

## Article

# Effect of Lignin Type as an Additive on Rheology and Adhesion Properties of Asphalt Binder

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**Abstract:** Utilization of alternative asphalt binders and additives from renewable sources, given the scale and the impact of the asphalt pavement industry, is an important step toward a sustainable future for the surface transportation infrastructure. Among several sources available for harvesting sustainable construction materials, bio-based materials from agricultural feedstock are known to be one of the most reliable, renewable, environmentally friendly, and economically feasible solutions to achieve this goal. Lignin, one of the most abundant materials in nature, is the byproduct of several industries, specifically pulp processing and biofuel production facilities. Given its physical properties, the use of lignin as a partial replacement for petroleum-based asphalt binder has been studied and proven promising. However, lignin's properties vary depending on its source and processing techniques. Therefore, incorporating lignin in asphalt binders can result in different mechanical properties, depending on its type and chemical composition. The present study was undertaken to evaluate the effect of three different lignin types, when used as an asphalt binder modifier, on the rheological properties of the asphalt binder, aging characteristics, and its adhesion to different aggregates. This study's findings showed that, when incorporated in an asphalt binder at the same amount, different lignin types have significantly different effects on asphalt binder blends' rheological, aging, and adhesion properties. Different rheological, aging, and adhesion properties of the binders result in different mechanical characteristics in asphalt mixes containing lignin-modified asphalt binders.



**Citation:** Ghabchi, R. Effect of Lignin Type as an Additive on Rheology and Adhesion Properties of Asphalt Binder. *Solids* **2022**, *3*, 603–619. <https://doi.org/10.3390/solids3040038>

Academic Editor: Joaquim Carneiro

Received: 20 September 2022

Accepted: 20 October 2022

Published: 26 October 2022

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**Keywords:** biomaterials; plant-based asphalt binder; lignin; rheology; adhesion; cracking; rutting

## 1. Introduction

Natural resource conservation, environmental concerns, fluctuating prices of fossil-based raw materials, and the need for a resilient construction material supply chain have spurred the pavement industry to research a new generation of construction materials that are environmentally friendly, renewable, and sustainable. Among different alternatives, in view of their competitive cost, low negative environmental impact, and renewable nature, plant-based biomaterials are identified as reliable and sustainable material feedstock [1]. Lignin, the second most abundant plant polymer found in nature, is the byproduct of several agricultural and forestry product industries, specifically pulp and biofuel processing facilities [2,3]. The paper pulping industry alone is responsible for the annual production of between 50 to 70 million tons of lignin [2]. Other than 2% of this amount which ends up in the chemical industry market, the remaining portion is used for applications, such as fuel, which do not utilize its full potential as a valuable raw material alternative. Lignin, a complex cross-linked phenolic polymer found in plants' cell walls, provides stiffness to the plant structure by binding cellulose and hemicellulose. The chemical structure of lignin, as a heterogenous phenylpropanoid macromolecule, consists of methoxy, phenolic hydroxyl, and terminal aldehyde groups as side chains [4].

Due to its inconsistent molecular structure and lack of well-defined characteristics, the application of lignin as a polymer in value-added products is found to be challenging. More

specifically, attributions such as difficulties associated with processing, high brittleness, low reactivity, and structural heterogeneity have limited its use as a polymer in advanced materials [5]. However, thanks to its chemical structure, lignin is used as an adhesive, an emulsifier, or a dispersing agent [6]. Additionally, given the physical properties and compatibility of lignin with asphalt, mainly due to similarities with the chemical structure of petroleum-based asphalt binder, its use as a partial replacement for asphalt or as an additive has been studied in the past and is proven promising [2,7–16]. Furthermore, due to its phenolic nature, the lignin's polymer acts as an antioxidant and provides a reduction in oxidation rate when present in asphalt binder [17]. The extent of its antioxidant effect largely depends on the lignin source and the processing technique used for harvesting it, affecting its chemical composition. Because of lignin's cross-linked polymeric structure, incorporating it in asphalt binders and mixes is found to raise the viscosity and high-temperature Performance Grade (PG) of the binder blends and improve their resistance to rutting [9–11,16,18]. While increasing the asphalt binder's stiffness was found to be advantageous for improving the resistance of the mixes to rutting, an elevated brittleness as a result of incorporating lignin was reported to negatively affect the resistance of the mixes to premature thermal and fatigue cracking [10,19,20]. It has also been reported that incorporating lignin in asphalt mixes can cause a delay in the oxidation process of the asphalt binder and, therefore, may be considered an antioxidant [9–11]. It can be concluded that the previous studies reported different ranges and extents of changes in the properties of asphalt binders as well as the mechanical and performance characteristics of asphalt mixes as a result of incorporating lignin. However, the source and the processes used for the production of lignin that highly affect its chemical compositions are not studied as important parameters influencing the properties of the lignin-modified asphalt binders.

To this end, this study was undertaken to investigate the effect of the utilization of different types of lignin, when used with the same amount as an asphalt binder modifier, on the binder's properties. More specifically, the effect of lignin type as an asphalt additive on rheology, Superpave PG, aging characteristics, adhesion to different aggregates, and moisture-induced damage potential of modified and unmodified asphalt binders was studied.

## 2. Objectives

The main objective of this study was to investigate the effect of the lignin type on the mechanical properties and the adhesion characteristics of the asphalt binder. The specific objectives of this study were as follows.

1. Determine the effect of incorporating three different types of lignin, namely L1, L2, and L3, used at the same rate in a non-polymer-modified asphalt binder (PG 58-28) on its rheology, Superpave PG grade, oxidative hardening, and resistance to rutting, fatigue, and low-temperature cracking.
2. Evaluate the effect of L1, L2, and L3 blended at the same rate with a PG 58-28 asphalt binder on its adhesion with different aggregate types, namely granite and quartzite, and binder-aggregate systems' resistance to moisture-induced damage.

## 3. Materials and Methods

### 3.1. Study Plan

Lignin, when incorporated in an asphalt binder, is known to enhance its stiffness, improve its resistance to rutting, delay aging, change adhesion behavior, and negatively affect its resistance to cracking [8,10,11,16,18–20]. However, the effect of the lignin type on the extent of the aforementioned observations is not well-established. It is expected that the changes observed in the properties of an asphalt binder, namely high-temperature (H-T) and low-temperature (L-T) PG, the propensity to aging, and adhesion to aggregates to depend on the lignin type and its composition when incorporated in a virgin asphalt binder. To study the effect of lignin type on the foregoing binder properties, a PG 58-28 asphalt binder (non-PMB), widely used in northcentral states, was selected. Three types of

lignin, namely L1, L2, and L3, having different compositions produced by using different processes, were collected from a biofuel production facility. Each lignin type (L1, L2, and L3) was blended separately with a virgin PG 58-28 binder at a rate of 20% by the weight of the binder. A dynamic shear rheometer (DSR, instrument model, Anton Paar USA Inc., Ashland, VA, USA) and a bending beam rheometer (BBR, BBR2s, Applied Test Systems, Butler, PA, USA) were used to evaluate the rheological properties of the lignin-modified and neat binder blends. As a result, complex shear modulus ( $G^*$ ), phase angle ( $\delta$ ), flexural stiffness ( $S$ ), creep compliance ( $m$ ), PG grade, and aging sensitivity of the binder blends were determined. In addition, the effect of lignin type on the sensitivity of the lignin-modified asphalt binder was determined. Finally, the effect of the lignin type incorporated in the virgin binder on its adhesion to different aggregates, namely granite, and quartzite, was determined. Additionally, the moisture-induced damage potential of the binder-aggregate systems was determined by conducting the binder bond strength (BBS) test on dry samples and those tested after moisture conditioning. Figure 1 presents the tested materials, applied methods, and the workflow pursued in this study.

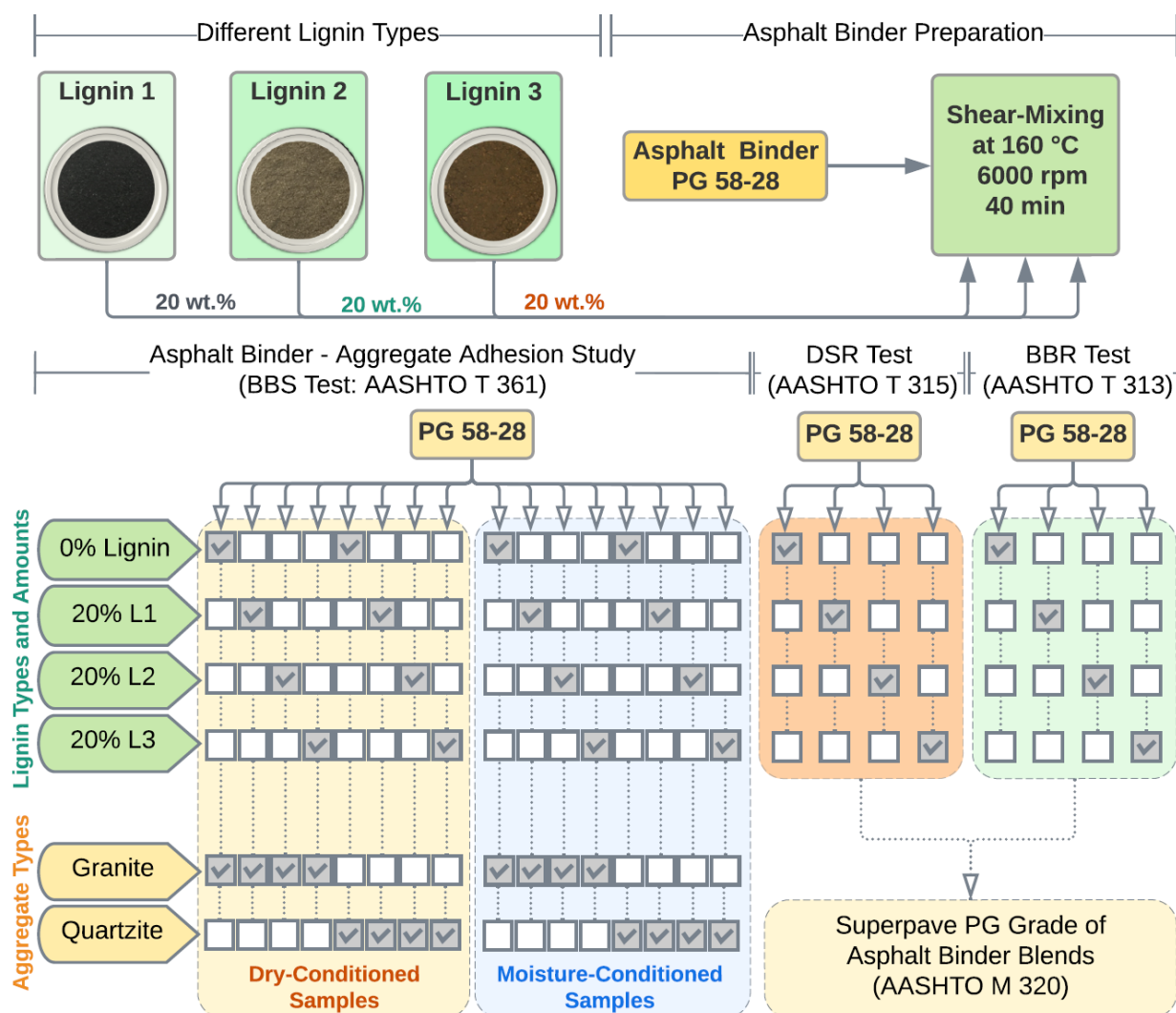
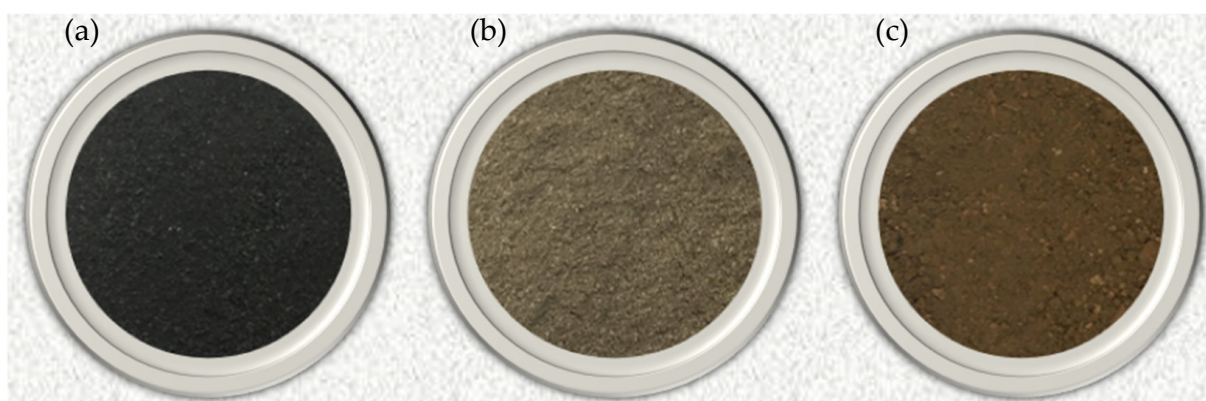


Figure 1. Work Flow and Test Matrix.

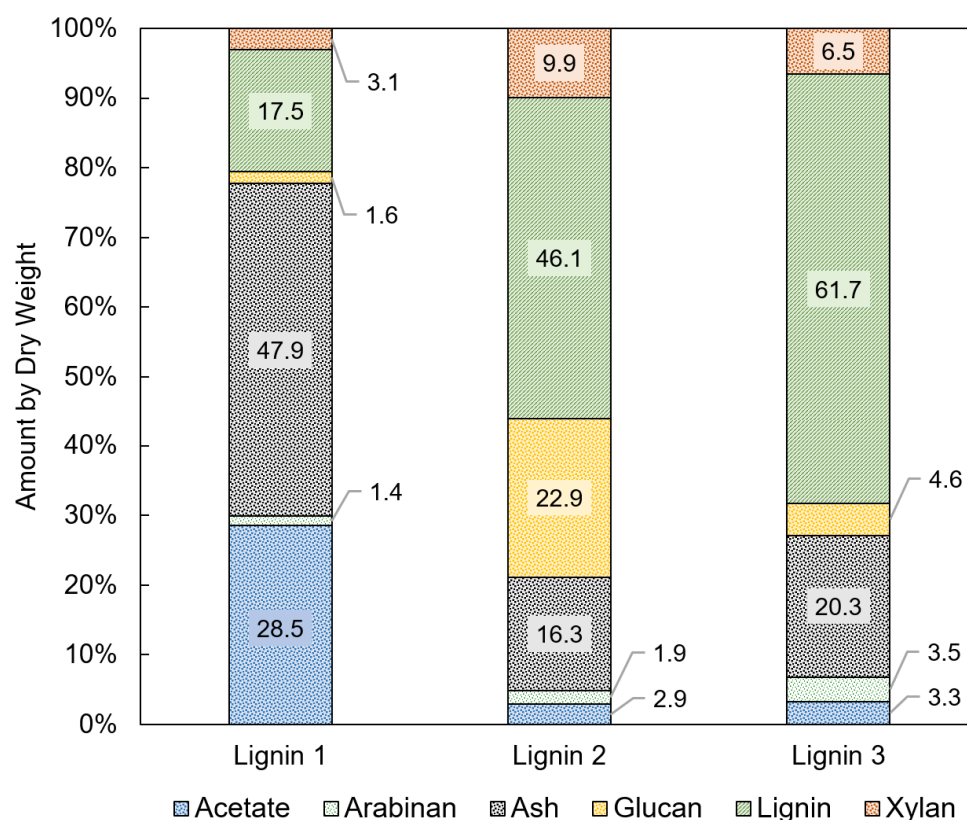
### 3.2. Materials

#### 3.2.1. Lignin

Three types of lignin, namely L1, L2, and L3, having different compositions produced by using different processes, were collected from a biofuel production and cellulosic biorefinery facility (Figure 2). A proprietary pretreatment followed by enzymatic hydrolysis was applied for the production of the L1, L2, and L3 as a part of the cellulosic ethanol process. Figure 3 summarizes the compositions of L1, L2, and L3 based on their dry weights.



**Figure 2.** Photographic Views of (a) Lignin 1; (b) Lignin 2; and (c) Lignin 3.



**Figure 3.** Compositions of Lignin 1, Lignin 2, and Lignin 3.

As can be observed from Figure 3, six important components present in lignin are acetate, arabinan, ash, glucan, lignin, and xylan. Acetate is mainly present in the form of cellulose acetate, among others. It is a photochemically degradable naturally occurring plastic. Arabinan is a neutral pectic side chain and an important cell wall constituent that plays an important role as a reinforcing matrix material [21]. The major constituent of ash is



silica. While ash can react with acids, it is considered an impurity that needs to be treated, as its presence in high amounts can result in extra wear and tear in the equipment used for grinding it [22]. Glucan, a polysaccharide, depending on its source, can be found in both water-soluble and insoluble forms. Glucans are major plant cell wall structural constituents in many plant cell types [23]. Lignin is an important polymer playing an essential role in the formation of a plant's stability and structure. More specifically, the rigidity and durability of the plant's wood and bark are because of the lignin's mechanical properties and chemical stability. Xylan and cellulose are the major constituents of hemicellulose (more than 30% by weight), which are found in the cell walls of land plants. Hemicelluloses' function is to interact with cellulose and lignin in land plants resulting in strengthening their cell walls [24].

### 3.2.2. Asphalt Binder

The asphalt binder considered in the study (PG 58-28) was collected from a local material supplier and used to prepare asphalt binder blends. The specific gravity of the collected asphalt binder reported by the asphalt refinery was 1.032.

### 3.2.3. Incorporating L1, L2, and L3 in Asphalt Binder

Asphalt binder blends were prepared by heating the PG 58-28 asphalt binder in different containers in an oven at 160 °C. Predetermined amounts of L1, L2, and L3 (20% by the weight of asphalt binder) were added to the liquid binder while being shear-mixed at 6000 rpm for 40 min, consistent with the procedure recommended by Gao et al. [10]. It should be noted that the aforementioned process was also applied to shear-mix the neat binder (PG 58-28 without any lignin). This process was adopted to exclude the effect of mixing and aging when comparing the neat binder with the other binder blends containing different types of lignin. To minimize the aging of the asphalt binders during the heating and mixing processes, the asphalt containers were kept covered when possible.

### 3.2.4. Aggregates

Granite and quartzite rocks were collected from local aggregate quarries, sawn into tiles approximately 25 mm thick and used for sample preparation and conducting aggregate-binder adhesion tests. The collected rocks were selected to have a minimum dimension of 300 mm to provide sufficient surface for preparing the test specimens.

## 3.3. Test Methods

### 3.3.1. Rolling-Thin Film Oven Procedure

Short-term aging of asphalt binder blends was carried out in the laboratory using a rolling-thin film oven (RTFO) in accordance with AASHTO T 240 standard method [25] to replicate asphalt binder oxidation in an asphalt plant and during construction. For this purpose, an approximate mass of  $35 \pm 0.5$  g of asphalt binder was poured into special glass bottles and aged at 163 °C using the RTFO equipment for a duration of 85 min. RTFO-aged asphalt binder was then collected and used for testing or long-term aging.

### 3.3.2. Pressure Aging Vessel Procedure

Pressure aging vessel (PAV) procedure in accordance with AASHTO R 28 standard practice [26] was carried out on the RTFO-aged asphalt binder samples to simulate five to ten years of long-term in-service aging. For this purpose,  $50 \pm 0.5$  g of RTFO-aged asphalt binder blend was poured into the PAV plates and aged at a temperature of 100 °C for 20 h while kept under 2.07 MPa air pressure.

### 3.3.3. Dynamic Shear Rheometer Test

A dynamic shear rheometer (DSR) was used to test asphalt binder blends in accordance with AASHTO T 315 standard method [27] to measure their complex shear modulus ( $G^*$ ) and phase angle ( $\delta$ ) at different temperatures. The rheological parameters measured at

high temperatures were used for the calculation of the rutting factor ( $G^*/\sin\delta$ ), and those measured at intermediate temperatures were used for determining the fatigue parameter ( $G^*\sin\delta$ ). In addition, calculated rutting factor and fatigue parameters were used to determine binder blends' high-temperature (H-T) grade and H-T Superpave PG in accordance with AASHTO M 320 standard specifications [28].

### 3.3.4. Bending Beam Rheometer Test

The PAV-aged asphalt binder blends were tested in a bending beam rheometer (BBR) in accordance with AASHTO T 313 standard method (AASHTO, 2019). Asphalt binder blends' creep compliance ( $m$ ) and their flexural stiffness ( $S$ ) measured 60 s after load application ( $m_{60}$  and  $S_{60}$ ) were used to determine the continuous low-temperature (L-T) grade and L-T Superpave PG in accordance with AASHTO M 320 standard specifications [29].

### 3.3.5. Binder Bond Strength Test

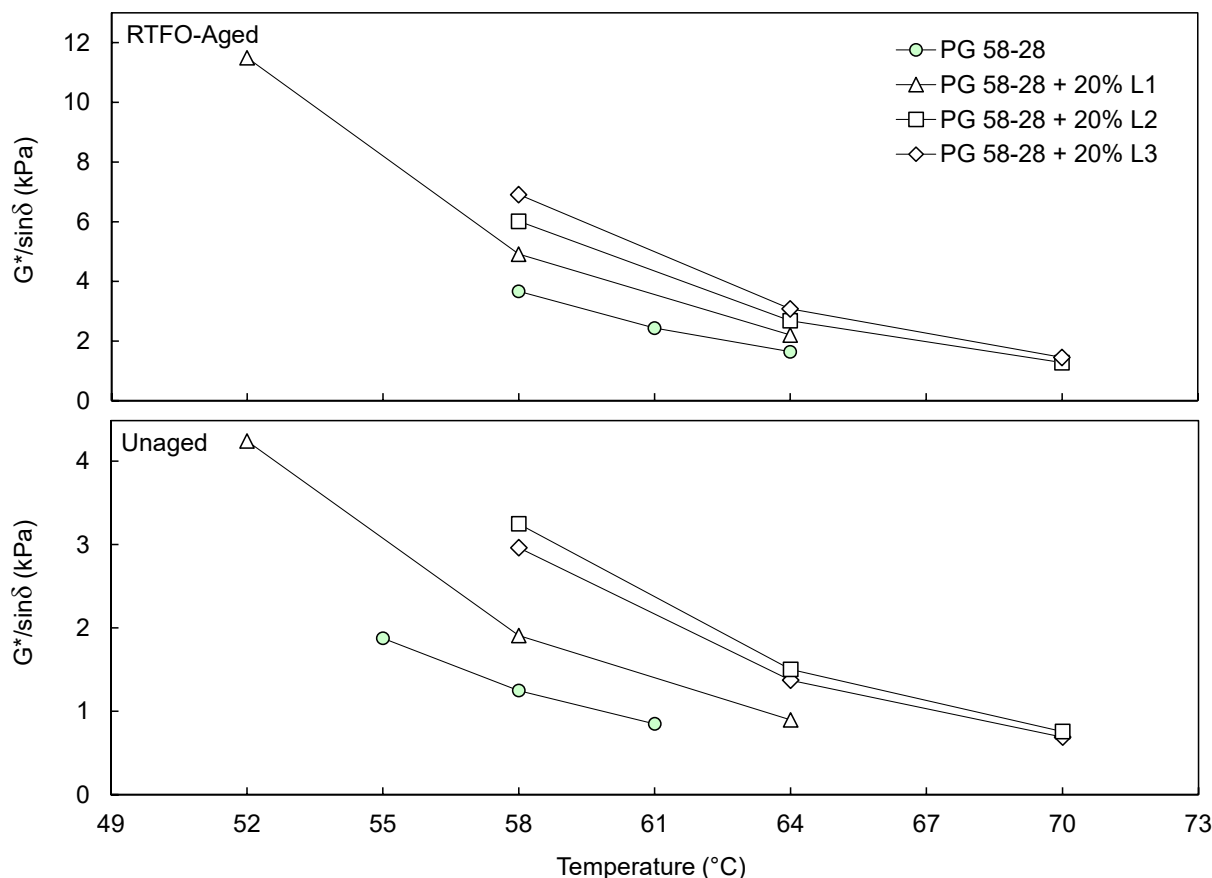
Binder bond strength (BBS) tests were conducted in accordance with the AASHTO T 361 standard method [30] to quantify the adhesion present in the asphalt binder-aggregate interface. In this method, the pull-off strength (POS) measured by testing binder-aggregate systems in dry ( $POS_{Dry}$ ) and moisture-conditioned states ( $POS_{Wet}$ ) were used to determine the adhesion quality and the susceptibility of the asphalt-aggregate systems to moisture-induced damage. Tests on both dry and moisture-conditioned specimens were conducted at 25 °C. To avoid false POS readings due to the effect of the capillary pressure formation in the asphalt binder-aggregate interface, pull-off tests on the moisture-conditioned specimens were carried out while they were submerged in the water. To ensure the test repeatability and data quality, a minimum of ten acceptable POS values measured for each asphalt binder-aggregate system were separately recorded for dry-conditioned and moisture-conditioned specimens, a total of 160 tests.

## 4. Results and Discussion

### 4.1. Resistance to Rutting

Figure 4 presents the rutting factors determined at different temperatures for unaged and RTFO-aged PG 58-28 asphalt binders containing 20% L1, 20% L2, and 20% L3. From Figure 4, it is evident that incorporating 20% of any type of lignin (L1, L2, or L3) resulted in an improved rutting factor compared to neat PG 58-28. This observation indicates an enhanced resistance of asphalt binder to rutting as a result of incorporating lignin in it. For instance, the rutting factor of the unaged PG 58-28 measured at 58 °C (1.25 kPa) was found to increase by 53%, 160%, and 137% due to mixing it with 20% L1, 20% L2, and 20% L3, respectively. Similarly, testing RTFO-aged binder blends at 58 °C indicated that incorporating 20% L1, L2, or L3 in PG 58-28 resulted in 34%, 64%, and 88% improvement in rutting factor of the neat binder (3.67 kPa), respectively. As a polyphenylpropanoid macromolecule containing aromatic monomers and branched side chains, lignin contains several different functional groups, which enables it to interact with asphaltenes present in the binder [31] and increase its resistance to deformation. This is consistent with the findings reported in the literature [9,32–34]. However, the extent of the improvement in the resistance of the asphalt binder to rutting as a result of incorporating lignin in it was found to be a function of the lignin type. For example, L2 was found to be the most effective lignin type in improving the rutting resistance of the unaged asphalt binder. However, when the binder blends were RTFO-aged, L3 was found to be the most effective lignin in improving the rutting resistance. In other words, while incorporating L2 in unaged PG 58-28 resulted in the highest rutting factor among other binder blends, after conducting RTFO aging, asphalt binder containing L3 exhibited the highest rutting factor. This observation indicates that L2 was capable of reducing the aging rate of an asphalt binder more effectively than the other lignin types. The higher effectiveness of L2 in improving the resistance to rutting compared to other lignin types and its superior aging retardation performance was attributed to its high lignin, xylan, and glucan contents; these three compounds are known to have high

structural stiffness and an antioxidant effect that contribute to superior rutting performance and aging retardation. This clearly indicates that the lignin type and its composition largely affect the extent to which it acts as an anti-aging compound and its rutting performance.



**Figure 4.** Rutting Factors ( $G^*/\sin\delta$ ) of Unaged and RTFO-aged PG 58-28 and that Containing Different Amounts of L1, L2, and L3 Measured at Different Temperatures.

#### 4.2. Sensitivity to Aging

Figure 5 presents a graphical comparison between the rutting factors of different asphalt binder blends measured at different temperatures before and after undergoing the RTFO procedure as an indicator of the sensitivity of asphalt binders to short-term aging. It was observed that the rutting factors measured for RTFO-aged asphalt binder blends at different temperatures were 1.7 to 3 folds higher than those of unaged asphalt binder blends. In addition to the qualitative aging susceptibility comparison shown in Figure 4, asphalt binder blends' sensitivity to oxidative aging was determined by calculation of each binder blend's Oxidative Hardening Index (OHI) from Equation (1) [35].

$$OHI = \frac{\sum_{i=1}^n (G_i^* \sin\delta_i)_{RTFO}}{\sum_{i=1}^n (G_i^* \sin\delta_i)_{unaged}} \quad (1)$$

where  $n$  is the number of temperatures under which the complex shear modulus and phase angles were measured, and  $G_i^*$  and  $\delta_i$  are the complex shear moduli and phase angles, respectively, at the  $i$ th test temperature. Equation (1) and the  $G^*$  and  $\delta$  values obtained from conducting the DSR tests were used to determine the OHI values of asphalt binder blends. A summary of the OHI values determined for the asphalt binder blends is shown in Figure 6.

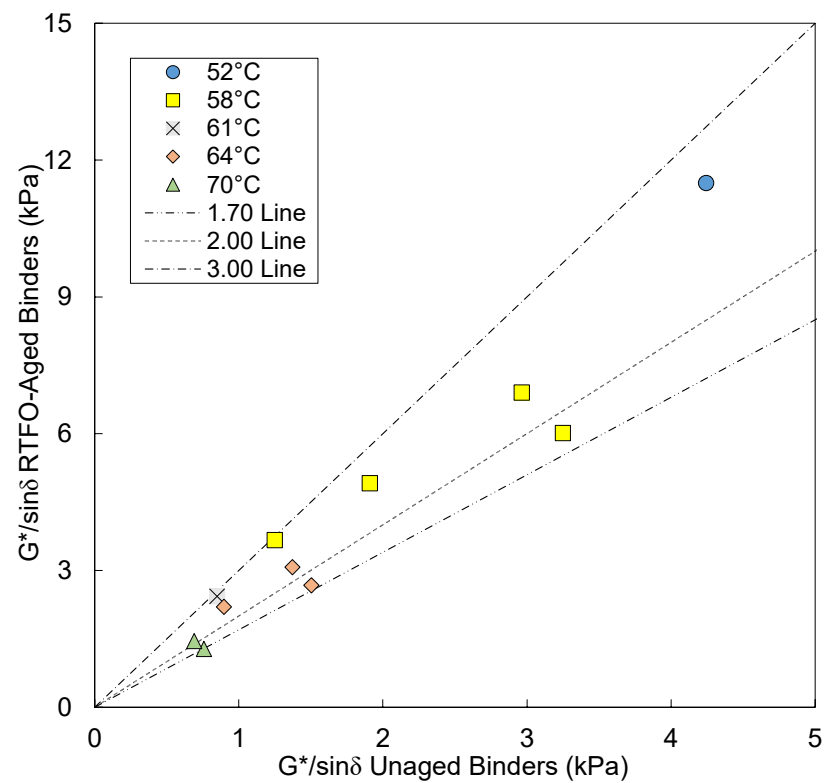


Figure 5. Rutting Factors of Unaged and RTFO-aged Binders at Different Temperatures.

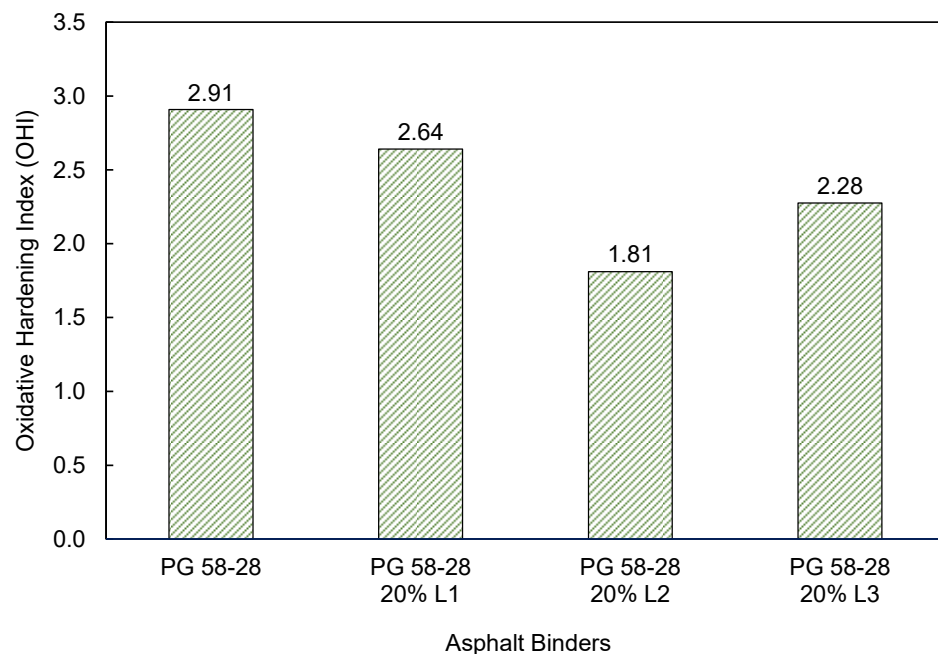


Figure 6. Oxidative Hardening Indices of Binder Blends.

From Figure 6, it was concluded that the OHI value of the PG 58-28 asphalt binder containing no lignin additive (2.91) was higher than that of other binder blends containing 20% of different lignin types. A higher OHI value indicates a higher potential for oxidative hardening. In other words, under the same aging condition, PG 58-28 oxidizes, ages, and embrittles at a higher rate compared to binder blends containing different lignin types. In contrast, a blend of PG 58-28 asphalt binder and 20% L2 exhibited the lowest OHI (1.81) among the tested binder blends. In other words, PG 58-28 containing 20% L2 is



expected to have the least susceptibility to aging when compared to other binder types. This observation is consistent with the data presented in Figure 4. From Figure 4, the binder blend containing 20% L2 exhibited the highest rutting factor before aging. However, after aging, its rutting factor measured at different temperatures ranked as the second highest among tested binder blends. The foregoing discussion suggests that incorporating L2 in PG 58-28 binder effectively reduced the oxidation rate in asphalt binder and had a superior aging retardation performance compared to other lignin types (L1 and L3). It should be noted that the aging rate reduces by a reduction in oxidation rate in maltenes due to a reduced formation of carbonyls (carboxylic acids, ketones, and anhydrides) as a result of the antioxidants present in the lignin [16]. However, the OHI values suggest that the lignin type heavily affected the extent of the anti-aging effect achieved by incorporating it in asphalt binder. This is attributed to the different amounts of phenolic compounds, known for their antioxidant effect, present in different lignin types. The phenolic compounds are mainly found in xylan and lignin. In addition, glucan is known to contain antioxidants [36] which can contribute to the reduction in oxidation rate. From Figure 3, it can be observed that L1, L2, and L3, respectively, contained 22.2%, 78.9%, and 72.8% cumulative amounts of glucan, lignin, and xylan as active ingredients. In other words, based on the foregoing discussions, L2 is the most effective, and L1 is the least effective antioxidant. This observation is consistent with the ranking of lignin types based on their OHI values observed in Figure 6. From Figure 6, it is evident that the OHI values of the PG 58-28 containing L1, L2, and L3 are 2.64, 1.81 and 2.28, ranking the oxidation-retarding effect of the lignin type with the same order as their composition suggests. An increase in the resistance of asphalt binder to aging is known to reduce the propensity of an asphalt mix to premature oxidative embrittlement leading to improved resistance to cracking.

#### 4.3. Resistance to Fatigue Cracking

Fatigue parameters ( $G^* \sin \delta$ ) of the PAV-aged asphalt binder blends considered in this study determined at different intermediate temperatures (16 °C, 19 °C, 22 °C and 25 °C) are summarized in Figure 7.

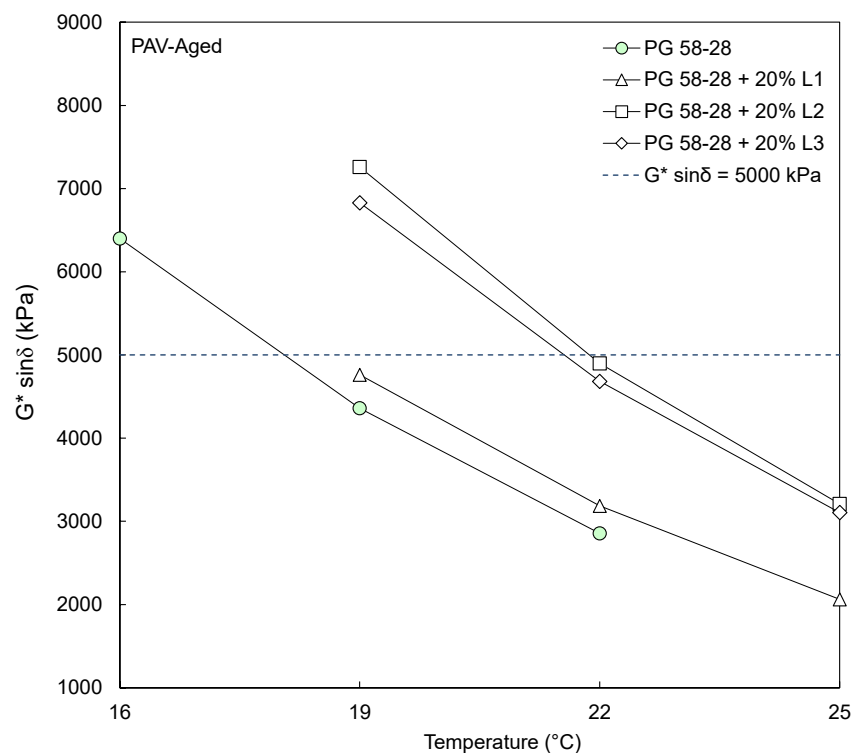
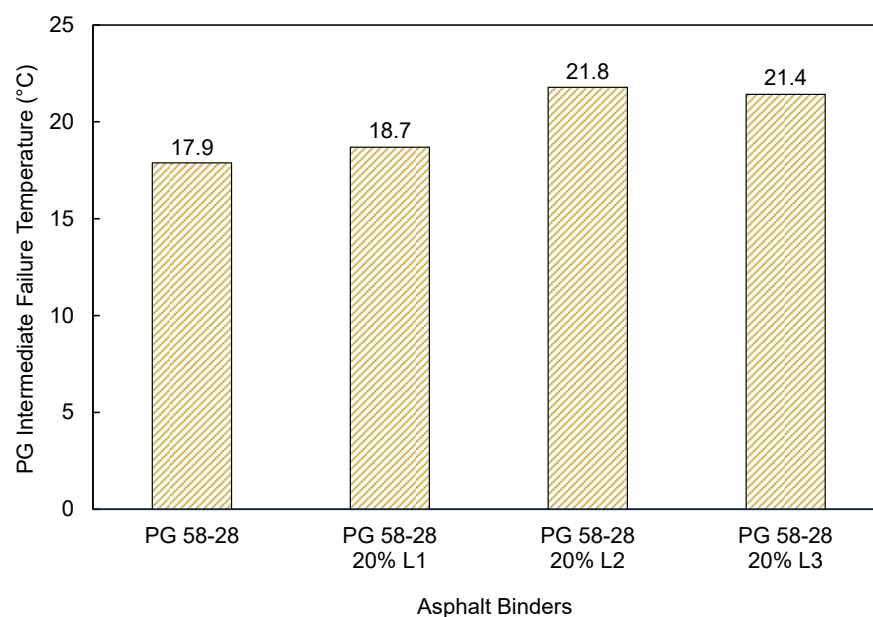


Figure 7. Fatigue parameters of PAV-Aged PG 58-28 Binder Blends.

It was observed that the fatigue parameter of the PG 58-28 binder measured at 19 °C and 22 °C increased by 9% and 12%, respectively, as a result of incorporating 20% L1. This observation was expected as the addition of the L1 to PG 58-28 was also observed not to contribute to the over-stiffening of the binder blend, as discussed in Section 4.1. In contrast, incorporating 20% L2 in neat PG 58-28 resulted in a 67% and 72% increase in its fatigue parameters at 19 °C and 22 °C, respectively. The observed increase in fatigue parameters of neat PG 58-28 as a result of blending it with 20% L2 was the highest among other blends, mainly attributed to the lignin's composition, as discussed in Section 4.2. This indicates a higher susceptibility of the PG 58-28 binder containing 20% L2 to fatigue cracking compared to other binder blends. Additionally, the aging retardation effect observed when the binder blends underwent short-term aging did not sustain when the long-term aging was carried out. For example, an asphalt binder blend of PG 58-28 and 20% L2 showed the highest stiffness before and the second highest after RTFO aging, indicating an anti-aging characteristic (Figure 4). However, the same blend after PAV-aging exhibited the highest fatigue parameter. As a result, it was concluded that despite an early-age improvement in aging retardation due to incorporating L2 in asphalt binder, this effect was not present when binder blends were subjected to long-term aging. This can be due to the chemical decomposition of some compounds present in L2 as a result of exposure to long-term aging. However, this can only be verified by conducting an elemental analysis of L2 before and after different aging procedures, which was beyond the scope of the present study. Finally, from Figure 7, the fatigue parameter of the neat PG 58-28 binder measured at 19 °C and 22 °C was found to increase by 57% and 64%, respectively, as a result of blending it with 20% L3. Therefore, one can conclude that while the latter lignin type had some adverse effect on the resistance of the PG 58-28 binder to fatigue cracking, it was not as severe as that observed when 20% L2 was blended with the neat binder. However, the rutting parameter measured for the RTFO-aged asphalt binder containing 20% L3 was the highest among other binder blends (Figure 4). This is an important observation, as incorporating L3 in a neat binder was capable of significantly improving rutting resistance with an intermediate adverse effect on the resistance of the blend to fatigue cracking.

Superpave asphalt binder specifications apply a PG intermediate failure temperature (IT) concept corresponding to the fatigue parameter of a PAV-aged asphalt binder at 5000 kPa. The lower the IT value, the higher the resistance of an asphalt binder to fatigue cracking. The IT values of the PAV-aged asphalt binder blends considered in this study were determined and summarized in Figure 8.

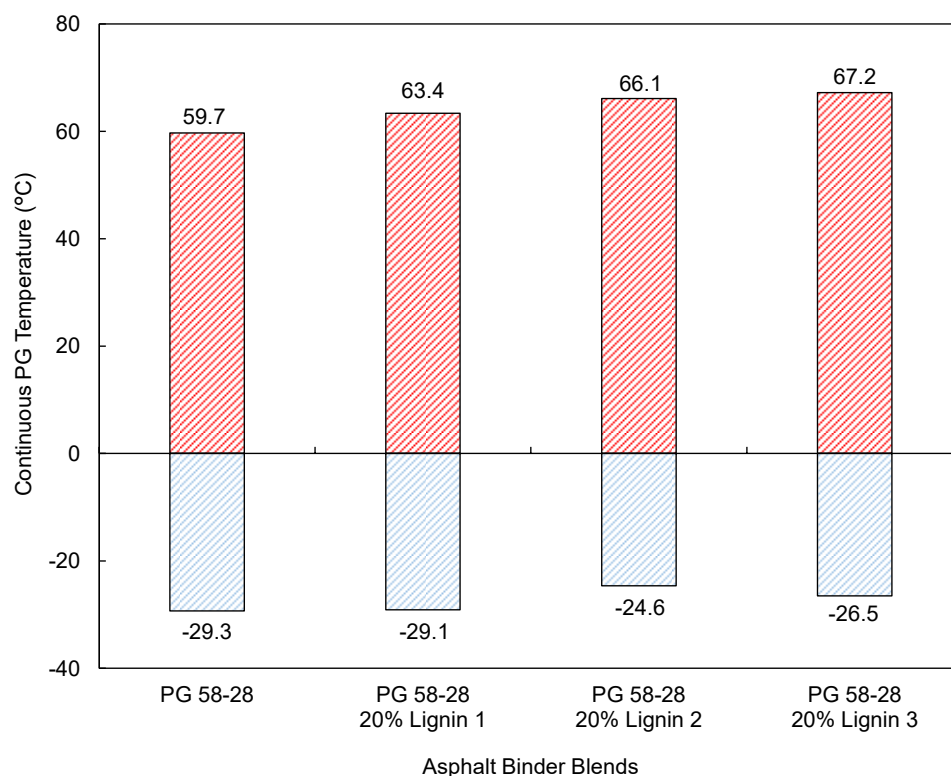


**Figure 8.** Intermediate Failure Temperature (IT) Values of PAV-Aged Binder Blends.

From Figure 8, it was observed that the IT value of the neat PG 58-28 asphalt binder (17.9 °C) slightly increased (by 5%) as a result of incorporating 20% L1 in it. This observation indicates that blending the neat binder with 20% L1 may not result in a meaningful reduction in the fatigue life of an asphalt mix compared to that prepared with a neat PG 58-28. In addition, it is evident that blending neat PG 58-28 with 20% L2 and 20% L3 resulted in TI values of 21.8 °C and 21.4 °C, respectively. This observation reveals that incorporating 20% of L1 or L2 in the binder may result in a more than 20% increase in TI of the neat binder, a significant adverse effect on resistance to fatigue cracking compared to the neat binder. High TI values measured for the two latter binder blends indicate they are not suitable for a climate where a PG 58-28 asphalt binder should be used. However, they may be acceptable candidates for regions with a higher intermediate temperature.

#### 4.4. Superpave Performance Grade of Binder Blends

Results of DSR and BBR tests conducted on asphalt binder blends were analyzed in accordance with AASHTO M 320 standard specifications [28] to determine continuous H-T and L-T grades of the neat PG 58-28 binder and that containing 20% L1, 20% L2, and 20% L3. A summary of the continuous PG temperatures of binder blends is presented in Figure 9.



**Figure 9.** Continuous Performance Grades of Binder Blends.

It was observed that incorporating 20% L1 in PG 58-28 asphalt binder resulted in a 3.7 °C and 0.2 °C increase in its H-T and L-T continuous grades, respectively. In other words, while incorporating 20% L1 in PG 58-28 resulted in a significant desirable increase in its H-T grade, at the same time, its L-T grade experienced an insignificant undesirable increase. It can be concluded that L1 as an asphalt binder additive has the potential to be used for a balanced improvement in the rutting resistance of an asphalt binder without significant adverse effects on its resistance to thermal cracking. While both H-T and L-T grades of the neat binder increased due to blending it with 20% L1, it did not result in any alteration in the Superpave PG grade of the neat binder. However, the blend resulted in a PG 58-28 binder, which was more resistant to rutting than the neat PG 58-28. In addition, it contained 17% less nonrenewable petroleum-based asphalt binder, which was replaced by

plant-based renewable lignin. This finding is significant as it shows the feasibility of partial replacement of an asphalt binder with a plant-based product with potential savings in cost and carbon footprint of pavement construction. In addition, from Figure 9, blending neat PG 58-28 asphalt binder with 20% L2 led to a 6.4 °C and 4.7 °C increase in neat binder's H-T and L-T grades, respectively. A significant increase in both H-T and L-T grades indicates an improved resistance to rutting and reduced resistance of the neat asphalt binder to low-temperature cracking after blending it with 20% L2. The Superpave PG grade of the binder blend containing PG 58-28 and 20% L2 was characterized as a PG 64-22. It underlines the fact that an asphalt binder suitable for a colder climate can be modified by blending it with 20% L2 to obtain another binder suitable for a warmer climate while reducing the use of petroleum-based binder by 17% through a partial binder replacement with the L2 lignin. Finally, incorporating 20% L3 in PG 58-28 asphalt binder resulted in a 7.5 °C and 2.8 °C increase in its H-T and L-T continuous grades, respectively. This indicates an increase in the H-T grade of the neat binder (an improved resistance to rutting) as a result of incorporating 20% L3 in the neat binder, which was by 1.7 times more than its adverse effect on its L-T grade (reduced resistance to cracking). The Superpave PG grade of the binder blend containing PG 58-28 and 20% L3 was characterized as a PG 64-22. However, the latter PG 64-22 binder's resistance to both rutting and thermal cracking was higher than the PG 64-22 binder produced by mixing PG 58-28 with 20% L2. It can be concluded that incorporating 20% L3 in PG 58-28 resulted in an asphalt binder blend with superior H-T and L-T grades compared to that produced by using 20% L2 in the same neat binder. In addition to the performance benefits, 17% savings in petroleum-based binders and carbon footprint can be achieved.

Table 1 presents a summary of the Superpave PG grades of the neat asphalt binder that contains 20% of different types of lignin, namely L1, L2, and L3.

**Table 1.** Superpave PG Grades of the Studied Binder Blends.

Base Binder	Lignin 1 (%)	Lignin 2 (%)	Lignin 3 (%)	Virgin Binder Replacement (%)	Superpave PG of Blend
PG 58-28	-	-	-	-	PG 58-28
PG 58-28	20	-	-	17	PG 58-28
PG 58-28	-	20	-	17	PG 64-22
PG 58-28	-	-	20	17	PG 64-22

#### 4.5. Adhesion of Binder Blends to Aggregates

Adhesion tests through applying a pull-out mechanism utilizing a BBS device were conducted on asphalt binder-aggregate systems to quantify the effect of incorporating different types of lignin (L1, L2, and L3) in the neat PG 58-28 asphalt binder on its adhesion to two different types of aggregates, namely granite and quartzite. The BBS tests were conducted on both dry and moisture-conditioned specimens, and their pull-off strength in dry condition ( $POS_{Dry}$ ) and after moisture conditioning ( $POS_{Wet}$ ) were determined. The measured pull-off strengths in dry and after moisture-conditioning the samples were used to determine the pull-off strength ratio (PSR) values ( $POS_{Wet}/POS_{Dry}$ ). The PSR values are analogous to the tensile strength ratio (TSR) values obtained from conducting indirect tensile strength tests on asphalt mixes [37]. Unlike the TSR test, which is used to evaluate the overall moisture susceptibility of an asphalt mix, PSR values can be used to spot incompatible binder-aggregate pairs at the component level.

##### 4.5.1. Adhesion to Granite

Average measured  $POS_{Dry}$ ,  $POS_{Wet}$ , and corresponding PSR values determined by conducting the BBS tests on binder-aggregate samples prepared using granite substrate and neat PG 58-28, and that containing 20% L1, 20% L2, and 20% L3 are summarized in Figure 10.

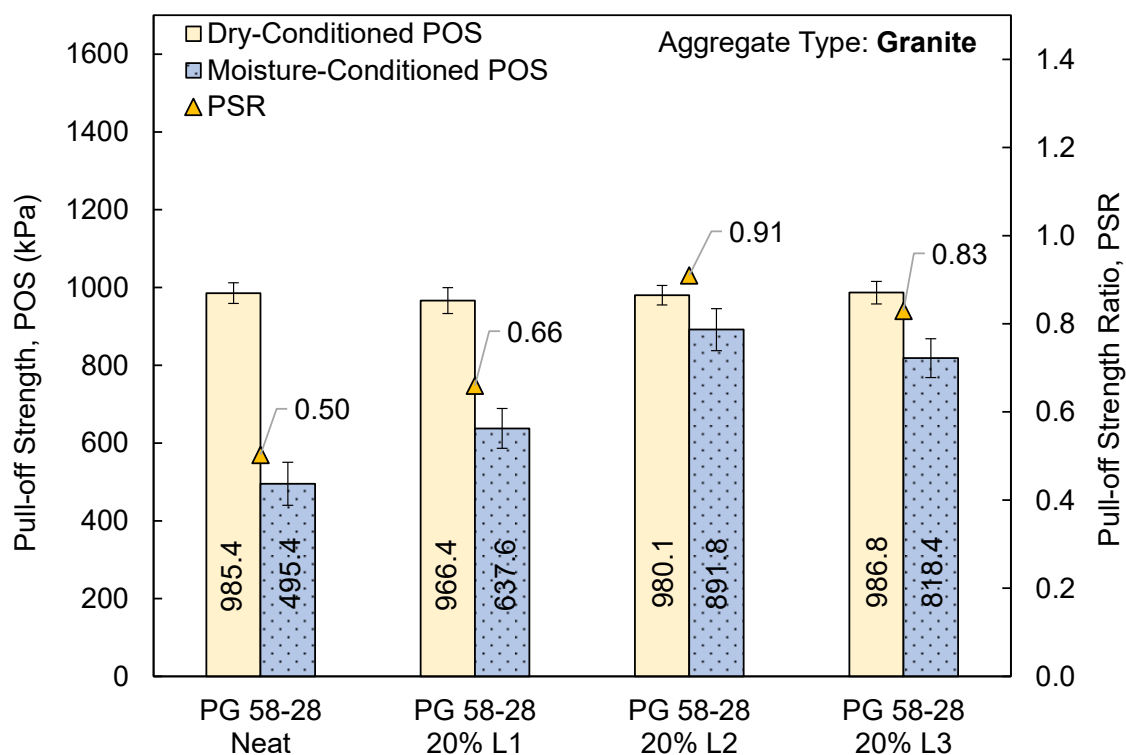


Figure 10. Average  $POS_{Dry}$ ,  $POS_{Wet}$ , and PSR Values of Binder Blend with Granite Aggregate.

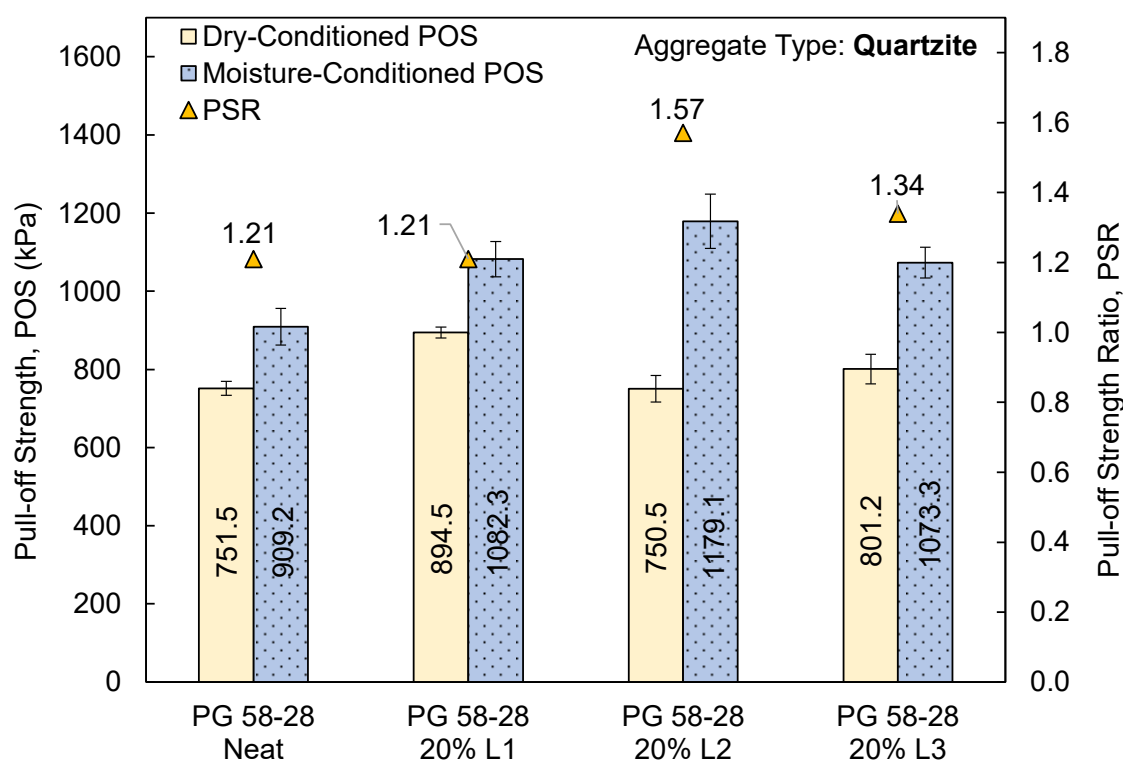
It was observed that the measured  $POS_{Dry}$  value of the samples prepared using neat PG 58-28 asphalt binder with granite (985.4 kPa) reduced by 2%, 1%, and 0%, after incorporating 20% L1, 20% L2, and 20% L3, respectively. Given the variability of the measured  $POS_{Dry}$  values, as shown by the error bars in Figure 10, it can be concluded that the  $POS_{Dry}$  value of the neat binder with granite remained statistically unchanged as a result of blending it with 20% L1, 20% L2, and 20% L3. Therefore, the resistance of the asphalt mixes containing PG 58-28 and granite considered in this study to adhesion-related distresses are not expected to be significantly altered as a result of using 20% of L1, L2, or L3 in asphalt binder. Asphalt binder-aggregate adhesion is known to be correlated with the resistance of asphalt pavements to raveling and its overall durability [38,39].

In addition, from Figure 10, it can be seen that incorporating lignin in the neat PG 58-28 resulted in an increase in its PSR value measured with granite (0.50) regardless of lignin type. More specifically, blending PG 58-28 with 20% L1, 20% L2, and 20% L3 and testing them with granite led to PSR values of 0.66, 0.91, and 0.83, respectively. This indicates that while aggregate-binder systems comprised of neat PG 58-28 asphalt binder and granite lost 50% of their pull-off strength as a result of moisture conditioning, the same system containing 20% of L1, L2, or L3 lost only 34%, 9%, and 17% of its tensile adhesive strength, respectively, due to the effect of moisture. Overall, one can conclude that the incorporation of 20% of different types of lignin considered in this study and granite was capable of improving the resistance of the binder blends with granite aggregate to moisture-induced damage compared with that of the specimens prepared with neat PG 58-28. It was also observed that L2 was the most effective lignin type in improving the resistance of the binder-granite system to moisture-induced damage.

#### 4.5.2. Adhesion to Quartzite

Average measured  $POS_{Dry}$ ,  $POS_{Wet}$ , and corresponding PSR values determined by conducting the BBS tests on binder-aggregate samples prepared using quartzite substrate and neat PG 58-28, and that containing 20% L1, 20% L2, and 20% L3 are summarized in Figure 11.





**Figure 11.** Average  $POS_{Dry}$ ,  $POS_{Wet}$ , and PSR Values of Binder Blends with Quartzite Aggregate.

From Figure 11, it was observed that the measured  $POS_{Dry}$  value of the samples prepared using neat PG 58-28 asphalt binder with quartzite (751.5 kPa) increased by 19%, 0%, and 7% after incorporating 20% L1, 20% L2, and 20% L3, respectively. Therefore, it can be concluded that the resistance of the asphalt mixes containing PG 58-28 and quartzite considered in this study to adhesion-related distresses is expected to increase as a result of using L1 and L3 in the blend and remain unchanged after incorporating L2 in the neat asphalt binder. Therefore, one can expect that the resistance of the asphalt mixes containing a PG 58-28 and quartzite aggregate to raveling improve as a result of using 20% L1 and L3 in the binder and to remain relatively unchanged when 20% L2 is used. It should be noted that lignin is a binding compound present in nature as a part of the plant structure. Foregoing observations show the compatibility of the lignin with the asphalt binder and the aggregates, making it an ideal plant-based additive for asphalt binder.

In addition, from Figure 11, it can be seen that incorporating lignin in the neat PG 58-28 resulted in no change or an increase in its PSR value measured with quartzite (1.21) regardless of lignin type. More specifically, blending PG 58-28 with 20% L1, 20% L2, and 20% L3 and testing them with quartzite led to PSR values of 1.21, 1.57, and 1.34, respectively. This indicates that while aggregate-binder systems comprised of neat PG 58-28 asphalt binder and quartzite gained 21% additional pull-off strength as a result of moisture conditioning, the same system containing 20% of L1, L2, or L3 also gained 21%, 57%, and 34% additional tensile adhesive strength, respectively, due to the effect of moisture. Overall, one can conclude that incorporating 20% of different types of lignin considered in this study, namely L1, L2, and L3, and granite was capable of improving the resistance of the binder blends with quartzite aggregate to moisture-induced damage compared with that of the specimens prepared with neat PG 58-28. It was also observed that L2 was the most effective lignin type in improving the resistance of the binder-quartzite system to moisture-induced damage.

According to Liu et al. [40], lignin contains high amounts of phenolic hydroxyls, aromatic rings, and hydroxyls (Lewis acid groups) which result in an acidic surface characteristic. This results in an acidic interface with aggregate and binder, leading to a polar

nature of the blend, forming a strong bond with aggregate, which naturally have a higher polarity than the neat binder [41].

## 5. Conclusions

The effect of incorporating 20% (by the weight of the binder) of three different types of lignin (L1, L2, and L3) with different compositions in a PG 58-28 asphalt binder on its rheology and its adhesion characteristics with different aggregates (granite and quartzite) was studied. Asphalt binder samples' rheology was characterized by conducting dynamic shear rheometer (DSR) and bending beam rheometer (BBR) tests, and their cracking and rutting potentials were evaluated. In addition, the adhesion of the asphalt binder blends with granite and quartzite and their moisture susceptibility were characterized by conducting BBS tests on dry and moisture-conditioned specimens. Based on the observations and discussions presented herein, conclusions were drawn as follows.

Incorporating 20% of all three lignin types in the PG 58-28 asphalt binder was found to lead to a substantial increase in the rutting factor of the neat binder and an improved resistance to rutting. This improvement was more pronounced when L2 and L3 lignin types were used.

Binder blends containing PG 58-28 asphalt binder and 20% of any of L1, L2, or L3 were found to be more resistant to oxidative aging compared to the neat PG 58-28 asphalt binder. However, a binder blend containing 20% L2 was found to be considerably more resistant to short-term oxidative hardening compared with other binder blends.

Incorporating 20% of any lignin type (L1, L2, and L3) in PG 58-28 asphalt binder was found to increase the fatigue parameter of the neat binder and reduced resistance to fatigue cracking. However, the increase in the fatigue parameter was the lowest among other blends when 20% L1 was used.

Blending neat PG 58-28 asphalt binder with 20% L1 was found to result in an asphalt binder with the same Superpave PG grade with higher resistance to rutting (H-T PG of 63.4 °C). This showed the feasibility of replacing 17% of the petroleum-based virgin binder with plant-based lignin resulting in performance parameters equal to or superior to that of the neat binder. The incorporation of 20% L2 or L3 in PG 58-28 resulted in a binder blend with a Superpave PG grade of PG 64-22.

In general, incorporating 20% of L1, L2, or L3 in the neat PG 58-28 asphalt binder was found to improve its adhesion to quartzite and granite aggregates in both dry and moisture-conditioned specimens, improving resistance to moisture-induced damage.

This study showed the effects of different types of lignin on an asphalt binder's rheological characteristics and adhesion properties with different aggregate types. Also, the feasibility of replacing up to 17% of a petroleum-based asphalt binder with a bio-based sustainable alternative while obtaining a final product with equal or superior properties to those of the neat binder was shown. It is recommended to characterize the asphalt binders and aggregates used in this study by quantifying their surface-free energy components to gain a better understanding of the adhesion and debonding mechanisms present at the interface of the asphalt binder-aggregate systems containing lignin. It is recommended to expand the scope of this work by characterization of asphalt mixes containing asphalt binder blends prepared using L1, L2, and L3.

**Funding:** This research was funded by POET LLC, grant number SA1800512.

**Data Availability Statement:** The data presented in this study are available on request from the corresponding author.

**Acknowledgments:** The study presented herein was conducted with financial support received from POET LLC. Also, the help and comments received from Steve Bly and Alex McCurdy (POET LLC) are acknowledged. The authors would like to thank Musharraf Zaman, Kenneth Hobson, and Syed Ashik Ali for their invaluable help. The contents of this article are the opinions of the authors and are not necessarily those of the sponsor.

**Conflicts of Interest:** The author declares no conflict of interest. The funder had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

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