
<th>LTOA 5D</th>
<th>LTOA 7D</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mix 1</td>
<td>75.7</td>
<td>77.5</td>
<td>78.5</td>
<td>78.6</td>
</tr>
<tr>
<td>Mix 2</td>
<td>83.1</td>
<td>83.8</td>
<td>85.8</td>
<td>85.7</td>
</tr>
<tr>
<td>Mix 3</td>
<td>82.1</td>
<td>83.8</td>
<td>85.3</td>
<td>85.6</td>
</tr>
<tr>
<td>Mix 4</td>
<td>81.8</td>
<td>82.2</td>
<td>84.0</td>
<td>84.0</td>
</tr>
<tr>
<td>Mix 5</td>
<td>80.4</td>
<td>82.9</td>
<td>83.5</td>
<td>84.3</td>
</tr>
</tbody>
</table>

Note: PLM LTOA = long-term oven aging.
4.2.2. Frequency sweep results and G-R parameter

Figures 4.2-4.7 present the dynamic shear modulus (G*) and phase angle master curves for the four extracted asphalt binders. The master curves were constructed by fitting the shifted dynamic shear moduli |G*| to the Christensen-Anderson (CA) model [3] using the Excel solver function. Generally, the stiffness of asphalt binders increased with the aging duration. However, the difference in master curves of asphalt binders extracted from LTOA 5D and LTOA 7D aged mixtures was found negligible for all mixtures. Additionally, the LTOA 2D master curves were distinguishable from LTOA 5D and LTOA 7D master curves for the Mix 1-3. However, for the Mix 4 and 5 mixtures, difference among the three aged master curves (LTOA 2, 5, and 7D) was visually indiscernible.
Figure 4.2. $|G^*|$ and Phase Angle Master Curves of Binder Extracted from Mix 1 at Various Aging Levels

Figure 4.3. $|G^*|$ and Phase Angle Master Curves of Binder Extracted from Mix 2 at Various Aging Levels
Figure 4.4. $|G^*|$ and Phase Angle Master Curves of Binder Extracted from Mix 3 at Various Aging Levels

Figure 4.5. $|G^*|$ and Phase Angle Master Curves of Binder Extracted from Mix 4 at Various Aging Levels
Figure 4.6. $|G^*|$ and Phase Angle Master Curves of Binder Extracted from Mix 5 at Various Aging Levels

Using the master curves shown in Figure 4.2-4.6, the $|G^*|$ and $\delta$ values at 15°C and 0.005 rad/s of the asphalt binders were obtained. For each master curve, a fitted CA model with specific physical parameters were determined, as shown in Equation (2). As shown in Figure 4.2, the $G^*$ and phase angle value at the target frequency of 0.005 rad/s at 15°C was obtained. Then, the G-R parameter was determined using Equation (3), which was derived by Rowe [4].

$$G-R \text{ Parameter} = \frac{|G^*| \times \cos^2 \delta}{\sin \delta}$$ (3)

Figures 4.7-4.11 present the G-R parameters in the black space, where the $|G^*|$ values at 15°C and 0.005 rad/s of the extracted asphalt binders were plotted against the corresponding $\delta$ values. The black space diagram is a rheological plot of $|G^*|$ and phase angle $\delta$. Anderson et al. [5] proposed G-R thresholds of 180 kPa when non-load associated cracking begins and 600 kPa when there are significant cracking problems. Note the thresholds were proposed based on early Khandal study [6], in which PG 58-28 under Pennsylvanian climate condition. They might not
applicable to current PG 76-22 asphalt under Louisiana condition, while they were used as references and the G-R parameters were used as an aging tracking index in this study. A Black diagram is a graph of the magnitude (norm) of the complex modulus, G*, versus the phase angle, \( \delta \), obtained from a dynamic test. The two thresholds of G-R parameter were plotted at various phase angle \( \delta \) to create damage curves in black space, and the zone between these two curves is referred to as damage zone, as shown in Figures 4.7-4.11.

The black space plot of Mix 1 showed significant spread pattern and longer “travel” distance compared to other mixtures. The binders extracted from Mix 1 showed higher phase angle value of 67.8 at PLM aging level. However, it reduced to around 60°C after 2D aging, and further reduced to 57.2 eventually at LTOA 7D aging level. This behavior indicated that the binder of Mix 1 was more viscous at PLM aging level, but it was susceptible to laboratory aging and undertaken significant amount of oxidation aging effect. For four FHWA mixtures, it was observed that the WC mixtures plots were located closer to damage zone than BC mixtures at each aging level, which indicated the WC mixtures were more prone to cracking in terms of asphalt binder. A consistent trend was observed in that mixtures subjected to higher aging durations were located closer to the upper left corner in the black space diagram, indicating an increased stiffness and reduced ductility after aging. Moreover, for all four studied asphalt mixtures, it was noticed that the plots of LTOA 5D and LTOA 7D aging levels located at close position, indicating produced similar aging effects.
Figure 4.7. G-R Parameter Results in Black Space Diagram for Mix 1

Figure 4.8. G-R Parameter Results in Black Space Diagram for Mix 2
Figure 4.9. G-R Parameter Results in Black Space Diagram for Mix 3

Figure 4.10. G-R Parameter Results in Black Space Diagram for Mix 4
Figure 4.11. G-R Parameter Results in Black Space Diagram for Mix 5

Figure 4.12 presents the G-R parameters determined for all the mixtures along with the crack initiation threshold of 180 kPa. It was observed that G-R increased with the aging level for all mixtures, indicating the loss of ductility during the aging process [5]. Similar to the trend of continuous high PG grade results, the increasing trend of G-R stabilized after LTOA 5D. As expected, the Mix 1 exhibited lowest G-R parameter at PLM aging level but increased rapidly after aging conditioning was applied. Compared to the conventional mixtures, the plus AC mixtures were found to have lower G-R before and after long-term aging for both gradations, which indicated that the extra asphalt content was beneficial in maintaining the ductility of asphalt materials to a certain extent. The observation of more aging intensities induced in the WC mixtures as indicated by G-R parameters could be explained by the lower asphalt film thickness, Table 3.1. Results also indicated that the G-R parameter was able to capture the effect of extra asphalt content.
In addition to Glover-Rowe parameter, other rheological aging indexes were evaluated, patriotically these physical parameters obtained from master curves. The R index generally increased with aging ranging from 2.36-2.48 for modified asphalt binders and 1.87-2.37 for unmodified Mix1. The crossover frequency decreased with aging ranging from 0.38-1.17. The narrow ranges made them not suitable to be used as aging index track the aging intensities. On the other hand, the crossover modulus values corresponding to crossover frequency were calculated by inputting crossover frequency into the fitted CA model. As shown in Figure 4.13, the crossover modulus decreased along with aging. Except Mix 1 showed significant drop in crossover modulus, the decrease in other mixtures was found not significant in term of the

Figure 4.12. G-R Results at Different Aging Levels
magnitude. Thus, in the development of aging factor, crossover modulus was not used as an aging index.

![Crossover Modulus at Different Aging Levels](image)

Figure 4.13. Crossover Modulus at Different Aging Levels
4.3. Chemical Characterization Results

4.3.1. GPC test results

Gel permeation chromatography (GPC) identified the changes that occur in virgin asphalt blends upon the addition of crumb rubber and other additives. The use of GPC is well established as a procedure for following these modifications [7, 8]. GPC uniquely mirrors the quantitative distribution of all species present in a binder, such as maltenes, asphaltenes, and polymers. The instrument signal, viz., the difference between the refractive indices of the eluting solution containing the asphalt and that of the solvent (ΔRI), is plotted versus the eluting volume (mL), the molecules of larger size are excluded first, allowing the differentiation of asphalt species on the scale of apparent MW = 1M-100 Daltons. A correlation of the eluting volume with the apparent molecular weight of the eluting fraction is achieved using narrow molecular weight standards. Efforts to predict the properties of asphalts using GPC have been reported. Rather than estimate the actual molecular weight of the eluting fractions, the GPC chromatograms have been divided into three regions: large molecular size (LMS), medium molecular size (MMS), and small molecular size (SMS). Researchers stated that the LMS and SMS regions are significant with respect to predicting pavement performance [9-13]. As shown in the following, using molecular weight regions, it is possible to divide the LMS fraction into ranges which change when the asphalt ages or is modified.

When the calibration curve is made using polystyrene standards GPC a chromatogram can be divided into four slices based on the apparent molecular weight of the eluting species: Fractions of apparent molecular weights greater than 45,000 Daltons (representing polymers), of apparent molecular weights 45,000>MW>19,000 (representing low MW polymer species and associated asphaltenes), of apparent molecular weights from 19,000 to 3,000 (representing asphaltenes), and
molecular weights less than 3000 (representing maltenes). Figure 4.14 shows the GPC chromatograms of PG 76-22 asphalt binder divided in this way. Quantitative data can be obtained (using Origin software, https://www.originlab.com) by determining the area under the curve [7]. Another way is to deconvolution of the GPC chromatogram to determine the contributions of the asphalt components. This gives more information about the different molecular weight species embedded in the curve as shown in Figure 4.14. Both types of quantification were used to understand various parameters.

Figure 4.14. Determination of Maltenes and Asphaltenes Content of PG 76-22 Binder Based on the Molecular Weight Regions of The GPC Curve.
Earlier determinations by osmometry indicated that the average molecular weight MW of maltenes (as heptane soluble binder fraction) is 700-900 Daltons and that of asphaltenes (as heptane insoluble binder fraction) ranges between 2,000 and 10,000 Daltons [14]. These MW data have been confirmed by GPC method which became a routine technique in Louisiana for analysis of asphalt binders[7]. Since the MW of polymers used in asphalt industry is higher than 10,000 Daltons, the polymer and asphalt components of polymer modified asphalt cements could be separated completely with accurate determination of molecular weight of species achieved by calibration with standard polystyrenes of narrow MW, as shown in Figure 4.15.

Asphaltenes, as the higher MW component, by virtue of their molecular size are the bodying agent for the maltenes and have a significant influence on asphalt performances [9]. The largest "molecules" are assemblies of smaller molecules held together by one or more intermolecular forces. The polarity of the solvent used in the analysis probed the ability of the samples to undergo self-assembly by different interactive mechanisms[10]. Therefore, by
analyzing distribution of components of asphalt paving combinations, such as asphalt cements and binders, with or without polymers (SB, SBS or crumb rubber), one might correlate the physical performance of mixtures containing these materials with the content and MW magnitude of asphalt species.

Table 4.3 presents the GPC results of the extracted asphalt binders at different aging levels. In general, aging of the mixtures was accompanied by a reduction of maltenes content, with a corresponding increase in asphaltenes and the associated asphaltenes species as well as a reduction of the species.

First of all, it was surprising to find the percentages of high molecular weight (HWM) of binders extracted from Mix 1 at different aging levels were distinguishably low. According to literature review and consulting with chemical experts, the HWM percentage for modified binder is commonly within the range of 1.5-2%. The GPC chemical characterization for Mix 1 indicated that the binder applied was not appropriately modified with polymer by manufacturer. Lower cracking resistance and higher aging susceptibility was expected to be seen for Mix 1. To verify this observation, a comparison was conducted as shown in Figure 4.16. In the HMW region, there was not molecular pieces were detected by GPC test.
The asphalt binders extracted from the other WC mixtures exhibited slightly higher HMW species content than those of the BC mixtures at all aging levels. This observation may be explained by the lower RBR of the WC mixtures (in other word, higher modified virgin asphalt binder ratio), as shown in Table 3.1, since the base binder (PG 76-22m) was polymer modified. The HMW species were found decreased with the aging durations, while the asphaltenes exhibited increasing trend. Meanwhile, the percentages of maltenes generally decreased with an increase in aging level. The results were found to be consistent with other studies [15, 16].
Table 4.3. GPC Test Results

<table>
<thead>
<tr>
<th>Mixture</th>
<th>Aging Level</th>
<th>HMW* Polymer 300K-45K, %</th>
<th>Associated Asphaltenes 45-19K, %</th>
<th>Asphaltenes 19-3K, %</th>
<th>Maltenes &lt; 3K, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mix 1</td>
<td>PLM</td>
<td>0.25</td>
<td>0.86</td>
<td>19.77</td>
<td>79.12</td>
</tr>
<tr>
<td></td>
<td>LTOA-2D</td>
<td>0.31</td>
<td>1.36</td>
<td>20.93</td>
<td>77.12</td>
</tr>
<tr>
<td></td>
<td>LTOA-5D</td>
<td>0.35</td>
<td>1.41</td>
<td>23.17</td>
<td>75.08</td>
</tr>
<tr>
<td></td>
<td>LTOA-7D</td>
<td>0.40</td>
<td>1.39</td>
<td>23.34</td>
<td>74.87</td>
</tr>
<tr>
<td>Mix 2</td>
<td>PLM</td>
<td>3.01</td>
<td>2.96</td>
<td>22.1</td>
<td>71.94</td>
</tr>
<tr>
<td></td>
<td>LTOA-2D</td>
<td>2.93</td>
<td>3.15</td>
<td>22.8</td>
<td>71.16</td>
</tr>
<tr>
<td></td>
<td>LTOA-5D</td>
<td>2.82</td>
<td>3.22</td>
<td>23.1</td>
<td>70.88</td>
</tr>
<tr>
<td></td>
<td>LTOA-7D</td>
<td>2.55</td>
<td>3.49</td>
<td>23.3</td>
<td>70.65</td>
</tr>
<tr>
<td>Mix 3</td>
<td>P-PLM</td>
<td>3.03</td>
<td>3.13</td>
<td>22.7</td>
<td>71.09</td>
</tr>
<tr>
<td></td>
<td>LTOA-2D</td>
<td>2.91</td>
<td>3.07</td>
<td>22.7</td>
<td>71.36</td>
</tr>
<tr>
<td></td>
<td>LTOA-5D</td>
<td>2.75</td>
<td>3.36</td>
<td>23.1</td>
<td>70.81</td>
</tr>
<tr>
<td></td>
<td>LTOA-7D</td>
<td>2.69</td>
<td>3.55</td>
<td>23.9</td>
<td>69.91</td>
</tr>
<tr>
<td>Mix 4</td>
<td>PLM</td>
<td>2.67</td>
<td>3.26</td>
<td>21.3</td>
<td>72.78</td>
</tr>
<tr>
<td></td>
<td>LTOA-2D</td>
<td>2.45</td>
<td>3.34</td>
<td>21.9</td>
<td>72.29</td>
</tr>
<tr>
<td></td>
<td>LTOA-5D</td>
<td>2.32</td>
<td>3.55</td>
<td>22.9</td>
<td>71.23</td>
</tr>
<tr>
<td></td>
<td>LTOA-7D</td>
<td>2.37</td>
<td>3.75</td>
<td>23.2</td>
<td>70.63</td>
</tr>
<tr>
<td>Mix 5</td>
<td>PLM</td>
<td>2.90</td>
<td>3.42</td>
<td>22.1</td>
<td>71.55</td>
</tr>
<tr>
<td></td>
<td>LTOA-2D</td>
<td>2.91</td>
<td>3.41</td>
<td>22.3</td>
<td>71.33</td>
</tr>
<tr>
<td></td>
<td>LTOA-5D</td>
<td>2.66</td>
<td>3.62</td>
<td>22.6</td>
<td>71.10</td>
</tr>
<tr>
<td></td>
<td>LTOA-7D</td>
<td>2.39</td>
<td>3.56</td>
<td>22.9</td>
<td>71.16</td>
</tr>
</tbody>
</table>

Note: HMW = high molecular weight; LTOA = long-term oven aging.
4.4. Development of Aging Factor and Cracking Resistance Prediction Model

4.4.1. Development of Aging Factor

The G-R parameter describes the ductility property of asphalt binders, which relates closely to cracking resistance. Moreover, the G-R parameter has been used as an aging index to track the aging intensities induced in asphalt binders. The correlation between the asphalt binder aging index and fracture properties of asphalt mixtures has not been extensively explored. In this study, the relationship between the G-R parameter and intermediate-temperature cracking resistance of asphalt mixtures as represented by \( J_c \) was examined.

Noted that, Mix 1 was considered separately because the unmodified asphalt binder was used. Mix 2 and Mix 3 were grouped for the similar mixture characteristics, while the Mix 4 and Mix 5 were grouped together. As shown in Figure 4.17, strong correlations between G-R parameters and SCB \( J_c \) values were observed for each group. The \( R^2 \) values for each group were 0.94, 0.89 and 0.86, respectively.

![Figure 4.17. Correlation between G-R Parameter and SCB \( J_c \)](image)
It was interesting to note that mixtures with different gradations showed different relationships between G-R and \(J_c\). However, the Mix 1, Mix 2, and Mix 3 that contained same gradation showed similar decreasing rate in SCB \(J_c\) alone with the increase of G-R parameter (-0.0045 versus -0.0044). This observation indicated that the mixture gradation might be the dominate factor that affected sensitivity of cracking resistance to the aging intensities. Within the mixtures evaluated, cracking resistance of the mixtures containing BC gradation were found to be more susceptible to asphalt binder aging. Meanwhile, Mix 2 and 3 exhibited higher \(J_c\) values Mix 1 compared to at same G-R parameter value, which might attribute to the effect of polymer modification in asphalt binders.

Based on this observation, G-R parameter was considered as a reliable and cracking-sensitive aging index and was utilized to develop an aging factor. As shown in Equation (4), the proposed aging factor was calculated as following:

\[
\text{Aging Factor (G-R)} = \frac{\text{aged G-R parameter}}{\text{unaged G-R parameter}}
\]

The aging factors based on G-R parameter was calculated for each mixture shown in Figure 4.18. Mix 1 showed significant higher G-R aging factor of 4.0 compared to that of Mix 2-4, which might indicate that the higher aging susceptibility for unmodified asphalt binder. For mixtures containing modified binders, the aging factor based on G-R parameters ranges from 1.5 to 1.8 with an average of 1.62.
To evaluate the feasibility of G-R aging factor, the aged SCB $J_c$ values were predicted by multiplying unaged SCB $J_c$ (PLM) with corresponding aging factor, and then compared to the measured aged SCB $J_c$. As shown in Figure 4.19, mixtures containing modified asphalt binders (Mix 2-4) showed comparable prediction SCB $J_c$ to measured ones, while the Mix 1 with unmodified binder exhibited significant difference.

![Figure 4.18. G-R Aging Factors for SCB $J_c$](image)

![Figure 4.19. Comparison of Measured and Predicted SCB $J_c$](image)
Figure 4.20 illustrates the correlation between measured and predicted SCB $J_c$. This figure shows a good correlation. The root-mean-squared error (RMSE) was also calculated as a measure of the difference between the predicted and measured observations according to the following Equation (5):

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (y_i - y_p)^2}$$

(5)

Where,

- RMSE=root-mean-squared error;
- $y_i$=measured value;
- $y_p$= predicted value; and
- $n$= number of observations.

The calculated RMSE was 0.09 kJ/m$^2$, which indicated a good predictive accuracy in comparison with the SCB $J_c$ values ranging from 0.37-0.71 kJ/m$^2$.
Based on these observations, mixtures applied unmodified and modified binders showed different magnitude G-R aging factor of values. This might indicate that the polymer modification affects the aging process. Thus, aging factor should be developed separately for mixtures applied unmodified and modified binder. The G-R aging factor of mixtures applied modified binders were within a narrow range (1.6-1.8), the averaged value of 1.62 was then used for all polymer modified mixtures.

As shown in Figure 4.21, mixtures containing modified asphalt binders (Mix 2-4) showed comparable prediction SCB $J_c$ to measured ones. Additionally, Figure 4.22 illustrates the good correlation between measured and predicted SCB $J_c$ with a RMSE of 0.07 kJ/m$^2$.

![Figure 4.21. Comparison of Measured and Predicted SCB $J_c$](image)
Figure 4.22. Predicted SCB $J_c$ versus Measured SCB $J_c$ (Averaged Aging Factor)

According to above discussion, an aging factor based on G-R parameter of 1.62 was proposed to transfer unaged SCB $J_c$ to aged SCB $J_c$ (LTOA 5D) for mixture containing polymer modified binders.
4.4.2. Development of Preliminary Cracking Resistance Prediction Model

Based on the above observations, it was motivated to the strong correlation to preliminarily develop a general function that describe the relationship between G-R Parameter and SCB $J_c$ by considering the potential affecting variables including surface area (count for gradation factor) and polymer content (asphalt polymer modification). After approximately 30 different model forms were attempted, the following Equation (6) represents the most accurate non-linear model form that was developed to predict the $J_c$:

$$J_c = 0.8 + 0.15 \times P - \frac{1500 \times GR}{A_s^4}$$

where,

$J_c$ = predicted critical strain release rate, kJ/m$^2$;

$P$= percentage of HMW in asphalt binder;

$A_s$= surface area of the mixture gradation, m$^2$;

GR = G-R parameter, kPa.

The parameters used for model development were obtained from mixtures at aging level of PLM to LTOA 7D, and the values of these parameters are as follows:

Percentage of polymer content is between 0-3.0 % including an unmodified asphalt;

G-R parameter is between 30-171 kPa;

SCB $J_c$ is between 0.36-0.95;

Figure 4.23 illustrates the correlation between the predicted and measured $J_c$ based on the developed non-linear regression model. The model showed an excellent $R^2$ value of 0.92 with an RMSE value of 0.05 kJ/m$^2$. Recall that the range of measured $J_c$ values used for model development was between 0.36 and 0.95 kJ /m$^2$ representing cracking resistance at unaged and significantly aged status. Similarly, a 95% prediction interval, which indicates the
range of values that is likely to contain the response value of a single new observation in the model, was developed. The root-mean-squared error (RMSE) was also calculated as a measure of the difference between the predicted and measured observations according to the Equation (5). The calculated RMSE was 0.05 kJ/m², which indicated a good predictive accuracy in comparison with the SCB $J_c$ values ranging from 0.36-0.71 kJ/m². The validation of the preliminary cracking resistance prediction model needed.

![Figure 4.23. Predicted SCB $J_c$ versus Measured SCB $J_c$ (Prediction Model)](image)

4.4.3. Preliminary Validation on Cracking Resistance Prediction Model

To validate the developed $J_c$ prediction model, data from literature was used. Due to the model being developed for the asphalt binder with G-R parameter ranging from 30-171 kPa, only the asphalt binders containing G-R parameter within this range were applied for the validation. Four mixtures with different binder types, recycled materials content, and application of Warm-mix technology were selected from published papers [17, 18]. The cracking resistance SCB $J_c$, gradation, HMW (GPC) percentage, and G-R parameter information was obtained as shown in Table 4.4. Note that the G-R parameter was calculated based on the master curves, and the
surface area values were calculated based on the mixtures gradation using same methodology in this study. The predicted SCB $J_c$ values were compared measured SCB $J_c$ values, and the comparison was shown in Figure 4.24. The calculated RMSE was 0.08 kJ/m$^2$, which was comparable with RMSE of 0.05 kJ/m$^2$ from model development.

Table 4.4. Mixture Characteristics for Model Validation

<table>
<thead>
<tr>
<th>Mixture</th>
<th>Surface Area</th>
<th>HMW, % ($&gt;=$ 45K Dalton)</th>
<th>G-R Parameter, kPa</th>
<th>Measured $J_c$, kJ/m$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lane 3 64-22 20% RAS HMA</td>
<td>26.40</td>
<td>0.77</td>
<td>176</td>
<td>0.38</td>
</tr>
<tr>
<td>Lane 4 64-22 20% RAP WMA Evotherm</td>
<td>24.97</td>
<td>0.59</td>
<td>133</td>
<td>0.46</td>
</tr>
<tr>
<td>Lane 8 58-28 40% RAP HMA</td>
<td>23.96</td>
<td>1.14</td>
<td>133</td>
<td>0.47</td>
</tr>
<tr>
<td>Lane 11 58-28 40% RAP WMA Evotherm</td>
<td>24.47</td>
<td>1.24</td>
<td>120</td>
<td>0.59</td>
</tr>
</tbody>
</table>

Figure 4.24. Predicted SCB $J_c$ versus Measured SCB $J_c$ (Validation)
4.6. References


CHAPTER 5. SUMMARY AND CONCLUSION

Oxidative aging progression of asphalt binder dictates the deterioration of asphalt mixtures’ cracking resistance. This study evaluated the effect of laboratory aging on the intermediate-temperature cracking resistance of asphalt mixtures, and chemical and rheological properties of asphalt binders. Cracking resistance of asphalt mixtures with different aging levels (0-, 2-, 5-, and 7 days at 85°C) was assessed by SCB test. Rheological and chemical properties of the extracted asphalt binders were evaluated using the DSR and GPC tests, respectively. The following conclusions were drawn based on the findings in this study:

• SCB $J_c$ values indicated the cracking resistance of asphalt mixtures decreased with aging, while the SCB $J_c$ values at LTOA 5D aging level were comparable with that of LTOA 7D aging level. Further, the plus AC mixtures generally exhibited higher SCB $J_c$ values than conventional mixtures at the optimum AC content at each aging level.

• The G-R parameter revealed loss of ductility of asphalt binders during the aging process while the G-R parameters at LTOA 5D aging level were comparable with that of LTOA 7D aging level. Additionally, it was captured that the plus AC mixtures showed higher ductility compared to the conventional mixtures.

• GPC test results showed that aging yielded constantly increased asphaltenes and reduced maltenes fractions in the asphalt binder composition. The degradation of polymer degradation was also observed in for polymer modified binders in this study.

• A strong correlation between the asphalt binder G-R parameter and the mixture SCB $J_c$ parameter was observed. This relationship was found to be dependent on mixture’s gradation. The G-R parameter demonstrated the potential to be used as an aging index to
track the aging intensities in asphalt binders and correlate with cracking resistance of asphalt mixtures.

- An aging factor based on G-R parameter was proposed to be 1.62 for mixtures containing polymer modified binder. A preliminary cracking resistance prediction model was developed based on the aging index of G-R parameter and other affecting mixture characteristics. Good agreement between the measured and predicted cracking resistance SCB $J_c$ values was observed based on preliminary validation.

5.1 Future Work

Although a preliminary aging factor and cracking resistance prediction model was proposed based on the G-R parameter for asphalt mixtures’ cracking resistance, the following investigations are recommended:

- The developed aging factor and cracking resistance prediction model must be validated by asphalt mixtures with different characteristics. The mixture variability should include but not limited to:
  a) Asphalt binders with different grade (e.g. PG76-22, PG 70-22; and PG 67-22)
  b) Different gradation;
  c) Application of additives:
     - Warm-mixed asphalt mixtures;
     - Crumb rubber modified binders;
     - Re-refined engine oil bottom (REOB).

- Complete the chemical experimental including FTIR to fully understand the effect of laboratory aging on asphalt binders’ chemical properties. A chemical aging factor might be explored.
• Note the surface area of mixture gradation was used a surrogate for surface free energy to represent the aggregate-asphalt adhesion. The surface free energy ought to be measured in further study to verify the effect on cracking resistance.
VITA

Yucheng Shi was born in 1994 in Tianchang, Anhui Province, China. In 2016, he finished his Bachelor of Science in Civil Engineering from Hefei University of Technology. Motivated to obtain further and various education, he made the decision to join graduate school in the Department of Civil and Environmental Engineering at Louisiana State University. He expects to receive his master’s degree in Civil Engineering in December 2018, and plans to work for a certain duration within OPT. He still plans to pursue a Doctor of Philosophy (PhD) in close future.