Report No. FHWA-KS-16-17 ▪ FINAL REPORT ▪ January 2017

Evaluation of Lignin as an Antioxidant in Asphalt Binders and Bituminous Mixtures

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Kansas Department of Transportation
Bureau of Research

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Kansas
Department of Transportation

1 Report No. FHWA-KS-16-17	2 Government Accession No.	3 Recipient Catalog No.
4 Title and Subtitle Evaluation of Lignin as an Antioxidant in Asphalt Binders and Bituminous Mixtures		5 Report Date January 2017 6 Performing Organization Code
7 Author(s) Cliff Hobson, P.E.		7 Performing Organization Report No.
9 Performing Organization Name and Address Kansas Department of Transportation Bureau of Research 2300 SW Van Buren Topeka, Kansas 66611-1195		10 Work Unit No. (TRAIS) 11 Contract or Grant No.
12 Sponsoring Agency Name and Address Kansas Department of Transportation Bureau of Research 2300 SW Van Buren Topeka, Kansas 66611-1195		13 Type of Report and Period Covered Final Report 14 Sponsoring Agency Code RE-0395-01 SPR 73-1
15 Supplementary Notes For more information write to address in block 9.		

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The objective of this 2007 limited laboratory research project was to determine the potential of reducing the oxidation rate of HMA pavements by adding lignin to the asphalt binder used to produce the HMA mixture. The testing matrix for this study consisted of two binders, two basic aggregate mixtures, and one hardwood lignin at one concentration. Eight total mixtures with and without lignin were tested after long-term aging to determine if the lignin had any effects on the material properties. The test results from this project showed minimal differences between the lignin test specimens and the control specimens.

Based on the results from this limited study, there is no clear evidence adding lignin to HMA mixtures reduces oxidation or the lignin acted as an antioxidant.

17 Key Words Lignin, Oxidation, Antioxidant, Age-Hardening		18 Distribution Statement No restrictions. This document is available to the public through the National Technical Information Service www.ntis.gov.	
19 Security Classification (of this report) Unclassified	20 Security Classification (of this page) Unclassified	21 No. of pages 45	22 Price

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Final Report

Prepared by

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Kansas Department of Transportation
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A Report on Research Sponsored by

THE KANSAS DEPARTMENT OF TRANSPORTATION
TOPEKA, KANSAS

January 2017

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Abstract

The chemical process of oxidative age-hardening in asphalt pavements is one of the major distresses leading to hot mix asphalt (HMA) pavement failure as evidenced by fatigue and thermal (low temperature) cracking.

Research investigations at the Western Research Institute (WRI) prior to 2005 suggested the addition of lignin may reduce the oxidation rate of asphalt binder. Research efforts by Bishara, Robertson, and Mahoney (2006) at the Kansas Department of Transportation's Research Chemistry Laboratory in 2005 also suggested the addition of lignin to asphalt binder appeared to reduce the oxidation rate of the binder, and therefore, had the potential to be an antioxidant for HMA pavement.

The objective of this 2007 limited laboratory research project was to determine the potential of reducing the oxidation rate of HMA pavements by adding lignin to the asphalt binder used to produce the HMA mixture. The testing matrix for this study consisted of two binders, two basic aggregate mixtures, and one hardwood lignin at one concentration. Eight total mixtures with and without lignin were tested after long-term aging to determine if the lignin had any effects on the material properties. The test results from this project showed minimal differences between the lignin test specimens and the control specimens.

Based on the results from this limited study, there is no clear evidence adding lignin to HMA mixtures reduces oxidation or the lignin acted as an antioxidant.

Acknowledgements

The Kansas Department of Transportation (KDOT) Research Asphalt Team personnel produced the hot mix asphalt samples and specimens for testing and performed the mixture testing part of this project. The KDOT Chemistry Laboratory personnel performed the binder extraction and binder testing.

S.W. Bishara, R.E. Robertson, and Donna Mahoney (Bishara et al., 2006) for their research efforts on the initial phase of this research project which found the addition of lignin to asphalt binder appeared to reduce the oxidation rate of the binder.

Table of Contents

List of Tables

List of Figures

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Chapter 1: Introduction

The Kansas Department of Transportation (KDOT) uses hot mix asphalt (HMA) mixtures when paving approximately 85% of the total roadway miles maintained by the agency. The chemical process of oxidative age-hardening in asphalt pavements is one of the major distresses leading to HMA pavement failure as evidenced by fatigue and thermal (low temperature) cracking. Binder oxidation and hardening has been extensively studied and can be broken down into at least three main forces at work: Oxidation Chemistry, Oxidation Kinetics, and Oxidative Hardening; however, the mechanism of fatigue life decline with oxidation is not yet well understood, and data indicates it is a very important phenomenon and there can be significant differences between different mixture designs (Glover et al., 2009). Slowing the rate of oxidative aging of HMA pavement would increase its service life and create an economic advantage.

Antioxidants have been studied in asphalt pavements, but none have proven to be practical for incorporation into the asphalt industry (Williams & McCready, 2008). Lignin, a known antioxidant, is the second most available biological polymer on earth (Dizhbite, Telysheva, Jurkjane, & Viesturs, 2004). Lignin is found in many sources such as timber, grass, and corn (Glasser & Sarkanen, 1989). Lignin, a complex natural polymer, is also found in biomass residues such as wheat straw and corn stalks. Kansas was the 6th state nationally in its potential for biomass production according to the U.S. Department of Agriculture/Department of Energy (USDA/DOE) in 2004.

Research investigations at the Western Research Institute (WRI) prior to 2005 suggested the addition of lignin may reduce the oxidation rate of asphalt binder. Research efforts by Bishara, Robertson, and Mahoney (2006) at the KDOT Research Chemistry Lab in 2005 also suggested the addition of lignin to asphalt binder appeared to reduce the oxidation rate of the binder and, thus, had the potential to be an antioxidant for HMA pavement.

Chapter 2: Objective

The objective of this 2007 limited laboratory research project was to determine the potential of reducing the oxidation rate of HMA pavements by adding lignin to the asphalt binder used to produce the HMA mixture.

The studies mentioned in the introduction above (WRI and Bishara et al., 2006) were conducted on the asphalt binder only. This research was to study the use of lignin in the HMA mixture, not just the asphalt binder. Intuitively, if lignin is an antioxidant for asphalt binder, then the HMA mixture could also behave in a similar manner. This limited laboratory study was to determine if adding lignin to the mixture slows down the rate of oxidation of the HMA and, therefore, acts as an antioxidant.

This research was conducted using two different asphalt binders, one lignin at one concentration and two different aggregate mixtures.

Chapter 3: Project Description

Bishara et al. (2006) at the KDOT Research Chemistry laboratory used the "Reduced Sampling" method to determine the seven most frequently used unmodified PG 64-22 asphalt binders to pave KDOT agency roads in 2004. Those seven binders were subjected to a battery of chemical tests to rank them according to their propensity for aging. The two extreme binders, the least-likely-to-age and the most-likely-to-age, from the ranking were selected for testing with lignin. Two lignins, a softwood and a hardwood lignin, were mixed with the two extreme binders at concentrations of 0, 2, 4, 7, and 10% of the binder. Both lignins used were from a commercial source since Kansas-produced lignin was not available. Each combination was subjected to rheological testing, according to AASHTO specifications, at the high, intermediate, and low temperatures. The effectiveness of lignin as an antioxidant was determined by an improvement in the aging index (AI) @ 25 °C, provided the critical temperatures were not adversely affected.

Bishara et al. (2006) concluded lignin was an effective antioxidant for asphalt binders and a lignin concentration at the 7 percent level was most effective for the two binders using either of the two lignins. Also, the decrease in AI @ 25 °C was binder dependent rather than lignin dependent; however, the hardwood-lignin seemed to be more effective overall.

This research project built on the Bishara et al. (2006) effort to determine if lignin would also be an effective antioxidant for HMA mixtures. The same two binders and the hardwood lignin used by Bishara et al. were used in this project to provide continuity throughout. The binders used were from the original volume originally obtained by Bishara et al.; however, the original volume of hardwood lignin had been depleted. Therefore, additional hardwood lignin was obtained from the original vendor using the same product number. This lignin was from a commercial source since Kansas-produced lignin was not available.

Two different aggregate types and two existing KDOT HMA mixture designs were originally decided upon for this study. The first aggregate type was crushed gravel and the mix design was to be from a low-volume western Kansas pavement, and the second aggregate type was limestone and the mix design was to be from a higher volume eastern Kansas pavement. The mixture designs would both use an SM-12.5A Superpave gradation with PG 64-22 binders. In addition, the mixtures chosen wouldn’t use anti-strip additives, recycled asphalt pavement (RAP), or recycled asphalt shingles (RAS) to eliminate variables which could potentially influence the study. The specimen matrix used is shown in Table 3.1.

Table 3.1: Specimen Matrix

	Western Kansas Mixtures		**Eastern Kansas Mixtures**	
	Control	**7% Lignin**	**Control**	**7% Lignin**
Binder A	X	X	X	X
Binder H	X	X	X	X

Notes:
1. Binders = A & H
2. Lignin = B108

The eastern Kansas KDOT mix design designation used was 1G06013A. This mixture was developed for District 1 in 2006 for mainline use in Shawnee County. The design was for 4.2 million equivalent single axle loads (ESALs). The western Kansas KDOT mix design designation used was 6G06014A. This mixture was developed for District Six in 2006 for shoulder use in Finney County. The design was for 6.1 million ESALs. The western Kansas mix for this study was originally intended to be from a low-volume road; however, when selecting mix designs and obtaining materials, the aggregates for a low-volume mix design weren’t readily available.

One ESAL is known to cause a quantifiable and standardized amount of damage to the pavement structure equivalent to one pass of a single 18,000-pound, dual-tire axle with all four tires inflated to 110 psi. Design ESALs is a cumulative traffic load summary statistic. The statistic represents a mixed stream of traffic of different axle loads and axle configurations predicted over the design or analysis period and then converted into an equivalent number of 18,000-lb. single axle loads summed over that period.

Table 3.2 lists the aggregates used in the eastern Kansas mix. CS-1 (Crushed Stone) is crushed limestone, CH-1A (Chat) is a waste byproduct obtained during the mining of lead and zinc ores in northeastern Oklahoma which mainly consists of chert, MSD-1 is a manufactured crushed intermediate gradation limestone, and SSG (Sand-Gravel) is a mixture of natural sand and gravel formed by the disintegration of siliceous and/or calcareous materials.

Table 3.2: Eastern Kansas Mix Aggregates

1. Aggregate Producer Information			
Aggr. Desig. / Material Code	% of Mix (Dry)	% of Mix (Wet)	Producer Name / Producer Code
CS-1	40.00	37.800	Mid-States Materials
002010117			00843501
CH-1A	10.00	9.450	Bingham Sand
00201A803			00821811
MSD-1	32.00	30.240	Mid-States Materials
002010917			00843501
SSG	18.00	17.010	Meier's Sand
002010919			00820601
PG64-22		5.500	
	100.00	100.00	

Table 3.3 lists the aggregates used in the western Kansas mix. CG-1 and CG-5 (Crushed Gravel) are produced by crushing siliceous gravel containing not more than 15% non-siliceous material, SSG-2 and SSG-3 (Sand-Gravel) are a mixture of natural sand and gravel formed by the disintegration of siliceous and/or calcareous materials, and MFS-5 (Mineral Filler Supplement) is volcanic ash containing a minimum of 70% glass shards.

Table 3.3: Western Kansas Mix Aggregates

1. Aggregate Producer Information			
Aggr. Desig. / Material Code	% of Mix (Dry)	% of Mix (Wet)	Producer Name / Producer Code
CG-1	15.00	14.213	E.C.A. 3/4"
002010312			00833301
CG-5	10.00	9.475	E.C.A. 3/4 3A
002010512			00833301
SSG-3	10.00	9.475	Klotz Vib Sand
00201C919			00812605
SSG-2	62.00	58.745	Klotz 3/4 RG
00201B919			00812605
MFS-5	3.00	2.843	Marshall Silica
002020505			00836201
PG64-22		5.250	
	100.00	100.00	

The hardwood lignin used for this project was in free-flowing powder form and was brown in color with a vanilla-like odor. The particle-size distribution for the PC-1369 kraft hardwood lignin manufactured by MeadWestvaco was as follows:

Retained on sieve	Percent (%)
No. 50	1.8
No. 120	21.8
No. 200	23.4
No. 325	32.1
Passing No. 325	21.0

The lignin used in this project was treated as a replacement for fine aggregate in the HMA mixture design. However, the quantity of lignin was based on 7% of the binder quantity.

There were eight different mixes developed for the project: two binders, two aggregates, and two lignin options (with and without) for a 2 × 2 × 2 matrix = 8 mixes. The eastern Kansas mixes were labeled 1–4 and the western Kansas mixes were labeled 5–8. Specifically, the mix labels were:

- Mix #1 Eastern Kansas, Binder A, w/o Lignin, 5.5% AC
- Mix #2 Eastern Kansas, Binder A, w/ Lignin, 5.5% AC
- Mix #3 Eastern Kansas, Binder H, w/o Lignin, 5.5% AC
- Mix #4 Eastern Kansas, Binder H, w/ Lignin, 5.5% AC
- Mix #5 Western Kansas, Binder A, w/o Lignin, 5.25% AC
- Mix #6 Western Kansas, Binder A, w/ Lignin, 5.25% AC
- Mix #7 Western Kansas, Binder H, w/o Lignin, 5.25% AC
- Mix #8 Western Kansas, Binder H, w/ Lignin, 5.25% AC

It was critical that all HMA samples/specimens were created and treated equally during mixing and conditioning. The binder is what oxidizes and heat increases the binder oxidation. Therefore, to keep the amount of oxidation the same when producing the samples/specimens, the binder for all HMA samples/specimens was heated the same number of times and for the same length of time. The bulk binder was heated only once after the Research Asphalt Laboratory took possession and the required binder quantity for all specimens (plus a few extras) to be produced was poured into individual containers and let cool. The containers were then placed in plastic bags and sealed.

The specimens produced for the Chemistry Lab testing, the Gmm tests, and the Gmb tests were mixed as a group at one time. The specimens produced for the TSRST and Hamburg tests were mixed as a group at a different time. Those steps eliminated having to hold mixtures at compaction temperatures for long periods of time or reheat material prior to compacting into specimens which could create differing amounts of oxidation.

The individual binder quantities were heated and the lignin was slowly stirred in before mixing the binder with the aggregate. All binder quantities were heated and stirred for the same amount of time. The test specimens were made before the binder cooled to eliminate the need for re-heating. The control specimen binders (no lignin) were heated and stirred the same as those to which the lignin was added.

The loose mix material for the Gmm tests and the Chemistry Lab testing was compacted into plugs at 7–10% air voids with the Superpave gyratory compactor (SGC) for long-term aging. The effort to make all specimens was accomplished so as to store specimens less than 48 hours between making the specimens and long-term aging them.

All specimens were long-term aged per AASHTO R 30 (2002) to simulate 7–10 years of service for in-place compacted HMA pavement. The Gmm and Chemistry Lab loose mix material which was compacted into plug specimens for long-term aging was broken apart back into loose mix material for actual testing during the 16-hour R 30 cooling period to eliminate the need to be reheated. The specimens and loose mix material were then stored in the laboratory at ambient temperature in sealed plastic bags prior to testing and the testing was completed within 14 days.

The specimens were tested per standard HMA testing procedures and all specimens were treated equally, especially when reheating the specimens, to avoid skewing the results.

The KDOT Chemistry Laboratory performed the following binder extraction and tests using the AASHTO Standard Method of Test for:

- T 49 (2007), Penetration of Bituminous Materials
- T 164 (2006), Quantitative Extraction of Asphalt Binder from Hot Mix Asphalt (HMA)
- T 170 (2005), Recovery of Asphalt Binder from Solution by Abson Method
- T 201 (2007), Kinematic Viscosity of Asphalts (Bitumens)
- T 313 (2006), Determining the Flexural Creep Stiffness of Asphalt Binder Using the Bending Beam Rheometer (BBR)
- T 315 (2006), Determining the Rheological Properties of Asphalt Binder Using a Dynamic Shear Rheometer (DSR)

The KDOT Research Asphalt Laboratory performed the following tests:

- AASHTO TP 10 (1996), Standard Method of Test for Thermal Stress Restrained Specimen Tensile Strength (presently withdrawn)

- Kansas Test Method KT-15 (2007), Bulk Specific Gravity and Unit Weight of Compacted Hot Mix Asphalt (HMA)
- Kansas Test Method KT-39 (2007), Theoretical Maximum Specific Gravity of Asphalt Paving Mixtures
- Kansas air permeability test on the Gmb SGC plugs

The Hamburg Wheel Tracking Tests were performed by Kansas State University (KSU) using the Texas Tex-242-F (2000–2004) test procedure.

Chapter 4: Findings

Table 4.1 summarizes the data from the various tests performed on the specimens. All the specimens had been long-term aged per AASHTO R 30 to simulate 7–10 years of in-place service prior to testing, since the objective was to determine if adding lignin to the HMA mixture slowed down the oxidation of the mixture over time.

The addition of hardwood lignin to these HMA mixtures didn’t appear to reduce the oxidative binder aging based on the tests performed during this study. It does appear the lignin co-mingled with the binder and became part of the binder based on the increased binder content in the mixtures with lignin compared to those mixtures without lignin.

Table 4.1: Results Summary Table

Testing Results - Lignin HMA Mixture Study								
Mix #	1	2	3	4	5	6	7	8
	EKS Mix Binder A w/o Lignin	EKS Mix Binder A w/ Lignin	EKS Mix Binder H w/o Lignin	EKS Mix Binder H w/ Lignin	WKS Mix Binder A w/o Lignin	WKS Mix Binder A w/ Lignin	WKS Mix Binder H w/o Lignin	WKS Mix Binder H w/ Lignin
Testing Date:	6/22/07	7/20/07	7/23/07	8/2/07	8/15/07	9/12/07	9/12/07	9/27/07
% Asphalt	5.80	6.12	5.99	6.26	5.51	5.92	5.61	5.87
Penetration @ 77 °F (mm)	22	21	22	20	26	20	24	20
DSR Failure Temp (°C)	80.27	81.39	75.98	78.54	78.11	82.18	75.73	77.21
Viscosity @ 140 °F (poise)	62944	82880	29734	55579	45103	72474	24338	34204
BBR Creep Stiffness (MPa)	101	123	141	210	115	108	129	165
BBR Slope (M)	0.322	0.291	0.346	0.330	0.327	0.321	0.354	0.331
TSRST Failure Temp (°C)	-21	-22	-19	-17	-22	-22	-18	-19
Hamburg Rut Depth (mm)	7.16	4.67	8.1	9.34	16.22	6.63	20	20
Hamburg Wheel Passes	20000	20000	20000	20000	20000	20000	16395	17355
Gmm	2.412	2.407	2.424	2.412	2.422	2.415	2.430	2.424
Gmb	2.333	2.337	2.316	2.315	2.379	2.375	2.373	2.376
Air Voids	3.3	2.9	4.5	4.1	1.8	1.7	2.4	2.0
Air Permeability	1664	1640	1791	1903	1601	1922	1618	1996

Note: Charts generated from the test result data are included in the Appendix.

Chapter 5: Discussion of Findings

The powdered lignin appears to have co-mingled with the virgin binder and became part of the liquid binder based on the increased binder content in the mixtures with lignin compared to those mixtures without lignin. The lignin was a finely ground solid material and was added as an aggregate replacement based on 7% of the binder content. The extracted binder content increased in the four lignin mixtures between 4.5% and 7.4% compared to the control mixtures without lignin. Therefore, lignin may act as an asphalt binder extender in HMA mixtures based on the extracted binder results. The overall test results of the lignin specimens could have been influenced by the increased binder content and, therefore, future studies should investigate having the lignin added in place of binder rather than the aggregate.

The test data from this project showed minimal differences between the lignin test specimens and the control specimens for most of the tests. A statistical analysis (like ANOVA) was not performed on the data from the testing accomplished during this project. Some of the tests performed have precision and bias coefficient of variation ranges such that test results may appear to be slightly different, when in fact, the differences may be due to the precision or bias of the test method.

The HMA mixture test results will be discussed first since this project was to investigate the potential antioxidant properties of lignin in HMA mixtures. Binder testing was also done on binder extracted from the HMA mixture specimens created specifically for binder extraction and testing.

The maximum theoretical specific gravity of loose asphalt mixtures (Gmm) is the ratio of the mass of a given volume of HMA with no air voids to the mass of an equal volume of water, both at the same temperature. All four lignin mixtures had slightly smaller Gmm values than the control mixtures, as can be seen by the data in Table 5.1. The difference between the Gmm values of the lignin mixtures and the Gmm of the mixtures without lignin ranged from 0.005 to 0.012 less (0.5% max) for all four lignin vs. control mixtures. The single operator precision for the Gmm test procedure is indicated as having 0.011 as an acceptable range. There were no apparent differences in the Gmm values, as was expected.

Table 5.1: Maximum Theoretical Specific Gravity (Gmm)

Mix No.	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**
Gmm	2.412	2.407	2.424	2.412	2.422	2.415	2.430	2.424
Comparison	No Difference		No Difference		No Difference		No Difference	

The compacted mixture bulk specific gravity (Gmb) is the ratio of the mass of a given volume of HMA to the mass of an equal volume of water, both at the same temperature. The difference between the Gmb values of the lignin mixtures and the Gmb of the mixtures without lignin ranged from 0.001 to 0.004 less (0.2% max) for all four lignin vs. control mixtures as shown in Table 5.2. The single operator precision for the Gmb test procedure is indicated as having duplicate determinations within 0.031 of each other. There were no apparent differences in the Gmb values, as was expected.

Table 5.2: Bulk Specific Gravity (Gmb)

Mix No.	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**
Gmb	2.333	2.337	2.316	2.315	2.379	2.375	2.373	2.376
Comparison	No Difference		No Difference		No Difference		No Difference	

Air voids (Va) is the total volume of the small pockets of air between the coated aggregate particles throughout a compacted paving mixture, expressed as percent of the bulk volume of the compacted paving mixture. The Va of the compacted lignin mixtures was less than the compacted control mixtures, ranging from 0.1–0.4 less air voids as per Table 5.3. This was most likely caused by the higher asphalt binder contents of the lignin mixtures compared to the control mixtures, as was discussed previously. Small amounts of additional binder added to a mix design will typically reduce the air voids of the mixture. Too much binder can cause stability issues with the mix, especially rutting. The specimens in this study did not appear to have stability issues.

Table 5.3: Air Voids (Va)

Mix No.	1	2	3	4	5	6	7	8
Va (%)	3.3	2.9*	4.5	4.1*	1.8	1.7*	2.4	2.0*
Comparison	Mix 2 Lower*		Mix 4 Lower*		Mix 6 Lower*		Mix 8 Lower*	

* Mixtures 2, 4, 6, and 8 had higher extracted binder content

The Kansas air permeability test is adapted from the SoilTest Asphalt Paving Meter, Model AP-400A. This unit evaluates the rate at which air can be forced, at low pressure, through HMA laboratory compacted specimens or cores. A falling head of water is used to maintain a small, constant pressure drop through the specimen. Measurement of the water flow rate gives the air flow rate through the material. The air-flow rate correlates with the permeability and relative density of the material tested (Soiltest, Inc., 1963).

The air permeability data from this project is shown in Table 5.4. Typically, the values are assigned a permeability "relative value" of very low (0-100), low (101-500), medium (501-1000), or high (1000+). As can be seen from the table, all the air permeability values from testing the Gmb specimens are in the "high" relative value category. These high values of permeability are unusual for specimens with the low air voids these specimens have. Generally, when the air voids of the specimen are less than 4.0%, the air permeability is in the "low" to "very low" relative value category. The specimens for Mixes 1, 2, 5, 6, 7, and 8 all have air voids less than 3.0% and those for Mixes 3 and 4 are 4.5 and 4.1, respectively. The reason for the higher than typical air permeability values obtained on this project was not determined.

Table 5.4: Air Permeability Results

Mix #		1	2	3	4	5	6	7	8
Before Aging	A	1227	1164	1683	1715	n/a	1202	1267	1153
Before Aging	B	1636	--	1470	1683	1265	1622	n/a	n/a
After Aging	A	1231	1640	1917	1822	1133	1762	1557	1817
After Aging	B	2097	--	1665	1983	2069	2082	1678	2175
A & B Avg. Before		1432	1164	1577	1699	1265	1412	1267	1153
A & B Avg. After		1664	1640	1791	1903	1601	1922	1618	1996
Avg. Difference		233	476	215	204	336	510	351	843
Lignin – Control (After Aging)			-24		112		321		379

Notes: All values are $x10^{-9}$ cm^2
Two Plugs for each mix – A set & B set
Lignin – Control = Lignin A & B average minus Control A & B average (shaded row above)

The air permeability of the specimens before aging ranged between 1153 and 1699, and the range of the specimens after aging was 1601 and 1996. The permeability difference before and after aging was more for the western Kansas mixes than the eastern Kansas mixes, except for Mix #2, including both the control and lignin mixtures. Additionally, the mixtures containing lignin were more permeable after aging than the control mixtures (except Mix #2 which was basically the same as the control mix). More permeability after aging could imply binder loss or cracking within the specimen due to oxidation.

The TSRST specimens were tested as per AASHTO TP 10 (1996), Standard Method of Test for Thermal Stress Restrained Specimen Tensile Strength (presently withdrawn). This test determines the tensile strength and temperature at fracture of field cores or laboratory compacted bituminous mixtures by measuring the tensile load in a specimen which is cooled at a constant rate while being restrained from contracting. A compacted bituminous mixture specimen is attached at the ends to the platens of a test system and enclosed in an environmental chamber.

The specimen is cooled at a given rate by introducing liquid nitrogen into the chamber. The initial length of the specimen is held constant by automatic adjustment of the platens. This process continues until tensile fracture of the specimen occurs due to thermal contraction on the long axis. The work necessary to determine the precision of this test was never performed and no justifiable statement can be made on the bias of this test method because there are no reference values available. Some specimen properties (like air voids) can affect some of the output results from this test; however, the temperature at failure is generally not controlled by those specimen properties.

The TSRST specimens tested were 2-inch-diameter by 6-inch-long specimens. Three of these specimens were cored from a single SGC compacted plug. Table 5.5 includes the TSRST data.

Table 5.5: Thermal Stress Restrained Specimen Test (TSRST) Data

Eastern Kansas Aggregate

Binder A				Binder H			
	% Air Voids	Load @ Failure (Kg)	Temp. @ Failure (ºC)		% Air Voids	Load @ Failure (Kg)	Temp. @ Failure (ºC)
Sample	**Mix #1 w/o Lignin**			**Sample**	**Mix #3 w/o Lignin**		
1-A	7.5	393	-20.6	3-A	9.6	373	-21.7
1-B	6.2	393	-18.0	3-B	9.0	305	-17.5
1-C	7.3	414	-23.3	3-C	9.5	303	-16.8
Average	**7.0**	**400**	**-21**	**Average**	**9.4**	**327**	**-19**
Sample	**Mix #2 w/ 7% Lignin**			**Sample**	**Mix #4 w/ 7% Lignin**		
2-A	8.7	340	-21.7	4-A	7.1	352	-16.8
2-B	7.1	391	-21.6	4-B	8.1	343	-17.9
2-C	8.1	409	-22.4	4-C	9.0	345	-17.3
Average	**8.0**	**380**	**-22**	**Average**	**8.1**	**347**	**-17**

Western Kansas Aggregate

Sample	**Mix #5 w/o Lignin**			**Sample**	**Mix #7 w/o Lignin**		
5-A	8.0	497	-23.8	7-A	8.0	362	-17.6
5-B	7.7	385	-18.6	7-B	8.5	370	-18.9
5-C	8.1	472	-22.4	7-C	N/A	N/A	N/A
Average	**7.9**	**451**	**-22**	**Average**	**8.3**	**366**	**-18**
Sample	**Mix #6 w/ 7% Lignin**			**Sample**	**Mix #8 w/ 7% Lignin**		
6-A	7.7	397	-19.3	8-A	8.4	378	-19.5
6-B	8.2	433	-24.9	8-B	7.2	409	-18.8
6-C	7.3	403	-22.8	8-C	7.8	390	-19.0
Average	**7.7**	**411**	**-22**	**Average**	**7.8**	**392**	**-19**

Note: "Load @ Failure" and "Temp. @ Failure" values taken at specimen failure

The temperature at failure of the individual specimens for the eastern Kansas mix with Binder A and no lignin ranged from -18.0 to -23.3 ºC (-21 ºC average) and those with lignin ranged from -21.6 to -22.4 ºC (-22 ºC average). The eastern Kansas mix with Binder H and no lignin ranged from -16.8 to -21.7 ºC (-19 ºC average) and those with lignin ranged from -16.8 to -17.9 ºC (-17 ºC average). The western Kansas mix with Binder A and no lignin ranged from

-18.6 to -23.8 ºC (-22 ºC average) and those with lignin ranged from -19.3 to -24.9 ºC (-22 ºC average). The western Kansas mix with Binder H and no lignin ranged from -17.6 to -18.9 ºC (-18 ºC average) and those with lignin ranged from -18.8 to -19.5 ºC (-19 ºC average). There is typically some variability in the failure temperatures of specimens of the same mixture with this test. The three individual specimen “temperature at failure” values for each mix were averaged to arrive at a single temperature for the mixture. The eastern Kansas mixture with Binder A and lignin and the western Kansas mixture with Binder H and lignin had a 1 °C colder failure temperature than the control mixtures. The western Kansas mixture with Binder A and lignin and the control specimens had the same failure temperature and the eastern Kansas mixture with Binder H and lignin had a 2 °C warmer temperature than the control mixture.

A less oxidized mix should normally have a colder “temperature at failure” when considering the low temperature properties of an aged mixture. There is a possibility individual mixtures with lignin might oxidize less based on the eastern Kansas mixture with Binder A and lignin and the western Kansas mixture with Binder H and lignin. However, it would seem there was no real apparent difference between the lignin and control specimens, in general.

The Hamburg Wheel-Tracking (HWT) Test is used to determine the susceptibility of compacted HMA mixtures to fail prematurely, mainly by rutting and stripping (moisture damage). The test consists of placing specimens in a heated water bath and having a loaded steel wheel track back and forth over the specimen; then observing the deformation vs. the number of wheel passes. The testing was performed at KSU as per Texas standard Tex-242-F for this project. The test runs for 20,000 wheel passes unless the deformation reaches 20 mm, at which time the test is stopped. The heated water temperature was 50 ± 1 °C. Stiffer mixtures tend to have less rutting in this test, thereby indicating a good mixture; however, mixtures that are too stiff typically have a greater chance of being brittle and developing early-on cracking distresses. Therefore, too little rutting in this test could imply the mix is too stiff and may develop early-on cracking distress.

The HWT test data for this project is summarized in Table 5.6. Comparing the lignin specimens to the control specimens shows Lignin Mix #2 had less rut depth than Control Mix #1 (4.67 vs. 7.16 mm), Lignin Mix #4 had more rut depth than Control Mix #3 (9.34 vs. 8.10 mm), Lignin Mix #6 had less rut depth than Control Mix #5 (6.63 vs. 16.22mm), and Lignin Mix #8 had more wheel passes than Control Mix #7 (17,355 vs. 16,395) at 20-mm rut depth. Three of the pairs had basically the same test results (1&2, 3&4, and 7&8) and for the remaining pair (5&6), the results may imply the lignin mixture was stiffer since there was considerably less rutting (lignin = 6.63 mm vs. control = 16.22 mm). Again, it would appear there was no real apparent difference between the specimens.

Table 5.6: Hamburg Wheel Tracking (HWT) Test Data

Mix No.	Number of Passes		Average Number of Passes	Maximum Rut Depth (mm)		Average Rut Depth (mm)
	Left Wheel Path	Right Wheel Path		Left Wheel Path	Right Wheel Path	
1	20,000*	20,000*	20,000*	9.03	5.29	7.16
2	20,000*	20,000*	20,000*	4.37	4.97	4.67
3	20,000*	20,000*	20,000*	5.43	10.78	8.10
4	20,000**	20,000**	20,000**	6.48	12.20	9.34
5	19,600	20,000*	19,800	20.00***	12.26	16.22
6	20,000*	20,000*	20,000*	6.65	6.62	6.63
7	16,590	16,200	16,395	20.00***	20.00***	20.00***
8	18,510	16,200	17,355	20.00***	20.00***	20.00***

Notes: * Reached the specified number of wheel passes without reaching rut depth of 20 mm
** Emergency stop activated at 19,950 passes; extrapolated results
*** Reached the specified rut depth of 20 mm

The extracted binder test results are discussed in the following paragraphs. The binder was extracted from the HMA mixture specimens specifically created for binder extraction and binder testing to determine the binder properties of the binder in the mixtures.

Table 5.7 summarizes the binder penetration test data. The definition of this test, as stated in AASHTO T 49, Penetration of Bituminous Materials, is as follows: consistency of a bituminous material expressed as the distance in tenths of a millimeter that a standard needle vertically penetrates a specimen of the material under known conditions of loading, time, and temperature. Higher values of penetration indicate softer consistency. The temperature, load, and time used for this project were: 25 °C (77 °F), 100g, and 5 sec, respectively.

The penetration was higher for the binder from the control specimens without lignin added than the binder from the lignin specimens, indicating the control specimen binder was softer than the binder from the lignin specimens. The control and lignin binders from the eastern Kansas aggregate mixtures (Mixtures #1–#4) essentially had the same penetration values. The difference in penetration values for the control and lignin binders from the western Kansas aggregate mixtures (Mixtures #5–#8) was more pronounced. A less oxidized binder should tend to be softer or less stiff with a resulting higher penetration value.

Table 5.7: Binder Penetration Test Data

Mix No.	1	2	3	4	5	6	7	8
Penetration (x 0.1 mm)	22	21	22	20	26	20	24	20
Comparison	Mix 1 softer		Mix 3 softer		Mix 5 softer		Mix 7 softer	

The DSR was used to find the upper temperature limit of the binders extracted from the specimens. A summary of the DSR test results is included in Table 5.8. A higher upper temperature would indicate a stiffer binder. However, a slight increase in the upper temperature limit can be a good thing if the other binder properties at the lower temperatures aren't affected. A higher upper temperature limit could help prevent rutting during times when the temperature nears the upper limit.

Table 5.8: DSR Test Data

Mix No.	1	2	3	4	5	6	7	8
Failure Temp (°C)	80.3	81.4	76.0	78.5	78.1	82.2	75.3	77.2
Comparison	Mix 2 stiffer		Mix 4 stiffer		Mix 6 stiffer		Mix 8 stiffer	

The lignin in the mixtures increased the upper temperature limit between 1 and 4 °C (<5.5%) in all the lignin mixtures vs. the control mixtures. Thus, the lignin mixtures are stiffer at this upper temperature limit.

Viscosity is a measure of the resistance to flow of the liquid. The test method used in this study determines the viscosity of asphalt binder by vacuum capillary viscometers. A larger viscosity has more resistance to flow and would indicate a stiffer material. Table 5.9 is a summary of the viscosity data for this project.

Table 5.9: Viscosity at 60 °C (140 °F) Data

Mix No.	1	2	3	4	5	6	7	8
Viscosity (Poise)	62944	82880	29734	55579	45103	72474	24338	34204
Comparison	Mix 2 stiffer		Mix 4 stiffer		Mix 6 stiffer		Mix 8 stiffer	

The binders from all the lignin mixtures (Mixtures #2, 4, 6 and 8) had larger viscosities than the binders from the control mixtures and are therefore stiffer than the control mixtures. The viscosities of the lignin mixtures are significantly larger, ranging from 32% to 87% more.

The BBR test measures the midpoint deflection of a simply supported beam of asphalt binder subjected to a constant load applied to the midpoint of the beam. The test beam is placed in a controlled temperature fluid bath. The flexural creep stiffness determined from the BBR describes the low-temperature stress-strain-time response of the asphalt binder. The low-temperature thermal cracking performance of paving mixtures is related to the creep stiffness and the slope of the creep stiffness versus the logarithm of the time curve of the asphalt binder contained in the mix (AASHTO T 313). As surrounding temperatures drop, pavements contract and build up internal stresses. If this contraction occurs fast enough, the pavement may crack because it does not have time to relax those internal stresses. This type of crack is typically called a thermal or transverse crack. Pavements with high creep moduli and low m-values are more susceptible to low-temperature thermal distress. Table 5.10 lists the BBR stiffness and slope data for the mixtures from this project.

Table 5.10: BBR Test Data (at -12 °C)

Mix No.	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**
Stiffness (MPa)	101	123	141	210	115	108	129	165
Slope (m)	0.322	0.291	0.346	0.330	0.327	0.321	0.354	0.331
Comparison	Mix 2 stiffer		Mix 4 stiffer		Similar stiffness		Mix 8 stiffer	

A larger creep stiffness value indicates higher thermal stresses and a stiffer mixture. Three of the four mixtures with lignin added had larger creep stiffness values than their comparative control mixture. Mixture #6 with lignin had a slightly lower creep stiffness than the Control Mixture #5. The m-value is the slope of the stiffness curve and relates to the material's ability to relax through plastic flow. A larger m-value is associated with high relaxation abilities while a smaller m-value has lower relaxation abilities. The m-value of all lignin mixtures was smaller than the control mixtures. Overall, three of the four lignin mixtures were stiffer and one was similar to the control mixture.

The following summarizes the discussion of the findings above relating the lignin mixtures to the control mixtures:

- Maximum Theoretical Specific Gravity (Gmm)
  - No differences
- Bulk Specific Gravity (Gmb)
  - No differences
- Air Voids (Va)
  - Lignin mixes all less (probably due to higher binder content)
- Air Permeability Results
  - Three of four lignin mixes higher (more permeable)
- Thermal Stress Restrained Specimen Test (TSRST) Data
  - Two lignin mixes colder, one the same and one warmer
- Hamburg Wheel Tracking (HWT) Test Data
  - No real apparent differences

- Binder Penetration Test Data
    - Lignin mixes all have a harder consistency (stiffer)
- DSR Test Data
    - Lignin mixes all stiffer (higher temperature – can be good under the right circumstances)
- Viscosity at 60 °C (140 °F) Data
    - Lignin mixes all had more resistance to flow (stiffer)
- BBR Test Data (at -12 °C)
    - Three of four lignin mixes stiffer and one the same

Based on the test results from this limited study, there is no clear evidence adding lignin to HMA mixtures will reduce oxidation. However, this study used only one lignin at one concentration and two binders with limestone and gravel mixtures; therefore, more research would be required to confirm lignin does not act as an antioxidant in some HMA mixtures.

Chapter 6: Conclusions/Recommendations

The following conclusions/recommendations can be made following this limited study to determine if adding lignin to HMA mixtures would decrease the rate of oxidation of the mixture. These statements only apply to the specific binders, aggregates, and lignin used in this study. There was no clear cut evidence indicating adding lignin to HMA mixtures slowed down oxidation or the lignin acted as an antioxidant.

1. The HMA mixtures with lignin showed mixed results when compared to the control mixtures for degree of oxidation. The differences in the test results were small and most likely within the precision range of the tests; indicating similar levels of oxidation.
2. The extracted binders with lignin were slightly more oxidized than the extracted control mixture binders.
3. The overall results from this study do not support the theory lignin acts as an antioxidant in HMA mixtures.
4. Lignin may act as an asphalt binder extender in HMA mixtures based on the extracted binder and air void results from this study.
5. This research study was limited to laboratory testing and the number of test specimens was small. Additional research is required to confirm lignin does not act as an antioxidant in HMA mixtures and/or draw more specific conclusions. Similar experiments could be run using other types of lignin and/or with other materials typically added to HMA mixtures (RAP, RAS, etc.).

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Appendix

Charts generated from the test result data are included in this Appendix.

See Figures A.1 through A.3 for charts from the HMA mixture data, Figures A.4 through A.8 for charts from the extracted binder data, and Figure A.9 for a chart of the data from the various properties “with” lignin vs. the “without” lignin as a percent.

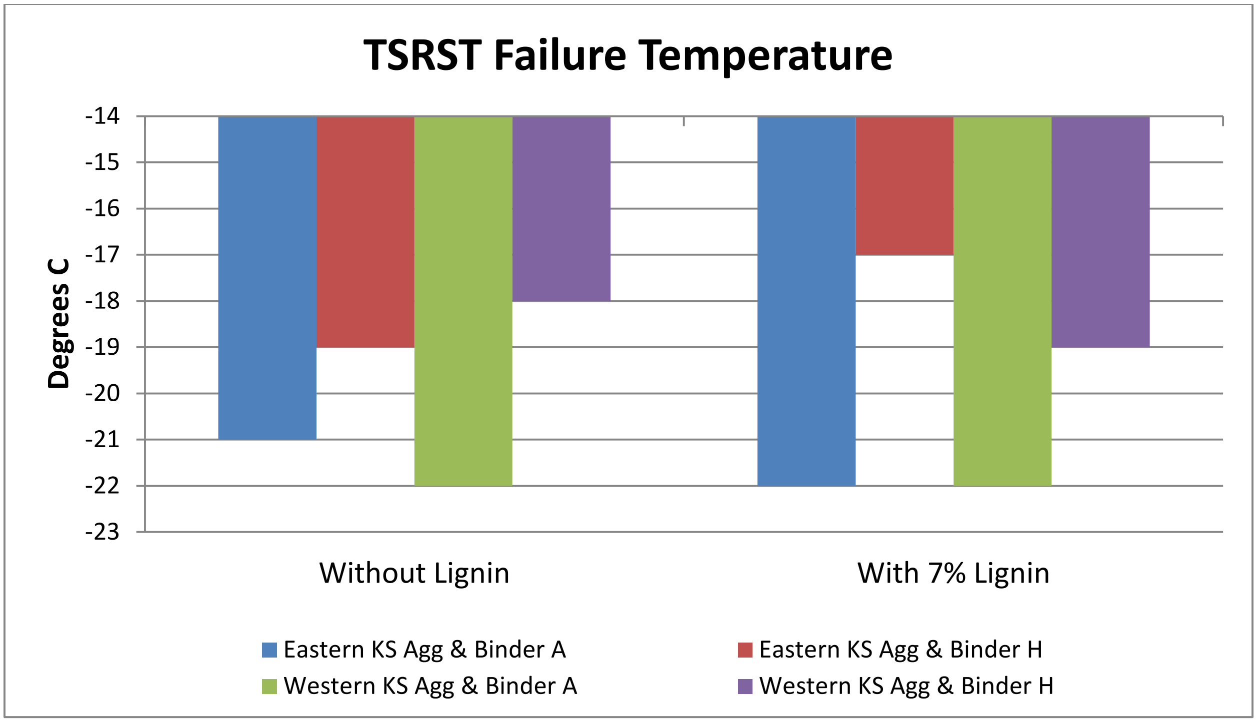

Figure A.1: Mixture TSRST Failure Temperature

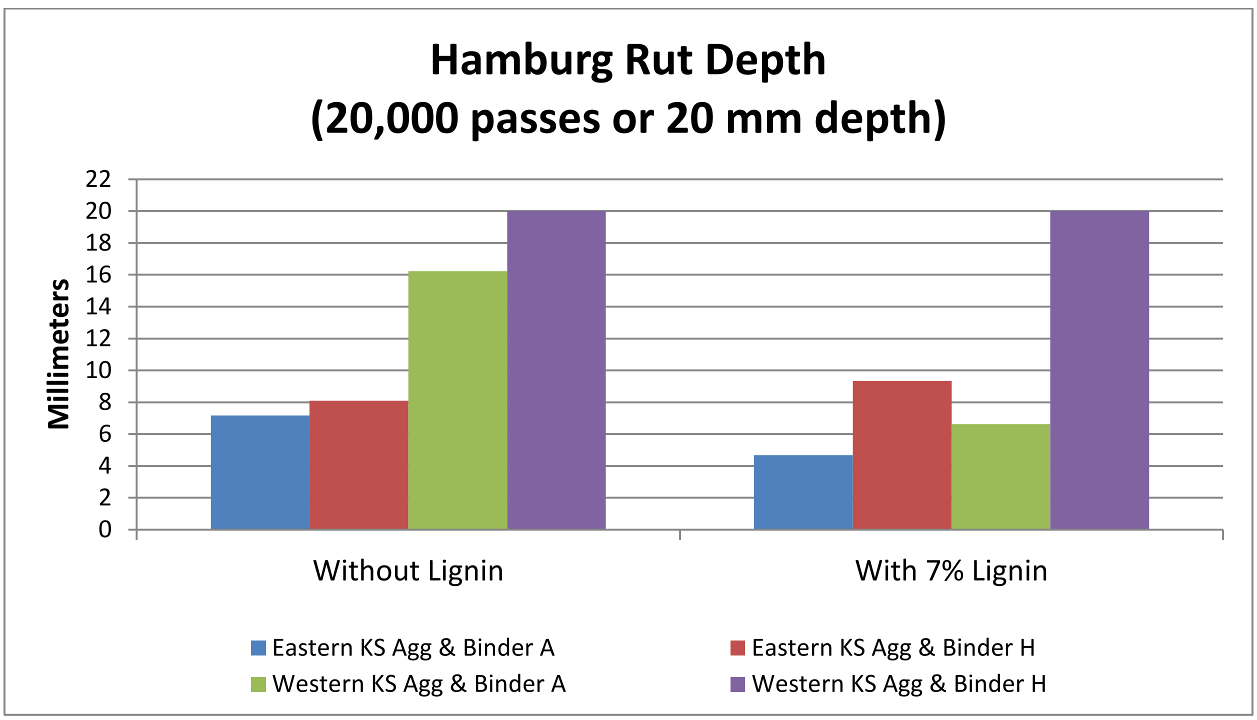

Figure A.2: Mixture Hamburg Rut Depth

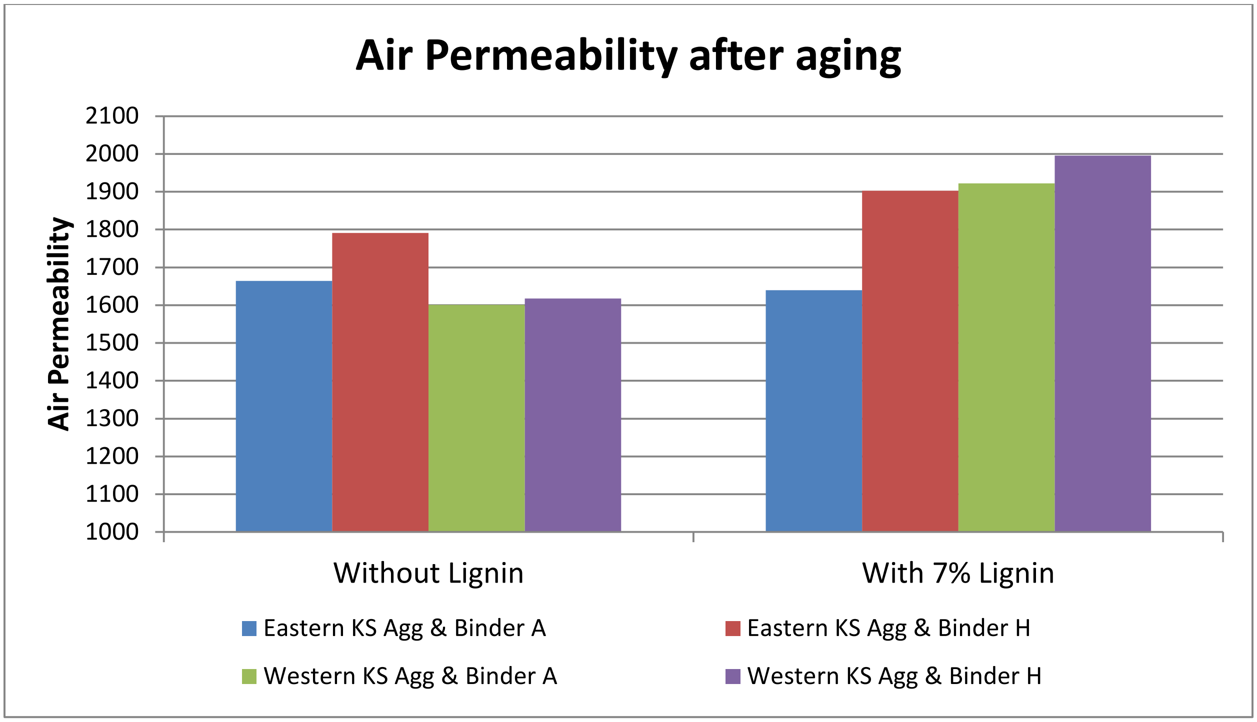

Figure A.3: Mixture Air Permeability after Aging

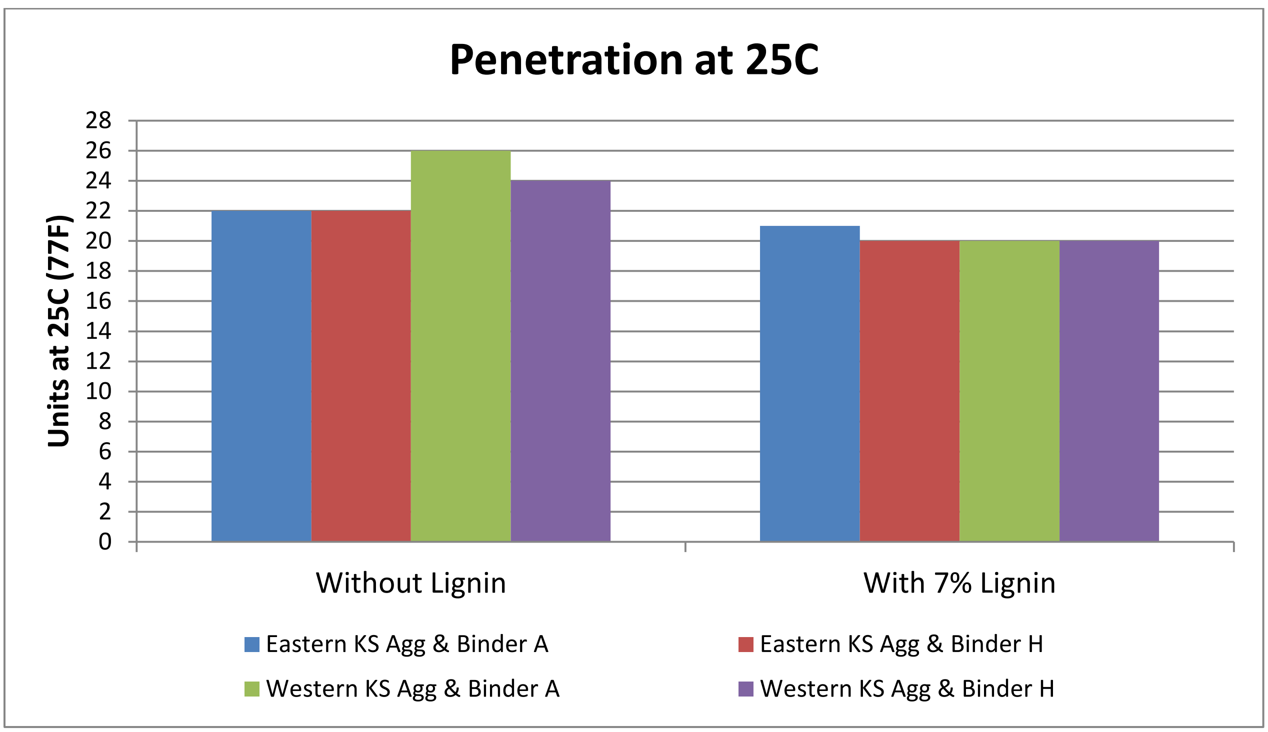

Figure A.4: Extracted Binder Penetration

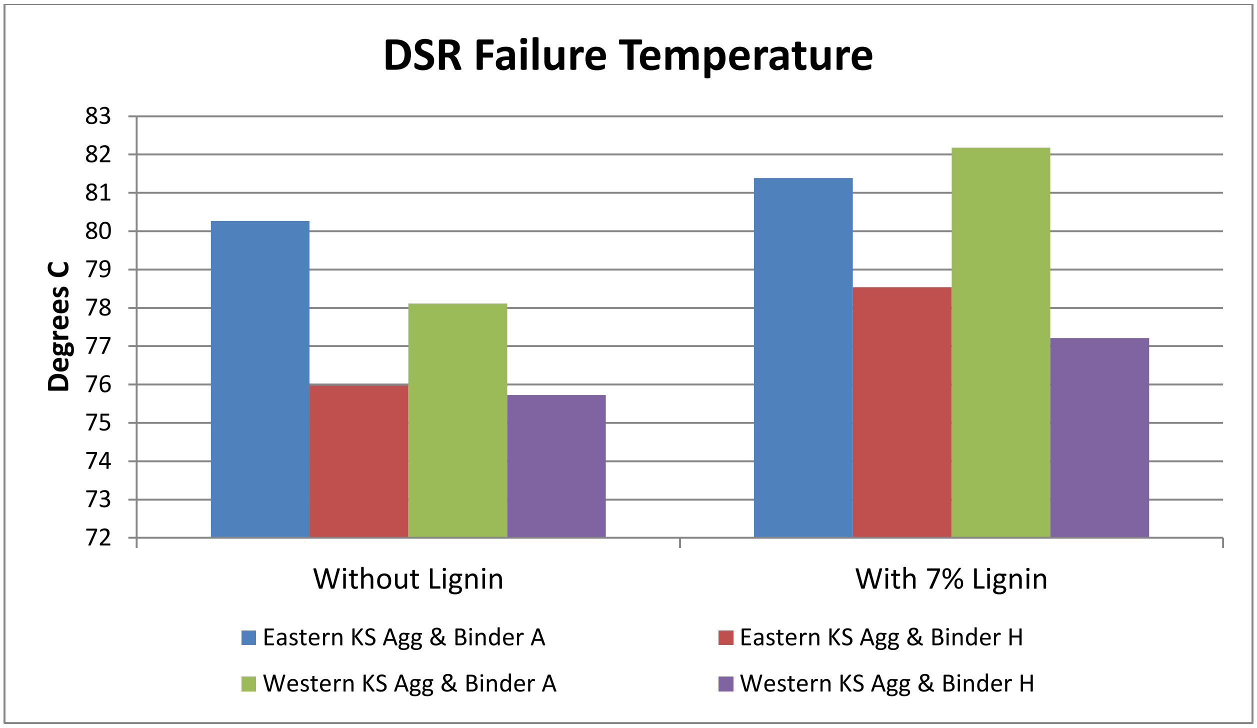

Figure A.5: Extracted Binder DSR Failure Temperature

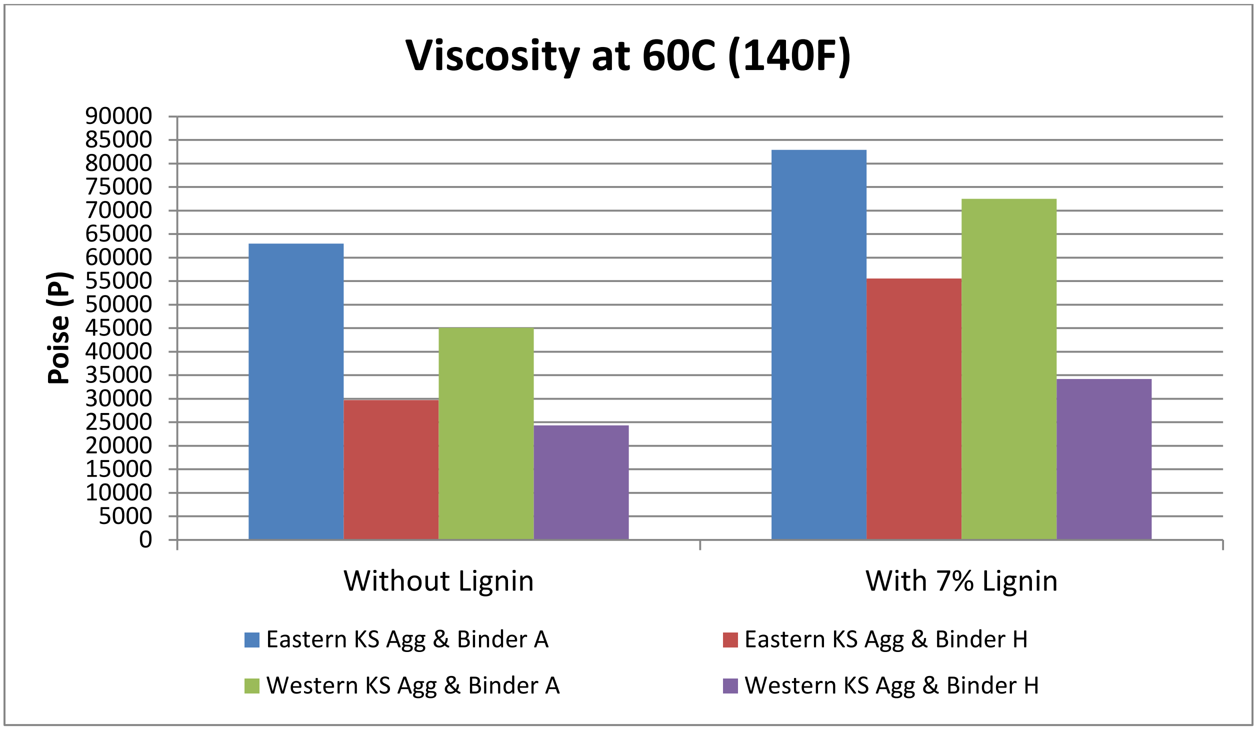

Figure A.6: Extracted Binder Viscosity

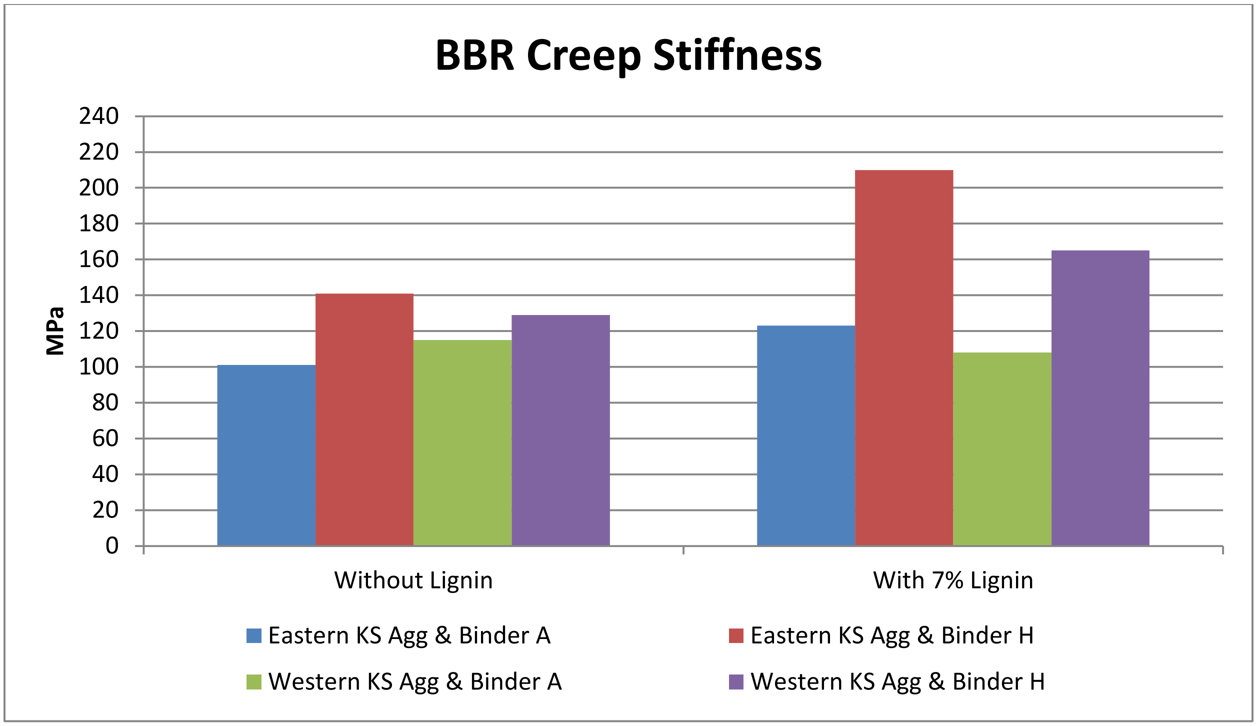

Figure A.7: Extracted Binder BBR Creep Stiffness

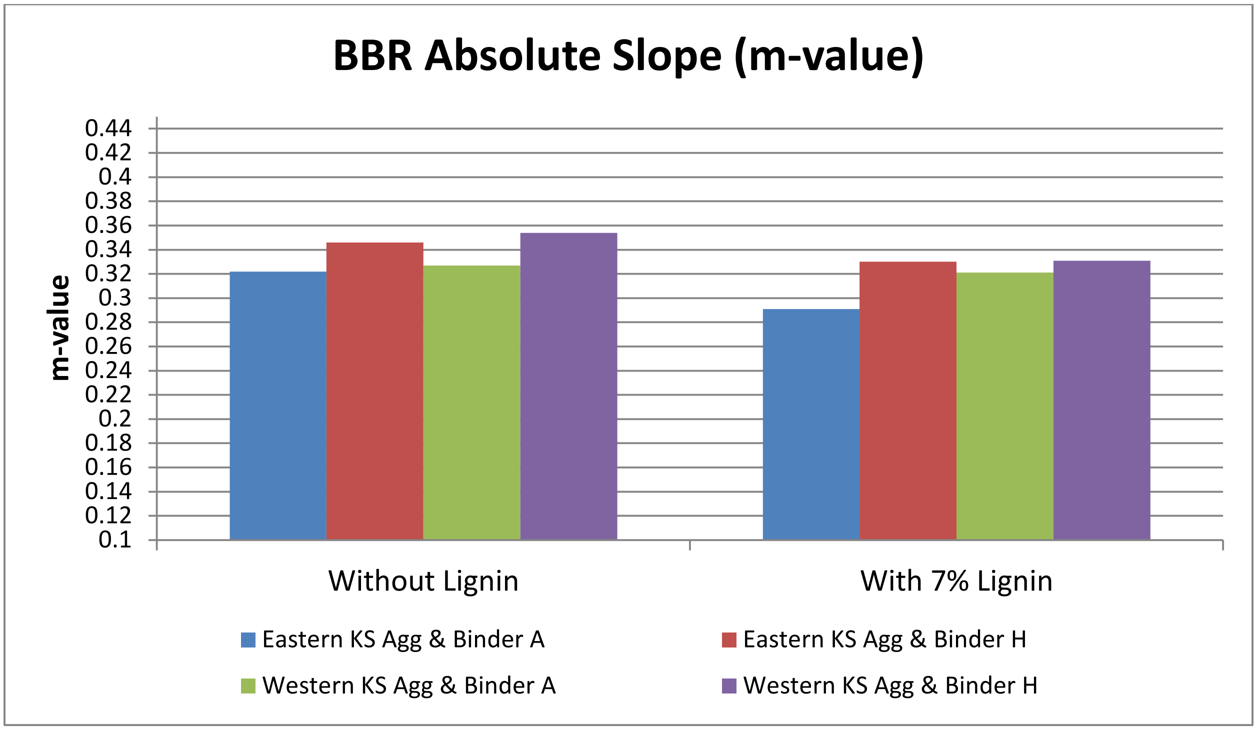

Figure A.8: Extracted Binder BBR Slope (m-value)

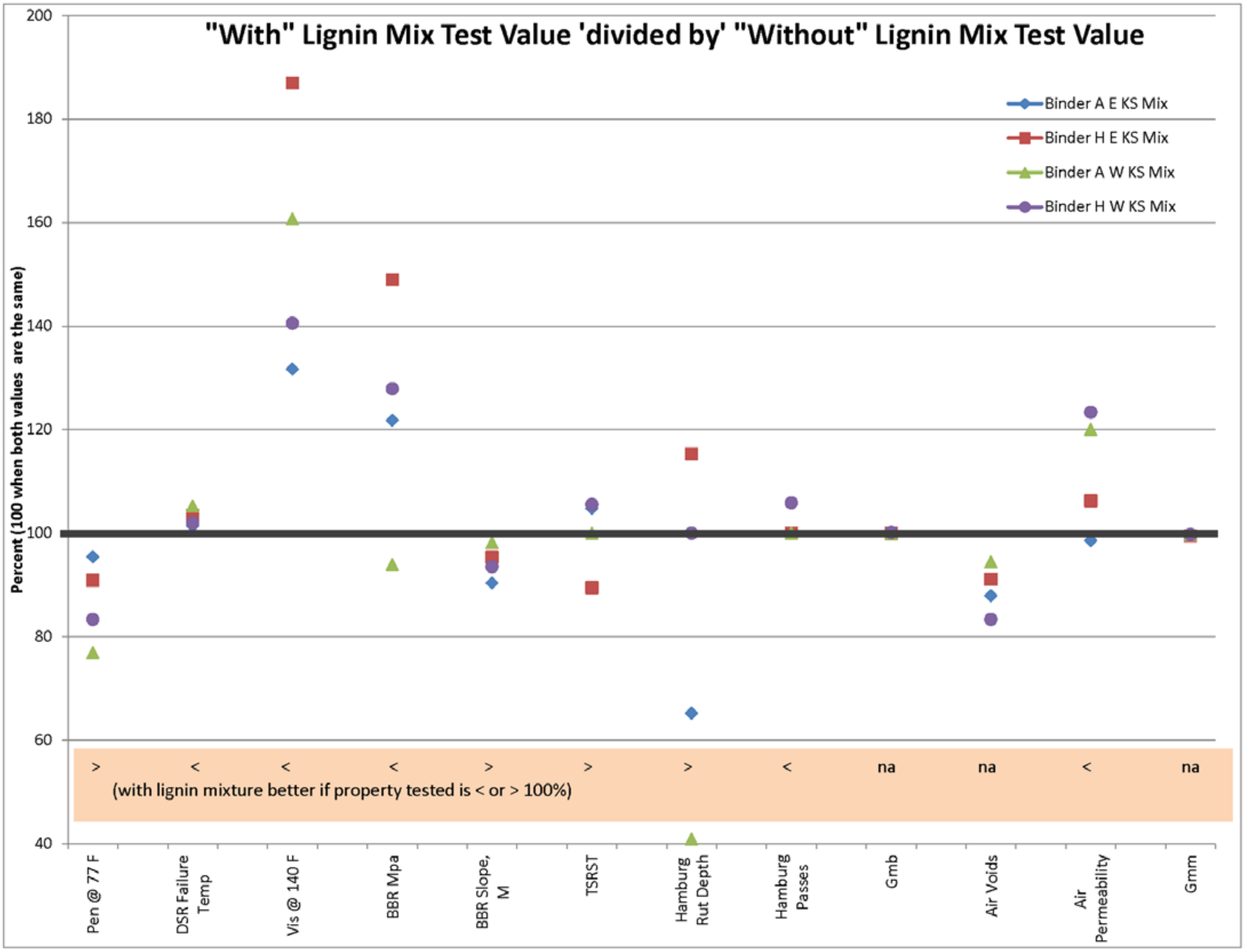

Figure A.9: With vs. Without Lignin Mixture Property Values as a Percent

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