Evaluation of Lignin-Bitumen Blend as a Sustainable Alternative for Asphalt Binder

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Abstract. To address the need to mitigate climate change and reduce carbon emissions, the road industry faces the challenge to reduce its dependence on fossil fuel-derived bitumen in asphalt pavement. This paper presents evaluation of lignin obtained from TNO FABIOLA^{TM} process as a sustainable alternative to traditional bituminous binders. DSC test determined the melting of the lignin, which aided in the selection of the blending temperature. The blending process achieved homogeneous dispersion and dissolution of lignin within bitumen, constituting 25 wt.% in the total binder. Rheological assessments, conducted using DSR revealed that the viscoelastic properties of the lignin-bitumen blend closely resembled those of penetration grade bitumen, suggesting the lignin potential as a viable substitute without compromising rheological performance. Furthermore, MSCR test results showed resistance to permanent deformation in the lignin-bitumen blend comparable to the penetration bitumen. Two asphalt mixtures were prepared, one with the reference PmB and another with the lignin-bitumen blend, as an alternative binder. Water Sensitivity Tests (WST) on both fresh and aged specimens revealed no detrimental effects on Indirect Tensile Strength Ratio (ITSR) values. These findings highlight the potential of lignin as sustainable alternative to bitumen, advancing road materials with reduced environmental impact.

Keywords: Lignin-bitumen blend · rheological properties · sustainable binder · multiple stress creep recovery (MSCR) test · indirect tensile strength ratio (ITSR) · stone mastic asphalt (SMA)

1 Introduction

The urgent need to combat climate change and reduce carbon emissions is driving innovation in the road construction industry. One of the challenges is to reduce the dependence on fossil fuel-derived bitumen. To address this, engineers and scientists are turning to biomass-derived materials, with lignin emerging as a promising alternative binder for asphalt pavement [1].

Recent research extensively explored the potential of lignin as a partial substitute for conventional bitumen in various applications [1–6]. Some studies reported lignin incorporation has shown promise in improving rutting resistance and low-temperature

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cracking, while others highlight the antioxidant properties of lignin and their impact on binder performance [2–4]. In 2015, a patent was filed for blending native and chemically modified lignin with bitumen, revealing the potential to partially replace bitumen to varying extents, with chemically modified lignin exhibiting characteristic rheological properties similar to polymer-modified bitumen [5, 6]. Increasing lignin content increased the viscosity of the blend, where 25 wt.% lignin content showed optimum viscoelastic properties [1].

Current study focused on preparing a lignin-bitumen blend using 'U10' lignin obtained from beech wood, partially substituting (25 wt.%) bitumen. DSC analysis revealed its melting point at 142 °C and the blending process ensured a homogeneous mixture. To assess the potential of the lignin-bitumen blend, a comprehensive rheological evaluations compared the blend to reference PmB and 70/100 penetration bitumen. The results showed that the viscoelastic properties of the lignin-bitumen blend closely resemble those of 70/100 bitumen, suggesting the viability of the lignin as a bitumen substitute without compromising the rheological performance. Furthermore, MSCR parameters predicted resistance to permanent deformation, with the blend showing improved performance compared to neat bitumen. The study extended its assessment to the asphalt level, evaluating Stone Mastic Asphalt (SMA-14) mixes. Water Sensitivity Tests (WST) on both fresh and aged samples revealed positive outcome in ITSR values.

2 Materials and Methods

2.1 Materials

Paving grade bitumen of penetration 70/100 and a polymer modified bitumen (PMB 65/105-60) provided by Breedongroup-Ireland were studied as reference bituminous binders in this study. Lignin-U10, processed from beech wood feedstock by TNO FABIOLA™ process was used for partial substitution (i.e., 25 wt.%) of bitumen. FABIOLA™ is an organosolv process that involves fractionation of biomass using low temperature acetone [7]. 'U10' is the number of the pilot run in which this specific lignin was produced. The lignin was brown fine powder at room temperature as delivered which could be used directly for blending without further drying or milling. The reference penetration 70/100 bitumen was used as a base bitumen to prepare the lignin bitumen blend. The blend is used to prepare Stone Mastic Asphalt (SMA) 14 slab and another reference mixture slab of SMA 14 was prepared using the PmB.

2.2 Methods of Preparation and Characterization

Blend Preparation: Lignin was blended with bitumen using a high shear mixing assembly followed by a low shear mixing according to the procedure described in the patent WO2015/137813. 2015 [5]. 25wt.% (i.e., by mass) lignin was added in penetration grade 70/100 bitumen. The bitumen was pre-heated in the oven for two hours at 140 °C and prior to blending was heated to 170 °C on a temperature-controlled heater plate. Lignin was gradually added using a sieve to control the particle size (<0.5 mm) and to improve the dispersion of the powder lignin during blending. After addition of the lignin, the lignin and bitumen are mixed at high shear for 5 min in 1000–1500 rpm followed by mixing at low shear for 30 min in 200–300 rpm.

Dynamic Shear Rheometer (DSR): The rheology of the original bitumen, a reference polymer modified bitumen and the lignin-bitumen blend were characterized by using DSR - Modular Compact Rheometer MCR 502 from Anton Paar. To obtain the master curves, two parallel plate test configuration was used with 8-mm plates of 2-mm gap for −10 to 30 °C and 25-mm plate of 1-mm gap for the temperatures 30–60 °C at a frequency of 0.1–400 rad/s. The DSR measurements were performed according to EN 14770–2022 where the measurements were carried out first by strain sweeps followed by frequency sweeps at different temperatures [8]. Parallel plate geometry of a diameter of 8 mm was used for the temperatures -10 to 30 °C and a diameter of 25 mm from 30 °C to 60 °C. Using the time-temperature-superposition principle, the frequency sweep data from different temperatures were shifted horizontally at a reference temperature of 20 °C to construct mastercurves of complex shear modulus |G*| and phase angle (δ) as a function of frequency.

MSCR Method: MSCR test was performed at 60 °C using DSR to evaluate the susceptibility of the binders for permanent deformation at high service temperature. The test was carried out according to the standard EN 16659 procedure in a plate-plate geometry of 25 mm diameter with a gap of 1 mm [9]. The binders were tested under shear creep and recovery at two stress levels of 0.100 kPa and 3.2 kPa at 60 °C, where the specimens were loaded at constant stress for 1 s, then the load was removed allowing to recover for 9 s. MSCR was performed first starting at the low stress level of 0.1 kPa for 10 creep-recovery cycles followed by an additional 10 cycles at 3.2 kPa. The MSCR parameters were calculated according to EN 16659 and presented in the result section.

Differential Scanning Calorimetry (DSC): DSC analysis was performed to determine the melting point of the lignin and to scan any additional thermal event. The DSC procedure for lignin was adapted from the ASTM standard -ASTM D4419-90(2021) [10]. To prepare a specimen, lignin sample of 10 ± 1 mg was taken into 50 µL DSC aluminium sample pans as shown in the Fig. 1.

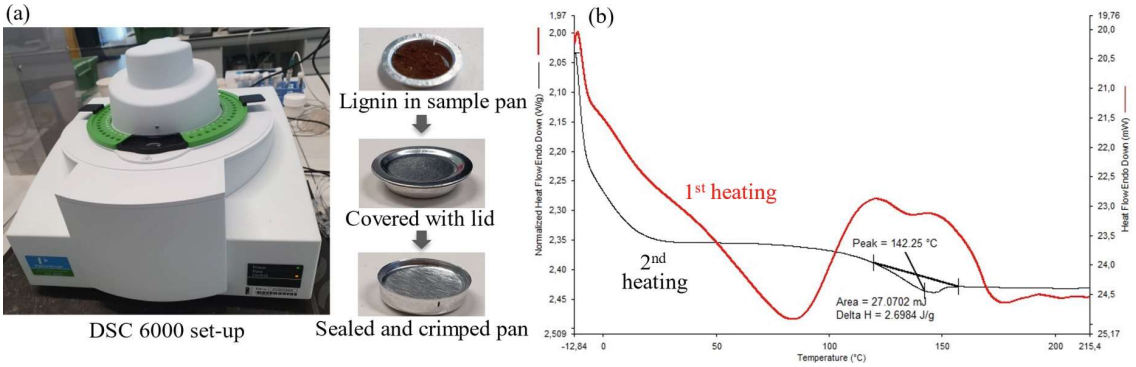

Fig. 1. (a) DSC set-up, sample preparation (left to right) to measure the thermal properties of lignin. (b) Thermal curve showing melting temperature of lignin derived from the heat flow as a function of temperature.

Prior to the DSC run the instrument was calibrated using a reference i.e. Indium sample and its melting temperature T_m (156.6 °C) was used as a calibration reference. The lignin was heated from −10 °C to 220 °C at a heating rate of 10 °C/ min. The heat

flow measured through the sample pan was compared with an empty reference pan and heat flow as a function of temperature was recorded.

Asphalt Preparation: In order to assess asphalt properties, two Stone Mastic Asphalt (SMA-14) slabs were prepared, one with PmB as a control mixture and the other using the lignin-bitumen blend with the same mix-composition. The asphalt mixtures were produced using the standard mix composition. Half of the asphalt specimens were subjected to long-term aging simulation at 85 °C in the oven for three weeks. Subsequently, a Water Sensitivity Test (WST) was conducted according to EN 12697-12:2018 on both fresh and aged specimens to evaluate their resistance to moisture damage.

3 Results

3.1 Binder Evaluation

Thermal Properties of Lignin-U10: Figure 1b showed heat flow curves from DSC temperature scans of U10-lignin. In the first heating, a broad evaporation endotherm indicated residual moisture and the second heating revealed the melting peak at 142 °C in the lignin. Melting point was a useful data to check and verify the mixing temperature for the lignin-bitumen blend preparation. Here the mixing temperature was 170 °C, above the melting point of lignin, ensuring proper dissolution into bitumen.

Mastercurves: The viscoelastic response of a 25wt.% lignin-bitumen blend overlapped with that of the original 70/100 bitumen (see Fig. 2a and b). This study demonstrated that the viscoelastic properties of the lignin blend were similar to those of neat 70/100 bitumen, with overlapping complex shear moduli and phase angles. U10-lignin replaced 25wt.% bitumen without compromising its viscoelastic properties, indicating good lignin and bitumen compatibility. The lignin-bitumen blend did not exhibit the characteristic plateau in the phase angle response at a low frequency or high-temperature region, as seen in the PmB response (Fig. 2b). This plateau is a result of elasticity in PmB, resulting from the polymer network in the bitumen. This suggests that the blend did not have a similar network structure as PmB, and as a result, it exhibited rheological properties similar to 70/100 bitumen.

MSCR: The MSCR test evaluated the high-temperature performance of the lignin-bitumen blend, comparing it to reference PmB and penetration bitumen and the results are summarized in Table 1. The creep compliance parameter "J_{nr}" quantifies a binder's non-recoverable portion due to creep-recovery cycles, indicating its contribution to permanent deformation at the asphalt mixture level. Additionally, the recovery value R reflects the delayed elastic response of the polymer in the binder. The parameters show that 70/100 bitumen exhibited a purely viscous response with no recovery at both stress levels. Whereas, PmB demonstrated a higher potential for recovery compared to 70/100 bitumen, displaying delayed elastic response and substantial recovery. The lignin-bitumen blend exhibited limited recovery at the low stress level (0.1 kPa) but a purely viscous response at 3.2 kPa. The J_{nr} values of PmB were lower than the other binders, indicating better resistance to permanent deformation due to its polymer network. Higher stress levels resulted in higher permanent deformation. The J_{nr} values in lignin-bitumen blend

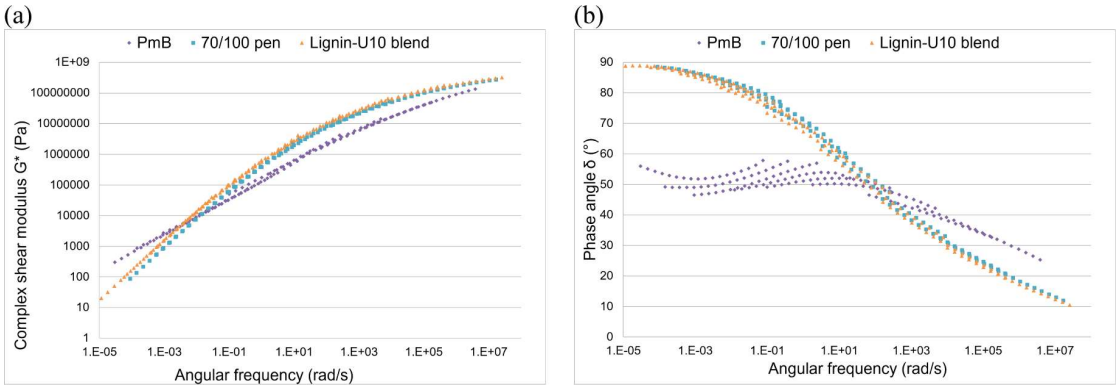

Fig. 2. Mastercurves of original PEN 70/100 bitumen, reference PmB and 25wt.% U10 lignin-bitumen blend as a function of angular frequency. (a) Complex shear modulus and (b) Phase angle.

were lower than those of 70/100 bitumen at both stress levels, demonstrating that the blend improved rutting resistance to some extent.

Table 1. MSCR test results of the binders.

Binder	Average recovery of the 10 cycles R (%)		Difference in recovery R_{diff} (%)	Average non-recoverable part of cycles J_{nr} (1/kPa)		Difference in non-recoverable compliance $J_{nr\text{-}diff}$ (%)
	@0.1 kPa	@3.2 kPa		@0.1kPa	@3.2 kPa	
Pen 70/100	0	0	-	7.0	7.72	10.1
PmB	87.2	86.5	0.73	0.1	0.13	14.2
Lignin-bitumen blend	3.1	0	-	5.3	6.2	16.3

3.2 Asphalt Evaluation

Indirect Tensile Test (ITT) were performed at 15 °C for both fresh and aged specimens of SMA-14 with reference PmB (SMA14-PmB) and lignin-bitumen blend (SMA14-70/100 bio). Indirect tensile strength ITS and ratio ITSR values are measured and the data are presented in Fig. 3.

With ageing, ITS values increases which is due to the hardening of asphalt as a result of the stiffening of bitumen. SMA14-PmB had a comparable wet and dry strength values. Whereas, SMA14-70/100 bio shows higher strength compared to SMA14-PmB and optimum ITSR values.

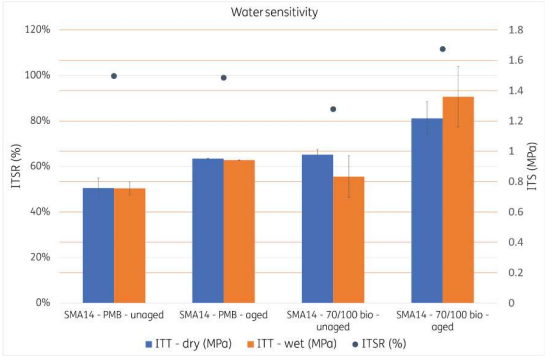

Fig. 3. ITS and ITSR values at dry and wet conditioning.

4 Conclusions

The thermal analysis revealed a melting point of 142 °C for U10 lignin, guiding the blending process to ensure effective lignin dissolution in bitumen. Using both high and low shear mixing processes at 170 °C resulted in a homogeneous blend with minimal lignin sedimentation. The complex shear modulus and phase angle mastercurves indicated that the lignin-bitumen blend possesses viscoelastic properties comparable to penetration 70/100 bitumen showing the potential to replace penetration bitumen without compromising rheological properties. The creep-recovery response aligned with penetration 70/100 bitumen, indicating favorable properties for rutting resistance. It showed some recovery at low stress level, while a fully viscous response at high stress level.

Evaluating the ITS values of SMA-14 mixes, the blend demonstrated optimum ITS and ITSR values, showcasing its potential for optimal asphalt mixture properties. This research showcases lignin as a promising and sustainable alternative to bitumen, with good compatibility, comparable viscoelastic behavior, and enhanced resistance to permanent deformation.

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