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Evaluation on Physical and Rheological Properties of Lignin as Modifier Asphalt Binder

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Abstract: The research is to evaluate the physical and rheological properties of lignin as a transformer asphalt binder with the workability of lignin as a renewable binding agent in asphalt mixtures. As the demand for petroleum-based binders is increasing from year to year, an alternative to replacing asphalt binders with natural binders has been studied by former researchers. This research investigates the workability of lignin as a renewable binding agent in asphalt binders. The feasibility of lignin was measured through several aspects such as its physical and rheological performance, for comparison between lignin transformer asphalt binder (LMAB) and conventional asphalt binder (CAB), and to determine the optimum percentage of lignin as a transformer in asphalt binder. A review analysis was performed based on previous studies to determine the feasibility of lignin as a renewable binder. Lignin compounds are high molecular weight aromatic polymer compounds that contain phenylpropane as the basic unit. Data was based on previous studies to find out the results based on the results of previous studies through review analysis methods. The laboratory tests involved to analyze physical and rheological performance (LMAB). Among the laboratory tests such as penetration tests and softening tests, which determine the physical performance. Other tests to determine rheological properties are the viscosity test, dynamic shear rheometer test (DSR), pressure aging vessel (PAV), rolling thin film oven (RTFO) which describe various aging conditions of LMAB and the most common test performed in the laboratory is bending beam rheometer (BBR) to find the effect of stiffness. Independent laboratory tests should be conducted in the future to study the use of lignin as a natural binder in asphalt mixtures as this will help future researchers obtain more accurate and specific results.

Keywords: Lignin, Modifier asphalt binder, Physical and rheological properties.

1. Introduction

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The demand for petroleum is increasing to the next decade, allowing the potential to find an alternative asphalt modifier or resource substitute [1]. Because of that, the price of petroleum has grown in recent years as a result of the gradual depletion of resources, resulting in a huge increase in expenses for road construction and maintenance [2]. Recent research on developing alternative asphalt binders for flexible pavement has begun, to reduce the cost, supply, and environmental impacts of using petroleum-based products [3]. Modification of the asphalt binder is one of the best ways to increase pavement workability [4]. Integrating new goods and technologies to combine greener resources, trash, and recycled materials into the asphalt mixture formula could help to boost the asphalt paving industry's resilience and cost-efficiency [5].

As the demand for petroleum-based binders is increasing from year to year, an alternative to replacing asphalt binders with natural binders has been studied by former researchers. The research aims to investigate the workability of lignin as a renewable binding agent in asphalt binders. The feasibility of lignin was measured through several aspects such as its physical and rheological performance, for comparison between lignin modifier asphalt binder (LMAB) and conventional asphalt binder (CAB), and to determine the optimum percentage of lignin as a transformer in asphalt binder. The analysis review was performed based on previous studies to determine the feasibility of lignin as a renewable binder. The previous study data to be taken care of on the laboratory tests involved to analyze physical and rheological performance (LMAB). Among the laboratory tests such as penetration tests and softening tests, which determine the physical performance. Other tests to determine rheological properties are the viscosity test, pressure aging vessel (PAV) and rolling thin film oven (RTFO) which describe various aging conditions of LMAB and the most common test performed in the laboratory is bending beam rheometer (BBR) to find the effect of stiffness.

2. Assessment of Concrete Structure

Since the need for petroleum-based asphalt binders is rising every year, the alternative to find another suitable binder getting intense [6]; [7]. Along with the rapid growth of industrial transportation and population, several types of natural binder have been identified to meet the requirement of asphalt binder to fully or partially substitute the asphalt binder.

Materials and Methods

2.1 Materials

Lignin was one of the promising materials to be used in the asphalt binder. Hence, the reviews were based on its origin and locality, as well as the advantages it would bring to hot mix asphalt (HMA) as an asphalt binder extender. Lignin can be found in most of the plant-based biomass and it acts as an adhesive to the plant's cell so that the stem stays rigid and still [1]. It is the most natural material that existed in the world and can be primarily found in pulp and paper industries by-products [14]. Lignin contains more than 25% of renewable organic carbon, and become one of the most wastes polymers, mostly from industrial activities [9]; [3]. Lignin can be a great promising alternative modifier in asphalt mixtures because of its similarity of chemical properties with petroleum-based asphalt. It has the potential to substitute the required amount of bitumen fully or partially in hot asphalt mixture [1]. This is because, through several laboratory tests, lignin as modifier asphalt binder (LMAB) shows great performances in terms of viscosity, aging resistance, and stiffness effect. Moreover, lignin can be used as an enhancing antioxidants additives to the pavement mixture with its high antioxidative properties [10]. Many kinds of lignin can be traced from waste, industrial processes, and plants. Lignin can be obtained from lignocellulosic materials such as corn cobs, leaves, wheat straw, and many more [11]. The content of lignin in these materials can be varied and dependent on the cellulose and hemicellulose contents.

2.2 Methods

From the analysis in the literature review, the research began by monitoring and looking for previous data to do the extraction and analysis process. Since one of the goals is to compare the performance between modified and conventional binders, the journals were studied thoroughly on the data obtained and analyzed the discussion and results to make the comparison. The comparison reported was based on the selection of tests performed and the materials used by the previous researchers in the experiment to get the data. Data extraction was conducted by organizing a table of previous studies. All the requirements in the table were filled with the findings found in journals. Hence, by the laboratory analysis data obtained from the former researchers, this study will be on the effectiveness of adding lignin in asphalt mix by their performances. The workability of lignin as a modifier asphalt binder will be measured by its physical and rheological properties. Among of lab tests are penetration tests, and softening tests, which decide on the physical performances. The other tests to determine the rheological properties would be viscosity test, dynamic shear rheometer test (DSR), pressure aging vessel (PAV) and rolling thin film oven (RTFO) which portray the various aging conditions of (LMAB) and the most common test conducted in the laboratory is bending beam rheometer (BBR) to find the stiffness effect.

3. Results and Discussion

The research is mainly on reviewing and monitoring previous researchers 'data, the research objectives were accomplished. The discussion on the laboratory result test by the former researchers in identifying the variety of physical and rheological performance of lignin modifier asphalt binder (LMAB). The performances were varied since there were several types and contents of lignin used in the experiments. Even though the type and proportion of lignin used were different, the outcomes of the result in terms of performances are still good due to the chemical properties and characteristics of the lignin itself.

3.1 The penetration test result of CAB and LMAB

Penetration value was measured by the experiment of penetration test accordingto ASTM D-5 standard. The grades were determined after the penetration tests which, for example, asphalt binder grades of B 50/70, B 100/150, and B 160/220. The meaning of 50/70 is the range of penetration value which to accurate 50 to 70. The result of the penetration test gave values that help to classify and grade the asphalt binder in the consistency's form. Table 1 and Figure 1 show the results of the penetration test conducted by Batista (2018) and Wu (2020).

Table 1: The penetration value of CAB and LMAB with different lignin content

Physical Properties	Penetration Value (0.1mm)	Lignin Content (wt.%)	Penetration Value (0.1mm)
Type of Asphalt Binder	CAB		LMAB
(Wu et al., 2020)	65.7	5.0	43.5
(Batista et al., 2018)	41.1	1.0	40.0
		4.0	39.7
		6.0	36.8

Table 1, incorporating lignin to the mixtures showed a significant decrease in the penetration value of LMAB compare to CAB. The test conducted by Wu et al., 2020 using asphalt binder grade 70 had the penetration grade of 65.7 (0.1mm) under the standard condition of (25°C/0.1 mm), and dropped to 43.5 (0.1mm) with the addition of 5 wt.% lignin concentration. The tabulated data were expressed in Figure 1 as a bar graph. Based on the data illustrated in Figure 1, the lignin content added to the mixtures gave lower penetration values at each increment of percentage. Obtained the result by different amounts of lignin added in the mixtures. At 6 wt.% of lignin, the penetration grade drop to 36.8 (0.1mm), while 39.7 and 40 (0.1mm) at 1 wt.% and 4 wt.% of lignin, respectively [12].

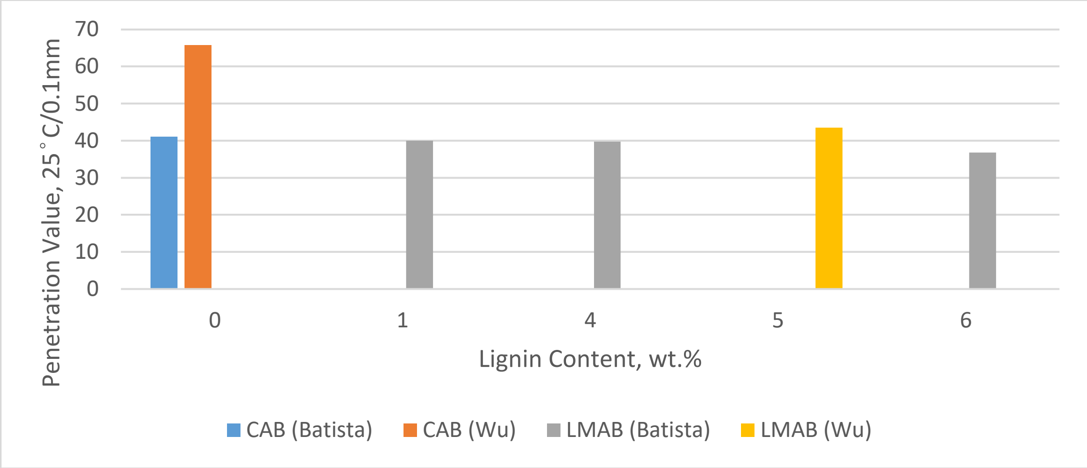

Figure 1: Penetration value of CAB and LMAB with different lignin content.

Based on figure 1, According to [13], the test has resulted in a stiffer asphalt binder, these lower penetration values, mainly at 6 wt.% showed higher stiffness of the mixture, hence making the binder harder and thicker. The sink depths of the samples asphalt binder decreases as the ductility also becomes poorer with the lignin concentration keep on adding. However, Wu (2020) found that equal volume of lignin and conventional binder in the HMA give higher penetration values with modification by lignin. Hence, the results showed the relationship of as the lignin content increases, the penetration value decreases.

3.2 The softening test result of CAB and LMAB

The softening point test was conducted to identify the softening point of the asphalt binder. The most common softening point test is the Ring and Ball test according to ASTM D-36 standard. Table 2 and Figure 2 showed the results of a softening point from the tests conducted by three former researchers.

Table 2: The softening point of CAB and LMAB with different lignin content.

Physical Properties	Softening Point (˚C)	Lignin Content (wt.%)	Softening Point (˚C)
Type of Asphalt Binder	CAB		LMAB
(Wu et al., 2020)	46.7	4.0	50.5
(Norgbey et al., 2020)	39.8	5.0 10.0	41.0 42.0
(Batista et al., 2018)	50.5	1.0 4.0 6.0	50.5 49.0 48.5

Test supervised by Norgbey (2020) showed the result of softening point a slight decrease as the lignin content increases. The conventional binder grade used had a 39.8°C softening points, then with an additional lignin content of 5 wt.% and 10 wt.%, dropped to 41.0°C and 42.0°C, respectively. The same results conducted by Batista (2018) portrayed a little decline on a softening point of LMAB, compared to CAB. The asphalt binder grade used was 50/70. Besides, in a test conducted by Wu (2020) in comparing the softening point between LMAB and a petroleum-based binder, the results show an increase in softening value for LMAB from 46.7°C to 50.5°C with 4 wt.% lignin. It was found that equal replacement of lignin and virgin binder increases the bitumen softening point and complexity of modulus. At higher temperatures, LMAB also showed great performance [14]; [15].

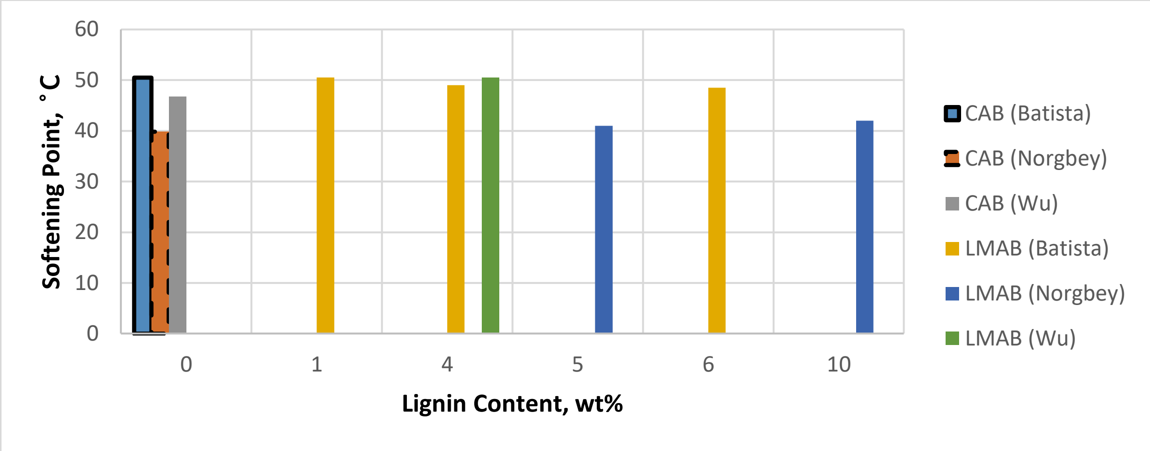

Figure 2: Softening point of CAB and LMAB with different lignin content

From analysis, the 5 wt.% to 6 wt.% of lignin concentration showed the lowest softening point. The values show also an inverse relationship to the viscosity of LMAB as if the softening point increases, the viscosity of LMAB will be decreased [13].

3.3 The viscosity test result of CAB and LMAB

There are four properties of rheological performances that have been analyzed in this research which are viscosity, aging, rutting resistance, and stiffening effect. Viscosity result of LMAB from the RV test is important to know the fluidity of the LMAB. The ability of a binder is measured by its viscosity, as it plays a significant role in the mixing and compaction process during road paving. The result obtained by the readings from rotational viscometer, following the standard of AASHTO T 316, ASTM D 4402, and ASTM D6373 according to the Superpave Standard. The data from the laboratory test conducted by Batista (2018) and Xu (2017) were shown in Table 3 and Figure 3.

Table 3: Viscosity of CAB and LMAB with different lignin content and temperature

Rheological Properties	Temperature (˚C)	Viscosity (Pa.s)	Lignin Content (wt.%)	Viscosity (Pa.s)
Type of Asphalt Binder		CAB		LMAB
(Batista et al., 2018)	135	0.34	1.0 4.0 6.0	0.36 0.38 0.43
(Batista et al., 2018)	155	0.17	1.0 4.0 6.0	0.18 0.20 0.19
(Batista et al., 2018)	177	0.05	1.0 4.0 6.0	0.06 0.07 0.08
(Xu et al., 2017)	135	2.2	5.0 10.0	2.40 2.80
(Xu et al., 2017)	155	1.5	5.0 10.0	1.60 2.0

The viscosity of an asphalt binder is identified by the rotational viscosity that indicates the workability of fluid to flow resistance. The test was organized at three different temperatures which were at 135°C, 155°C, and 177°C, that each with three distinct lignin content. The viscosity values of CAB between Batista and Xu were different at temperatures measured because both of them used different types of binder grades. However, the results still show that even though different types of original binder and lignin content were used, the performance and viscosity of LMAB keep increasing. The summary of the result showed the viscosity of LMAB was higher than CAB for all temperatures and the highest with 5.0 wt.% lignin at 135°C. There were not many changes in viscosity value at 177°C compared to the other two temperatures supervised. Hence, most of the time in real fieldwork, asphalt binder at 135°C is often used as it gives the best viscosity value, according to the ASTM standard. Moreover, the data showed the viscosity values decreases as the temperature increases. However, a binder with 1.0 wt.% lignin gave better performance than other concentration values, according to penetration and softening point test as the data obtained was the lowest viscosity value (Wang et al., 2019).

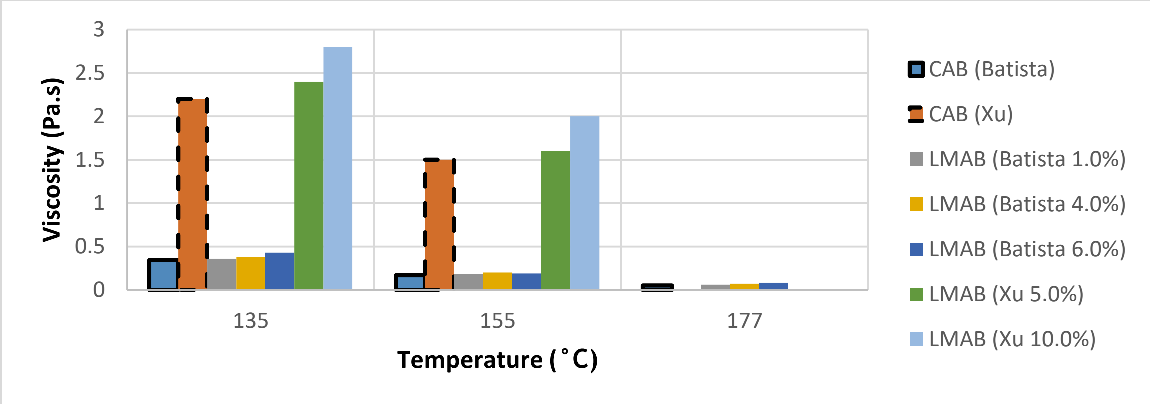

Figure 3: Viscosity of CAB and LMAB with different lignin content and temperature

The result indicates that the higher viscosity of a binder would need a higher temperature for the mixing process with aggregates. This is because the high consistency of fluid brings effects to the physical and rheological properties of the asphalt binder, making it more brittle and harder while its ductility and elasticity get reduced. According to [16], the values increase because of the lignin fibers that react to the light component of the asphalt binder. The existence of the lignin molecules in the asphalt binder obstructs the flowability of the mixtures, causing the viscosity to higher and temperature susceptibility to lowered. The viscosity of a binder has an inverse relationship with penetration value, as if the test showed a decrease in the value of penetration, the viscosity of the fluid must be increased. Moreover, it has a proportional relationship with the lignin content that is as the lignin content increases, the viscosity value of the binder increases.

3.4 The pressure aging vessel test result of CAB and LMAB

The aging of asphalt binder or asphalt can be determined by running specific tests in the laboratory. Usually, to show short-term aging, the RTFO test is conducted while for long-term aging, the PAV test is more suitable and accurate. The aging resistance will be measured by observing the carbonyl index and fourier transform infrared spectroscopy. Usually, the result will get to obtain after the sample had undergone weathering process during specific hours. Hence, the result of aging can be measured. Table 4 and Figure 4 showed the data of aging resistance from the experiment conducted by Batista (2018), and Table 4 and Figure 4 showed the results from Xu (2017), for comparison.

Table 4: Carbonyl Index of CAB and LMAB by Batista (2018)

Rheological Properties	Duration of Weathering (Hour)	Carbonyl Index	Lignin Content (wt.%)	Carbonyl Index
Type of Asphalt Binder		CAB		LMAB
(Batista et al., 2018)		0.18	1.0	0.04

	RTFO		4.0	0.04
			6.0	0.04
(Batista et al., 2018)	10	0.26	1.0	0.11
			4.0	0.10
			6.0	0.13
(Batista et al., 2018)	20	0.34	1.0	0.10
			4.0	0.15
			6.0	0.12
(Batista et al., 2018)	30	0.32	1.0	0.13
			4.0	0.14
			6.0	0.10
(Batista et al., 2018)	40	0.16	1.0	0.13
			4.0	0.14
			6.0	0.11
(Batista et al., 2018)	50	0.27	1.0	0.12
			4.0	0.12
			6.0	0.12

The test was monitored for 50 hours by using the aged bitumen sample from the RTFO test. From Table 4, the carbonyl index of CAB was increased as the duration of weathering exposure increased. However, with the addition of lignin to the mixtures, the carbonyl index started to fall. The carbonyl index of CAB at 10 hours of weathering showed 0.26 and with 1 wt.%, 4 wt.%, and 6 wt.% of lignin added, the data showed 0.11, 0.10, and 0.11, respectively. The carbonyl content of LMAB became constant during 50 hours of weathering as the values obtained with different lignin content were constant. Which was 0.12. LMAB results showed a lower carbonyl index except for the 4 wt.% lignin content after 50 hours of weathering test. Thus, from the data, a concentration of 6 wt.% lignin to the binder, mainly at 30 hours gave the lowest carbonyl index which was 0.10, therefore making it the higher aging resistance.

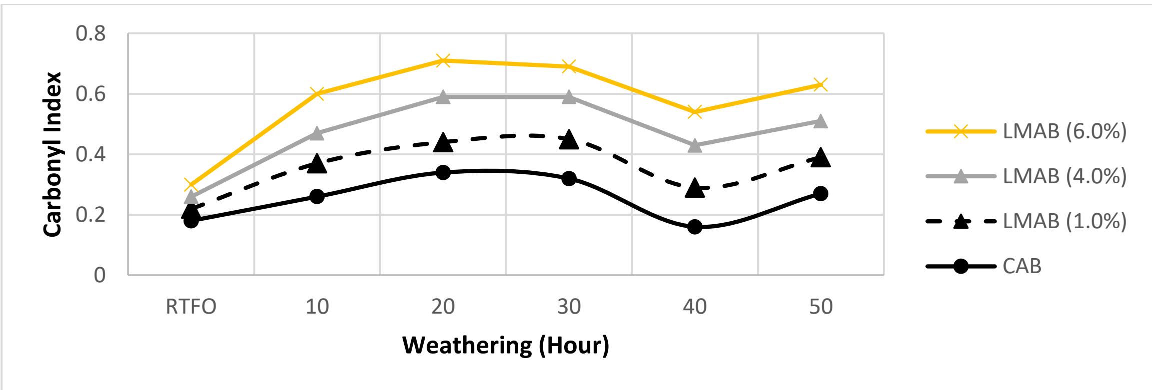

Figure 4: Carbonyl Index of CAB and LMAB by Batista (2018)

Different from Xu's experiment in Table 5 and Figure 5, the three different binder types were used to measure the carbonyl index. Two of the binder were samples of aged bitumen, from RTFO and PAV tests. The content of lignin added into the mixtures wwas5 wt.% and 10 wt.% signifies increment of carbonyl index of LMAB from the original value. The carbonyl index of CAB incorporated with lignin showed a steady increase compared to RTFO and PAV samples that showed a sharp increase from 5 wt.% to 10 wt.% of lignin. This indicates even though different types of the binder were used with different lignin content been added, the carbonyl index in the asphalt mixture incorporated with lignin increases. In measuring the resistance to aging of an asphalt binder, the carbonyl index value is important as the higher the value, the higher the aging resistance.

Table 5: Carbonyl Index of CAB and LMAB by Xu (2017)

Rheological Properties	Types of Binder	Carbonyl Index	Lignin Content (wt.%)	Carbonyl Index
Type of Asphalt Binder		CAB		LMAB
(Xu et al., 2017)	Base Binder	0.009	5.0 10.0	0.020 0.022
(Xu et al., 2017)	RTFO	0.013	5.0 10.0	0.018 0.100
(Xu et al., 2017)	PAV	0.015	5.0 10.0	0.020 0.018

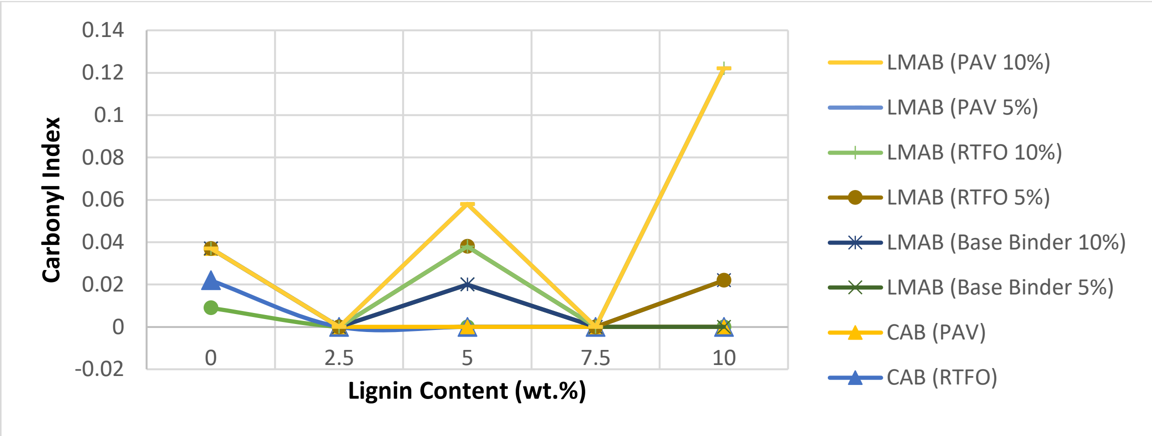

Figure 5: Carbonyl Index of CAB and LMAB by Xu (2017)

In addition, the aging of the LMAB getting higher than the conventional binder [17]. The carbonyl index group (C=O) which formed during the normal oxidation reaction of asphalt binder when exposed to certain temperatures is getting lower due to the incorporation of lignin in the mixtures. Research shows that lignin has antioxidant properties which help to neutralize the triggered radicals during the oxidation period of a binder. With the readings and reviews from journals, this study is assured that LMAB is workable as it is strong in aging resistance, thus able to slow down the aging process.

3.5 The dynamic shear rheometer test result of CAB and LMAB

The rutting resistance is usually measured by conducting a dynamic shear rheometer test. Table 6 and Figure 6 expressed the data from Xu (2017) and Batista (2018). According to Xu (2017), the rutting resistance of LMAB gradually increases as more lignin is added to the asphalt mixtures. The binder type used were PG 64-22 and PG 76-22 and even though they were different in grade, the result of rutting resistance between the two binders still showed the same increment. For example, at a temperature of 70°C for PG 64-22, the rutting resistance for CAB is 0.7 (G/sin ϕ) kPa, and with incorporating 10 wt.% lignin, the data showed 1.0 (G/sin ϕ) kPa.

Table 6: Rutting resistance of CAB and LMAB

Rheological Properties	Types of Binder	Temperature (˚C)	Rutting Resistance (G/sin ϕ) kPa	Lignin Content (wt.%)	Rutting Resistance (G/sin ϕ) kPa
Type of Asphalt Binder			CAB		LMAB
(Batista et al., 2018)		64	1.5	1.0 4.0	1.5 1.8

				6.0	1.9
	PEN 50/70		0.7	1.0	0.7
		70		4.0	0.8
				6.0	0.9
				5.0	1.6
		64	1.4	10.0	2.1
(Xu et al., 2017)	PG 64-22			5.0	0.8
		70	0.7	10.0	1.0
				5.0	1.1
		82	1.0	10.0	1.4
(Xu et al., 2017)	PG 76-22			5.0	0.65
		88	0.6	10.0	0.8

Batista that used a different binder grade, PEN 50/70 conducted the test at two different temperatures which at 64°C and 70°C also had the rutting resistance of LMAB increases. This assures that despite the difference in asphalt binder grade, adding lignin into the mixture of asphalt improved the rutting resistance hence showing its workability.

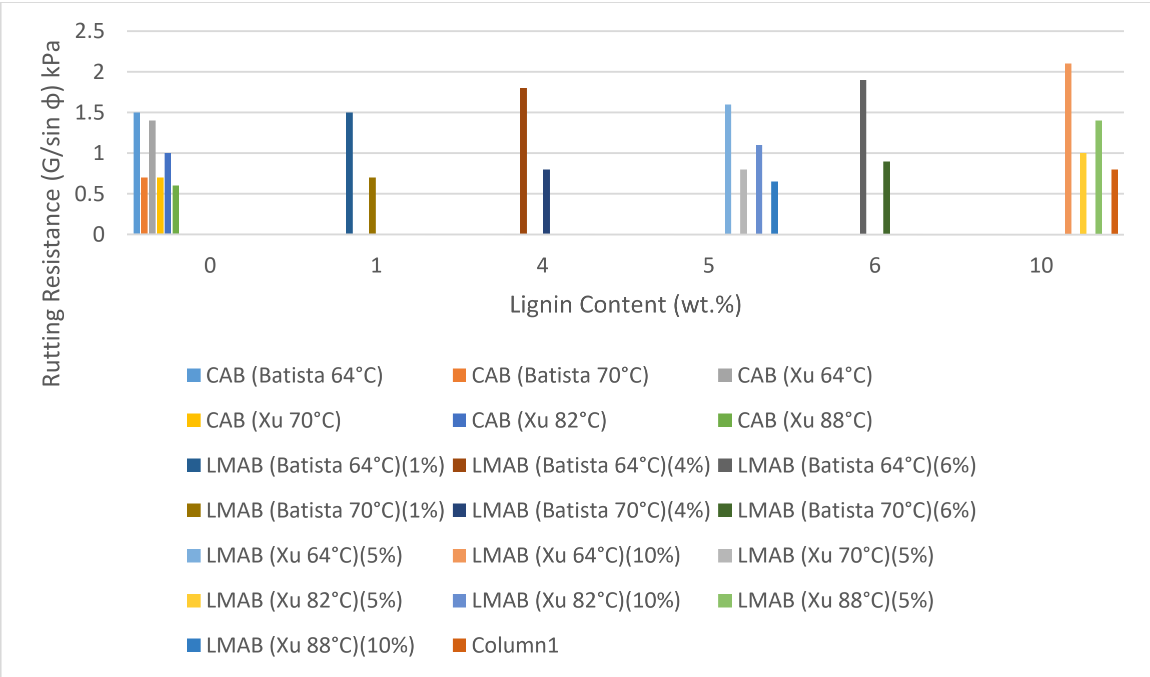

Figure 6: Rutting resistance of CAB and LMAB

Based on Figure 6 the LMAB showed a good rutting resistance at a higher temperature which is also stronger in resisting aging compared to CAB [18]; [19]. The relationship is that along with the temperature rising, as the lignin concentration in the binder increases, the rutting resistance also increases. This is also to specify that asphalt binder with a higher (G/sin ϕ) kPa value portrayed a higher rutting resistance. The rutting factor (G/sin ϕ) is the indicator of the binder's resistance towards rutting, as it means higher (G/sin ϕ) is the higher reluctance to rutting.

3.6 The bending beam rheometer test result of CAB and LMAB

The most common test conducted in the laboratory is the bending beam rheometer (BBR). The test measures the complex modulus and phase angle value of the asphalt binder at low temperatures. Table 7, Figure 7, and Figure 8 demonstrated the data from Wang (2019) and Batista (2018) laboratory tests to find the stiffness effect of Pen60/70 of raw asphalt binder as CAB and flocculent lignocellulose and granulated lignocellulose as LMAB.

Table 7: The stiffness effect of CAB and LMAB

Rheological Properties	Temperature (˚C)	Creep Stiffness (MPa)	Creep Stiffness (MPa)	m-value ($x10^{-2}$)	m-value ($x10^{-2}$)
Type of Asphalt Binder		CAB	LMAB (wt.%)	CAB	LMAB (wt.%)
(Wang et al., 2019)	-6.0	175.0	80.0	32.7	38.9
	-12.0	280.0	140.0	28.0	31.9
	-18.0	541.0	295.0	19.0	27.9
(Batista et al., 2018)	-6.0	70.0	55	44.0	45.0
	-12.0	125.0	125	35.0	36.0
	-18.0	325.0	260	25.0	28.0

Based on Table 7, Wang's stiffness of CAB was reduced from 175 MPa to 80 MPa after the addition of lignin at -6°C, whilst the m-value LMAB increases to 38.9 x10-2 from 32.7 x10-2 of CAB. The same pattern of results went to Batista's test, which also experienced decrement in creep stiffness value whilst increment in m-value. The binder at -12°C and -18°C also showed the same trend in which the stiffening effect was decreased and m-value increased. There is a relation that the binder with lesser stiffening effect, but greater m-value has thermal cracking resistance and good ability to overcome high thermal stresses. Figures 7 and 8 illustrated the data that adding lignin is able to form an anti-cracking performance at freezing temperature. This must be due to the lignin fibers that act as the binding agent, making the mixtures thicker and more viscous. According, Wu (2020) found that partial replacement of lignin to the binder can promote stiffness, hence increasing the cohesion between the molecule.

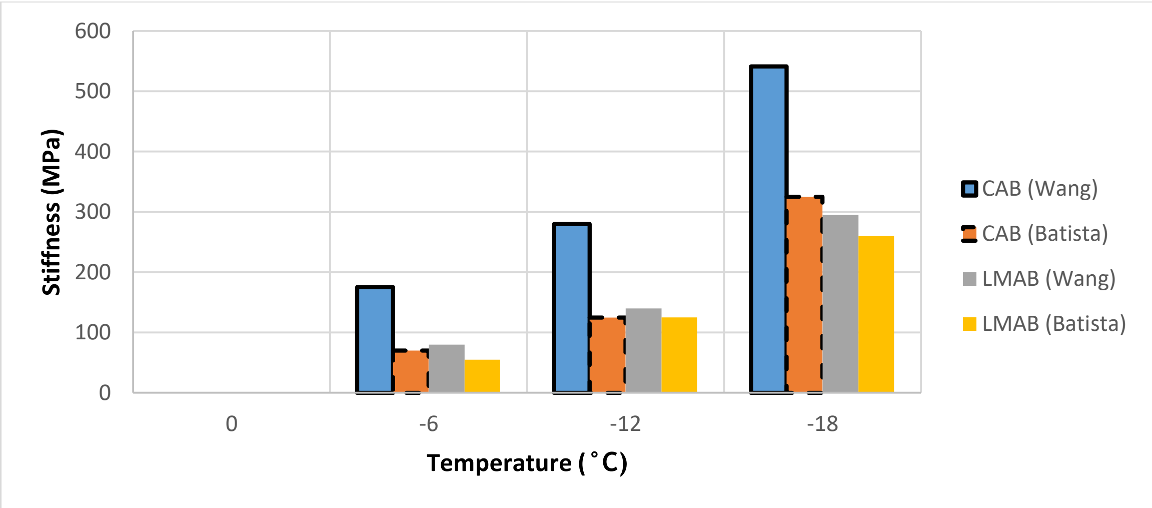

Figure 7: The stiffness effect (MPa) of CAB and LMAB

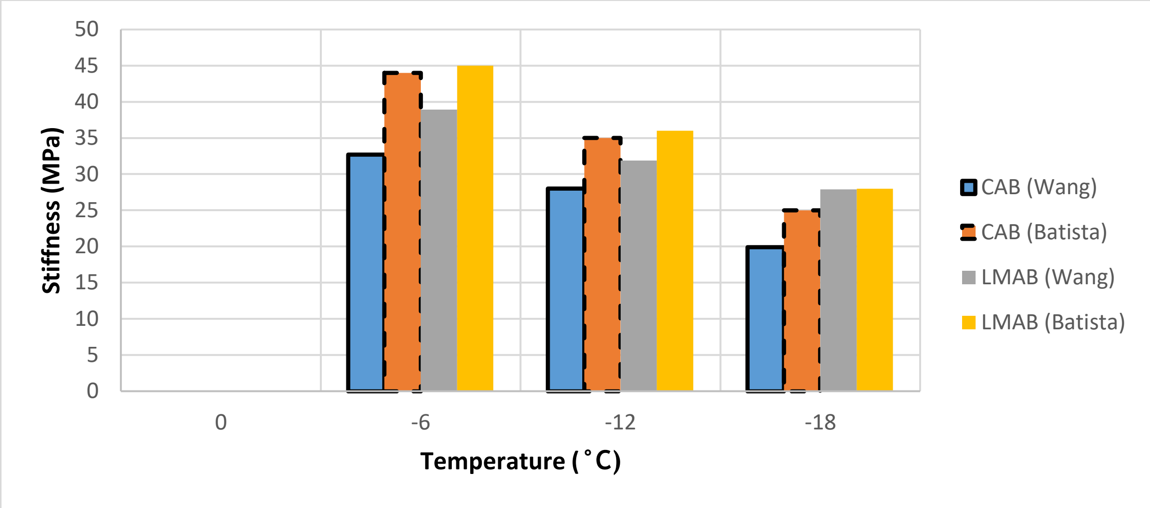

Figure 8: The stiffness effect (m-value) of CAB and LMAB

The stiffening effect is also related to penetration value and ductility that, as if the stiffness is higher, it would result in low penetration value and ductility. As more lignin concentration is added to the binder mixtures, the greater the stiffness would be [20]. The good in this significant increase of stiffening effect is that it helps to enhance the permanent deformation resistance without deteriorating other physical or rheological properties of the binder.

3.7 Discussion

This study comprehensively defines the workability of lignin as a renewable binder agent in asphalt mixtures through data collections from previous journals and articles of the same area of study. A series of physical and rheological performances have been analyzed from the results of laboratory tests conducted by former researchers to compare the properties of LMAB and CAB. Based on the results, the following findings have been obtained is the incorporation of lignin in asphalt binder, mainly at 6 wt.%, gives a lower penetration value to LMAB compared to CAB, hence increasing the stiffness effect, making the modified binder thicker. The softening point of LMAB has slight changes from CAB, however, as the lignin content increases, the softening point also increases. Moreover, LMAB is able to display better performance in higher temperatures. After that, LMAB shows higher viscosity than CAB, which indicates the higher temperature is required for the production and mixing process with aggregates, approximately at 1 wt.% lignin concentration.

However, incorporating lignin into the asphalt binder can still be recommended as a renewable binder. Lignin particles contain high anti-oxidants molecules which allow higher aging resistance to LMAB than CAB, as the lignin concentration in the mixtures increases. The low carbonyl index is able to slow down the speed of aging of bitumen in hot mix asphalt mixtures. LMAB has an increment in rutting resistance at high temperatures and a great stiffening effect at low temperatures. Considering lignin into the hot mix asphalt mixtures would result in higher performances if the manufacturing cost is not considered. The optimum concentration of lignin to be added into asphalt binder mixtures is between 1 wt.% to 6 wt.%. Nevertheless, an equal or partial replacement is also suitable regarding the virgin asphalt binder grade.

4. Conclusion

The conclusion is, the results of the study show the effectiveness of lignin as a renewable binding agent in asphalt binders, show better physical and rheological properties than conventional asphalt binders, and allow the determination of the optimum percentage of lignin as a modifier in asphalt binders., the addition of lignin positively affects the performances of LMAB by physical and rheological. It can act as a reinforcement that helps to bind the hot mix asphalt mixtures well and demonstrates better workability than the conventional binder. Thus, with lesser emission produced, lignin is assured to be an effective alternative binder replacement for existing petroleum-based asphalt binder.

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