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Original Research Paper

Chemical and rheological analysis of unaged and aged bio-extended binders containing lignin

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HIGHLIGHTS

- Chemical interaction between bitumen and lignin were detected by FTIR and SARA analysis.
- Rheological properties at low, medium and high temperatures were investigated.
- Evolution of the temperature range of Performance Grade is outlined.
- The role of lignin against oxidations effects were evaluated.

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ABSTRACT

The use of alternative renewable sources to totally or partially replace bitumen is one of the most current challenges in the road pavement sector. The growing energy cost and environmental concerns lead to the necessity to find alternative solutions, by supporting at the same time sustainability and circular economy principles. Within this framework, this paper presents the application of a powder lignin, a natural bio-polymer deriving from by-products of wood pulp and paper industry, to replace part of bitumen. The bituminous blend consisting in 70% of bitumen and 30% of powder lignin (by weight) was made in laboratory through the use of a high shear stirring mixer, and a reference plain bitumen characterized by a similar consistency (i.e., same penetration grade) was used as comparison. Then, an extensive investigation on chemical and rheological properties of the bio-binder is presented. Fourier transform infrared spectroscopy (FTIR), saturates, aromatics, resins and asphaltenes (SARA), bending beam rheometer (BBR), frequency sweep tests and multiple stress creep recovery tests (MSCR) with a dynamic shear rheometer (DSR) were performed. Moreover, unaged, short- and long-term aging conditions were considered. Results indicated that powder lignin dominates the rheological behavior of the bio-binder and, from chemical analysis, it seems that it partially acts as a filler and partially as a binder. This would result in improved performances at both low and high temperatures, leading to a wider temperature range of performance grade (PG). Moreover, despite a stiffening effect is recognized, lignin also offers an antioxidant potentiality, reducing the aging susceptibility of the investigated bio-binder.

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1. Introduction

The continuously increasing cost of raw material and the current environmental issues also affected road construction field. Bitumen is an essential constituent of road pavements, but its supply may become difficult because of its ever-decreasing availability and consequently growing cost (Zahedi et al., 2020). Researchers also have been looking for promising renewable alternatives based on sustainability and circular economy principles (Fakhri and Norouzi, 2022; Gao et al., 2020; You et al., 2022). Nowadays, a sustainable development means to give a balance between economic, social and environmental, reducing GHG emissions as well as saving energy and reusing end of life materials (Kuhlman and Farrington, 2010; Yaro et al., 2022; Zhang et al., 2022). In this context, the world of biogenic materials as potential alternative sources for the production of binder for road application could comply with all the sustainability principles. However, giving up good performance in exchange for sustainability in paving our roads is unacceptable (Arafat et al., 2019). On the contrary, a valid technical solution forces to investigate the chemical, rheological and mechanical behaviors of bio-extended binders which are partially replaced with renewable sources, in order to guarantee at least comparable performances to those of the common bituminous binders available on the market so far.

Nowadays, the major feedstocks used for bio-binders are plants residues, such as agriculture crops or vegetable biomass (soy, palm, linseed, rapeseed, sunflowers, olives and more) (Gaudenzi et al., 2021; Ingrassia et al., 2019; Su et al., 2018), animal manure (Fini et al., 2011) or algae (Chen et al., 2021). Among the several solutions identified, lignin seems to be very promising for several reasons. First, deriving from many plant species, it is available in large volumes and represents the second most abundant bio-polymer on the planet (Boerjan et al., 2003; Pérez et al., 2019). Moreover, it is generated from production process for wood pulp and paper industry and is an unwanted waste during the production chain. Given its hydrocarbon nature, lignin is potentially compatible with bitumen, showing a certain similarity to asphaltenes, which represent the heaviest and most polar part of bitumen (Min et al., 2013).

As a result of the mentioned chemical affinity, lignin can be treated as a bitumen modifier/replacement, which can significantly change the high- and low-temperature rheological behavior, fatigue response, aging susceptibility as well as durability of base bitumen (Yao et al., 2022). A conscious use of lignin in the production of bituminous binders could not only allow the reduction of environmental impact and reduction of waste to be disposed of in landfills, but would also reduce the dependence on petroleum, possibly with consequent economic saving.

Past studies on using lignin in asphalt binders have shown that from a chemical point of view, the molecular structure of lignin contains benzene ring, phenolic, hydroxide, aldehyde and methoxy groups, which have been proved to be more resistant against oxidation (Batista et al., 2018; Ren et al., 2021; Xu et al., 2017). Moreover, the same antioxidant property is also reflected in rheological tests (Gao et al., 2020; Ren et al., 2021; Pérez et al., 2020). According to the studies of Wu et al. (2021), when lignin is incorporated in bitumen it rises explicit chemical activities associated to the formation of additional functional groups, but divergent results were reported by Zhang et al. (2019). On the contrary, unequivocal results were found on the physical properties of lignin-modified binders, which showed increased values of stiffness and viscosity, and a reduced temperature susceptibility as compared to the conventional bitumen (Batista et al., 2018; Gao et al., 2020; Xu et al., 2017). The increase in stiffness leads to an improvement of high temperature properties, and the higher the lignin content, the higher the resistance to rutting (Fakhri and Norouzi, 2022; Norgbey et al., 2020; Pérez et al., 2020; Xu et al., 2017, 2021). The addition of lignin also produces a bio-binder having a more elastic behavior, but fatigue performances appeared overall penalized or at most comparable with those of conventional bitumens (Gao et al., 2020; Ren et al., 2021; Xu et al., 2021). At low temperature lignin-extended binder seem to be characterized by a lower ability to quickly absorb thermal stresses resulting in higher cracking potential (Ren et al., 2021; Xu et al., 2017), even though in this regard other studies did not find significant changes (Norgbey et al., 2020).

The discording results found in literature can be attributable in most of cases to the strictly dependence on the combination of base materials, that is type of bitumen, the raw material from which lignin derives, the production methods adopted to produce it, the lignin dosage and many other variables.

The extreme variability which characterizes each kind of lignin also can have an impact on the rheological behavior of bio-binder produced with lignin. Thus, in order to make the use of lignin in bituminous binders truly feasible, more in depth investigations are needed.

This paper focuses on the evaluation of the potentialities of a lignin-extended binder with respect to a conventional 50/70 pen grade bitumen commonly used in Italy for road applications. The blend was produced in laboratory by replacing a conventional bitumen with 30% of powder lignin, so obtaining a bio-binder characterized by the same consistency (i.e., penetration value) of the reference plain bitumen. Precisely, this work is supposed to be innovative because it contributes

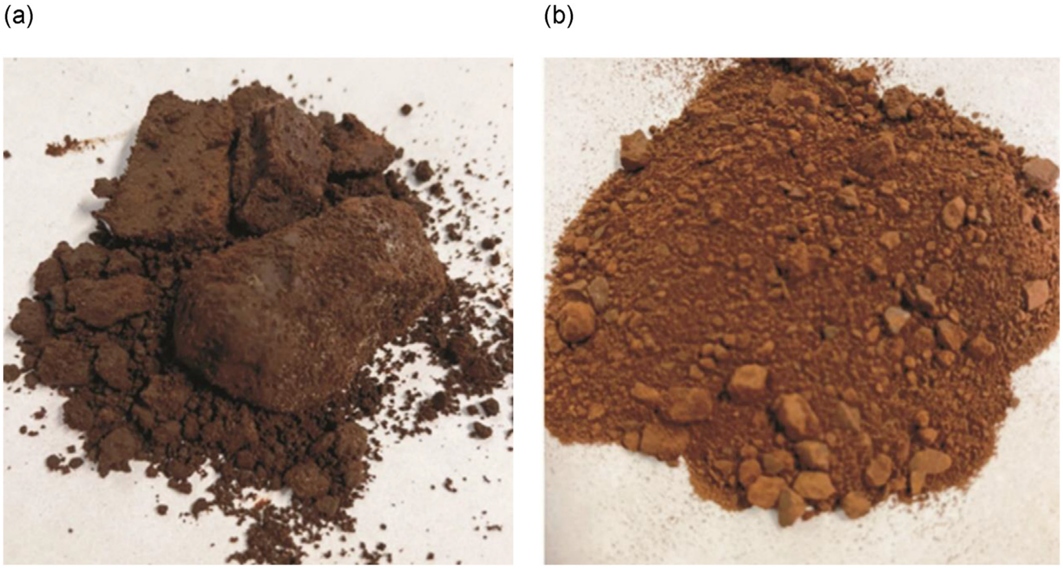

Fig. 1 – Lignin “S” in different moisture conditions. (a) Wet. (b) Dry.

results and practical experiences to world place challenges currently in place by addressing the effectiveness of lignin higher powder lignin contents than those currently available in the literature studies, with the aim to save petrol-derived source. Indeed, the idea embraces the main goals of the modern society, consisting in generate improving environmental and economic standards through the efficient and conscious use of renewable sources. The experimental investigation considers the study of the chemical components and rheological behavior of both binders. Specifically, the chemical analysis aims at clarifying the function of the lignin replacement, i.e., chemical bounds or only a physical interaction with bitumen. Moreover, a rheological analysis allows to provide a performance characterization, which is useful to predict the material behavior at different temperatures and loading conditions and considering the aging effects as well. Additionally, the performance grade (PG) of the bituminous binders was evaluated. In the first part, the chemical components of lignin, bitumen and bio-binder were characterized by Fourier transform infrared spectroscopy (FTIR) and saturates, aromatics, resins and asphaltenes (SARA) analysis. Then, rheological tests consisting in bending beam rheometer (BBR), multiple stress creep recovery (MSCR) and frequency sweep were performed.

2. Materials

In this experimental investigation, a conventional 50/70 penetration grade bitumen commonly used in Italy for road applications (ANAS, 2009) was selected as reference material. Whereas, a powder lignin of Swedish origins named as “S” deriving from some kind of thermochemical and/or enzymatic treatment of wood was used to replace 30% by weight of a 70/100 plain bitumen to obtain a bio-binder, coded as 70/100_S30. Before its use, lignin “S” was finely crumbled and dried at 105 °C to eliminate the moisture content, as showed in Fig. 1, while Table 1 sums up its characteristics. The main aim was to obtain a lignin-based binder characterized by physical properties comparable to that of the reference bitumen. For a better understanding of the chemical effects, the original 70/100 plain bitumen used to produce the lignin-extended binder was also investigated. The main properties of the binders selected are summarized in Table 2. These binders have been investigated previously with respect to their performance in asphalt mixtures (Gaudenzi et al., 2022a, 2023). The binder containing lignin was prepared in laboratory through a mechanical mixing by means of a high shear stirring mixer, setting the blending speed at 5000 rpm, and running the mixing at temperature of 150 °C for about 30 min to guarantee a homogenous dispersion of lignin within bitumen. Further details about the binder preparation can be found elsewhere (Gaudenzi et al., 2022a, b). Bio-binder and both plain bitumens were subjected to short- and long-term aging through the rolling thin film oven test (RTFOT) and pressure aging vessel (PAV), respectively. RTFOT procedure subjects the binders to a temperature of 163 °C for 75 min following the EN 12607-1 (CEN, 2007), whereas PAV procedure is performed on binders already aged with RTFOT by applying a temperature of 100 °C and a pressure of 2.1 MPa for 20 h in accordance with EN 14769 (CEN, 2013). In total, eight types of binders were tested.

Table 1 – Characteristics of the lignin “S”.

Origin	Lignin from thermochemical and enzymatic treatment of wood
Physical property	Solid
Moisture content (%)	50/60
Freezing point (°C)	−5 to 0
Color	Dark
Brown pH	4–6
Density (g/cm^3)	1.35

Table 2 – Main conventional properties of the investigated binders.

Characteristic	Value[a]	50/70	70/100	70/100_S30
Penetration at 25 °C (0.1 mm) (EN 1426)[b]	50.0/70.0	52.0	88.0	58.8
Softening point (°C) (EN 1427)[c]	45.0–65.0	51.5	46.5	50.8
Dynamic viscosity at 160 °C (mPa·s) (EN 13302)[d]	30–100	116	80	423
Increase in softening point (after RTFOT) (°C)	≤9.0	4.6	5.1	7.0
Retained penetration (%)	≥40	85	64	58

Note:
[a] Italian technical requirement (CEN, 2013).
[b] CEN, 2015a.
[c] CEN, 2015b.
[d] CEN, 2018.

3. Testing program and methods

Preliminarily, chemical analysis consisting of FTIR were carried out on basic materials (pure lignin, plain bitumens and bio-binder) to evaluate their composition, whereas SARA analysis was carried out on unaged and PAV aged binders to assess oxidation effects. Later, the rheological properties at medium and high temperatures of two binders in all aging conditions were evaluated through frequency sweep tests and MSCR tests, respectively, by using DSR. Finally, the low temperature behavior of unaged and PAV aged binders was assessed by means of BBR analysis. Table 3 summarizes the testing program.

3.1. *Chemical analysis*

The chemical composition of the pure lignin, lignin-extended binder and two plain bitumens in unaged condition were evaluated through FTIR, which allows to identify the various functional groups (Batista et al., 2018; Zhang et al., 2019). The samples were prepared by laying a very small amount of material on a thin diamond crystal plate of the spectrometer. The analysis was run in reflection mode with the attenuated total reflectance (ATR), by recording the absorbance between wavenumbers 500 and 4000 cm^{-1} with a resolution of 4 cm^{-1}. The obtained spectrum gives information about the functional groups corresponding to specific wavenumbers, in particular the carbonyl index ($I_{C=O}$) and the sulphoxide index ($I_{S=O}$) were considered as parameters for the analysis. Two repetitions for each material were performed at ambient temperature and the average was considered for the analysis.

The determination of SARA was performed by thin layer chromatography with flame ionization detection (TLC-FID, commonly named iatroscan) following the IP 469 standard (Energy Institute, 2001). The samples were prepared by dissolving about 0.1 g of binder in dichloromethane to reach a 2% ± 0.2% solution. Then, a small amount of solution (1 μm) was carefully applied on the pre-activated

Table 3 – Sum up of the testing program.

Test	Lignin	S50/70_unaged	70/100_unaged	70/100_S30_unaged	50/70_RTFOT	70/100_S30_RTFOT	50/70_PAV	70/100_PAV	70/100_S30_PAV
FTIR	X	X	X	X	–	–	X	X	X
SARA	–	X	X	X	–	–	X	X	X
Frequency sweep	–	X	–	X	X	X	X	–	X
MSCR	–	X	–	X	X	X	X	–	X
BBR	–	X	–	X	–	–	X	–	X

Note: X means test performed, – means test not performed.

quartz rods filled with active silica. An entire frame with 10 rods were analyzed for every binder and aging configuration. Saturates, aromatics and resins were separated by using heptane, toluene-heptane (80:20) and dichloromethane-methanol (95:5), respectively, while asphaltenes remained not eluted. An integration process of the obtained chromatograms allowed the determination of the percentage amount (by weight) of the four components. The lignin solubility was determined following the EN 12592 specification (CEN, 2014) by dissolving a small sample of lignin-extended bio-binder in 100 mL of dichloromethane. The solution was filtered through a filtering mat in a filtering crucible and the material retained on the filter was then dried for about 30 min in an oven at 90 °C and weighed, and subsequently left in a dehumidifier for about 20 min. At the end, the bitumen content as a percentage of the weight of the original binder was calculated and consequently the lignin solubility amount by the following equation.

$$x_S = 100 - \frac{m_A}{m_B} \times 100 \tag{1}$$

where x_S is the percentage of soluble matter, in percent, m_A is the mass of insoluble material and m_B is the mass of the dry sample. Both unaged and long-term aged bio-binders were investigated.

3.2. Rheological analysis

3.2.1. Bending beam rheometer test

BBR test was performed to access low temperature cracking propensity of the investigated binders according to AASHTO T313 (AASHTO, 2019).

The central deflection of the beam and the applied load were continuously monitored to determine the creep stiffness $S(t)$ and creep rate parameter representative of the stress relaxation capacity $m(t)$ of the binder as functions of the loading time and temperature.

Specifically, only the values of creep stiffness $S(60)$ measured at 60 s and the corresponding $m(60)$ value were considered for the result analysis for all materials and testing conditions.

In this analysis, BBR tests were carried out on unaged samples at testing temperatures of −6 °C and −12 °C and on long-term aged samples at the testing temperatures of −12 °C and −18 °C. Three replicates for each binder and testing conditions were tested and the average values were considered for the analysis.

The BBR results also allow the calculation of the $T_{\min}$ to define the performance grade (PG), which is described by the AASHTO MP1 specification (AASHTO, 1998) with the following Eq. (2).

$$T_{\min} = T_{\text{test}} - 10 \tag{2}$$

where T_{test} is the lowest test temperature able to satisfy the creep stiffness and creep rate requirements ($S(60) < 300$ MPa, $m(60) > 0.300$) at the same time. The test refers to the long-term aged binders.

Moreover, the BBR results are used to calculate ΔT_c, which is a binder parameter proposed to assess aging-associated surface cracking. Specifically, ΔT_c is calculated as the mathematical difference between the two PG critical temperatures, expressed in degrees (°C) as follows.

$$\Delta T_c = T_{c,S} - T_{c,m} \tag{3}$$

where $T_{c,S}$ is the critical temperature for creep stiffness ($S(60)$) and $T_{c,m}$ is the critical temperature for creep rate ($m(60)$) at the limiting values of 300 MPa and 0.3, respectively.

In an asphalt binder, the sign of ΔT_c, either positive or negative, indicates whether the performance grade of the binder is governed by its creep stiffness ($+\Delta T_c$) or creep rate ($-\Delta T_c$). The absolute magnitude of ΔT_c indicates the degree to which the binder is governed by either creep stiffness or creep rate. In general, a binder of more positive ΔT_c is more durable in terms of resistance against the aging-associated surface cracking, meaning that a better stress relaxation capability is ensured regardless of the higher aged stiffness.

3.2.2. Frequency-sweep test

Dynamic shear rheometer (DSR) device was used to evaluate the time and temperature dependence of the investigated binders by performing frequency sweep test according to EN 14770 (CEN, 2012).

Tests were performed in the temperature range from 4 °C to 100 °C with a step of 6 °C, by applying a loading frequency range between 0.159 and 15.9 Hz (corresponding to 0.1–100 rad/s) at each testing temperature. A plate-plate configuration was adopted, selecting 8 mm diameter and 2 mm gap for testing from 4 °C to 34 °C, and 25 mm diameter and 1 mm gap for test from 34 °C to 100 °C. A shear strain equal to 0.1% was selected in all tests to ensure the rheological measurements within the linear viscoelastic domain (LVE). Tests were performed on unaged, short- and long-term aged samples, and at least two repetitions were tested for each binder and aging condition.

Data from frequency sweep test allowed the evaluation of the complex modulus G^* on a wide range of temperature and loading frequency. In particular, the G^* master curve was obtained by applying shift factors based on the Williams-Landel-Ferry (WLF) law (Williams et al., 1955).

3.2.3. Multiple stress creep recovery (MSCR)

The MSCR analysis were carried out to evaluate the rutting behavior of the investigated binders. The MSCR tests were performed in accordance with AASHTO T350 (AASHTO, 2018a) using a dynamic shear rheometer in a 25 mm parallel plate configuration at 40 °C and 60 °C on binders in all aging conditions. Moreover, further tests were run on the short-term aged binders at 64 °C, 70 °C and 76 °C with the aim to verify the T_{high} for performance grade (PG) evaluation. The tests consisted of the application, in succession, of two constant stress creep phases (0.1 and 3.2 kPa) of 1 s duration followed by a zero-stress recovery of 9 s duration, for 20 and 10 cycles, respectively, at each testing temperature. For each stress level, the last 10 creep-recovery cycles were considered for the data analysis. Specifically, the non-recoverable creep compliance J_{nr} (ratio between the accumulated strain and the applied stress) and the percent of recovery parameter R (%) (related to the recovered strain) were calculated as good indicators of the high temperature behavior of binders in terms of the resistance to the accumulation of permanent deformations and of the ability to recover deformation, respectively.

At least, two repetitions for each binder, temperature and aging condition were performed.

4. Results and analysis

4.1. *FTIR*

FTIR was used to study the chemical composition and functional groups of lignin, reference bitumen (50/70_unaged) and lignin-extended binder (70/100_S30_unaged). Fig. 2(a) shows the fingerprint region between 500 and 4000 cm^{-1} wavenumbers, from which it can be observed that the shape of the spectra of both binders is very similar in the entire area considered. A small difference is found in correspondence of the carbonyl group (C═O) at 1700 cm^{-1} and aromatic (C═C) at 1600 cm^{-1}. In correspondence of the two wavenumbers, small peaks are recognized in the lignin-extended binder spectra, while a linear shape is visible in the reference 50/70 bitumen spectra. In similar way, also the range between 500 and about 1300 cm^{-1} shows some differences between the two binders. In particular, the same peaks are registered except for the range between about 1000 and 1300 cm^{-1}, mainly corresponding to C─H, C─O and O─H bonds deformations and vibrations. To understand the nature of the recognized variations, the 70/100 plain bitumen was also investigated. Since bio-binder containing lignin shows similar trend compared with the original 70/100 pen. grade, the small differences may be attributable to a possible difference in bitumen composition, and an only physical interaction can be recognized between bitumen and lignin. In fact, the absence of new peaks leads to the assumption that no

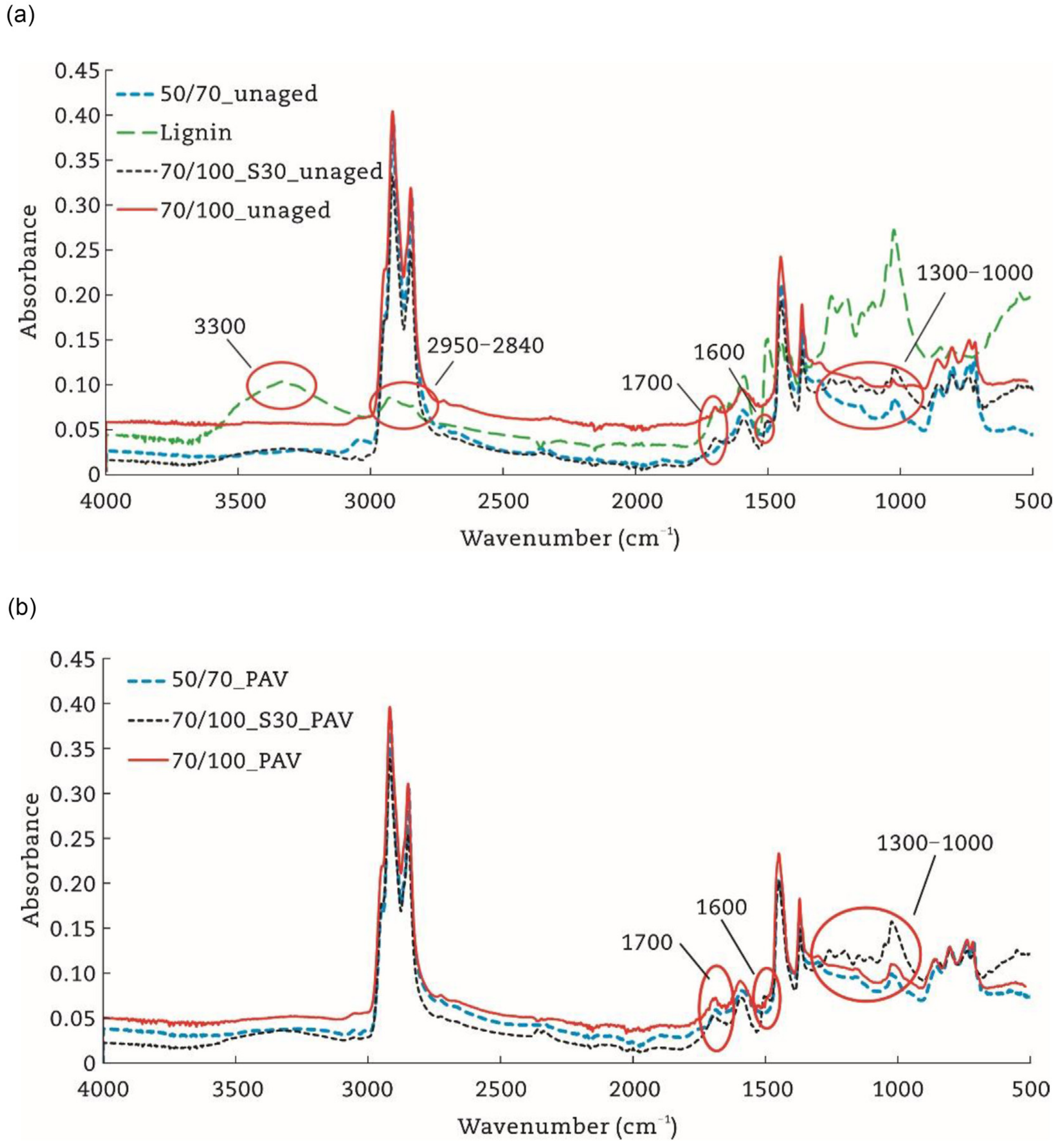

Fig. 2 – FTIR spectra. (a) Unaged. (b) PAV aged conditions.

chemical interactions occur. As far as the only lignin peaks are considered, given its hydrocarbon nature, a similar spectrum to these of binders is recognized, as in correspondence of C=O (1700 cm^{-1}), S=O (1030 cm^{-1}), CH_2 (1460 cm^{-1}) and CH_3 (1380 cm^{-1}), even if a difference in correspondence of N–H bond (3300 cm^{-1}), the lack of a defined peak in correspondence of the –C–H stretch (2950–2840 cm^{-1}) and an higher intensity at C–I and C–F bonds, corresponding to 500 cm^{-1} and 1030–1250 cm^{-1} wavenumbers were found. To study the influence of aging on the interaction mechanism, the spectra of the same binders were recorded also after PAV aging conditions. From Fig. 2(b) it was observed that no visible changes to peaks were found for the lignin-based binder, synonym of no substantial susceptibility against aging effects, while little differences were identified for 50/70_PAV and 70/100_PAV at 1700 and 1000–1300 cm^{-1}, corresponding to C=O and S=O bonds respectively.

4.2. *SARA fractions*

Typical chromatograms obtained in unaged and long-term aged conditions for the lignin-extended binder and the 50/70 reference bitumen are plotted in Fig. 3 as an example. For a more comprehensive understanding of the lignin interaction with bitumen, also the plain 70/100 has been analyzed. Results show, as expected, the same four peaks corresponding to saturates, aromatics, resins and asphaltenes are recognizable in all cases. A less pronounced change in peaks of bio-binder was observed after long-term aging, contrarily to the more consistent reduction of aromatics and increase of resins of both plain bitumen (reference 50/70 and original 70/100). These findings would support the hypothesis of the antioxidant potential offered by lignin (Luo et al., 2019; Pan, 2012; Ren et al., 2021). However, regardless the presence or absence of lignin, asphaltene peak of all the investigated binders changed its shape from a distinct peak in unaged conditions to a blurred area in PAV conditions with worse separation between resins and asphaltenes, in line with previous findings in literature (Ingrassia et al., 2019; Stangl et al., 2006).

Table 4 quantify the percentages of the four fractions (saturates, aromatics, resins, and asphaltenes), which was claimed to play a key role in forming a sol-gel structure in asphalt binder (Zheng et al., 2021), and affect both elastic and

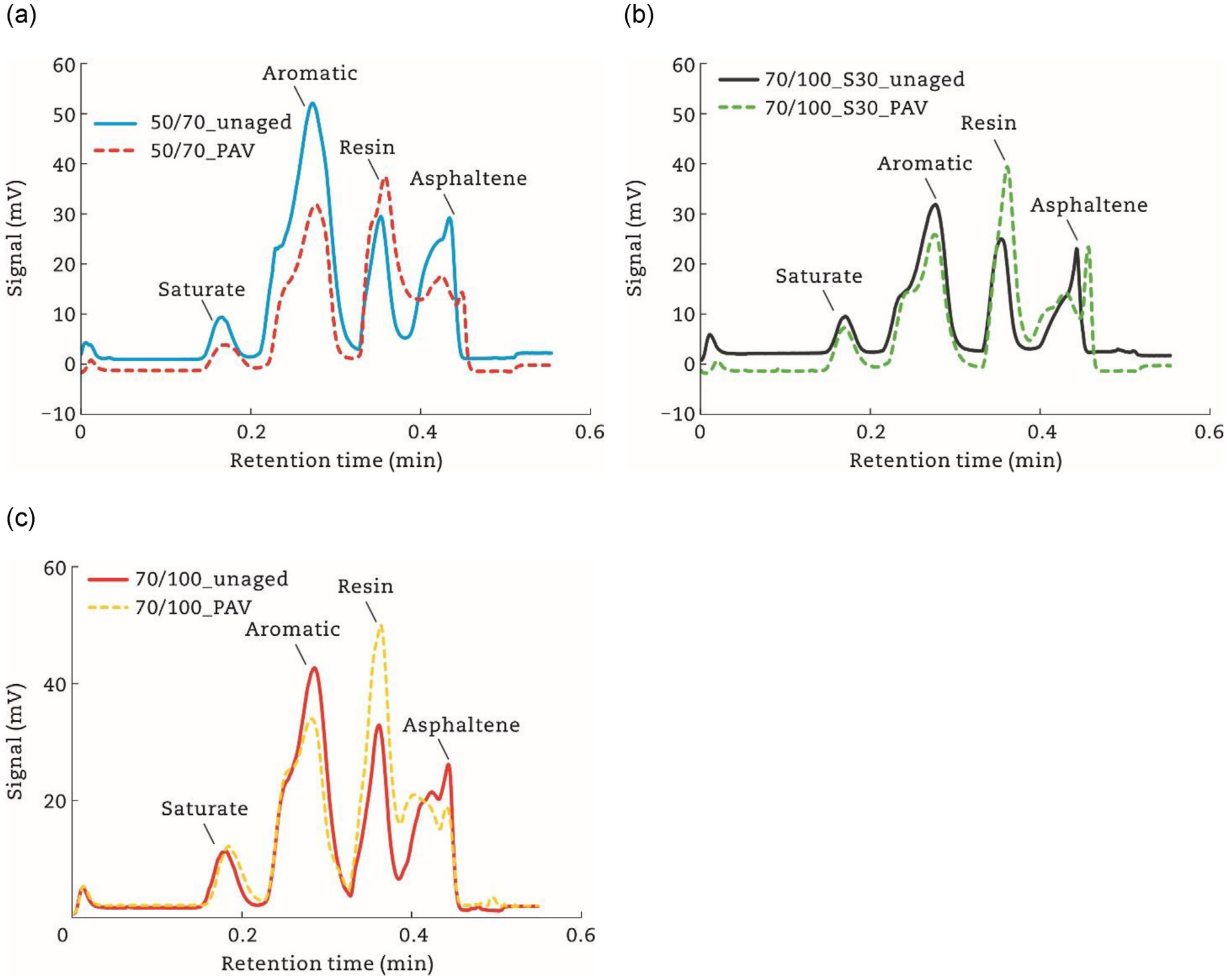

Fig. 3 – Chromatograms comparison between unaged and long-term aged conditions. (a) Reference 50/70. (b) Lignin-extended binder. (c) Original 70/100 binders.

Table 4 – SARA analysis: main components and colloidal index of the investigated binders in unaged and long-term aged conditions.

Material	Saturate (%)	Aromatic (%)	Resin (%)	Asphaltene (%)	Resin + asphaltene (%)	Colloidal index
70/100_S30_unaged	6.5	50.1	21.6	21.9	43.5	0.39
50/70_unaged	4.6	58.0	15.4	21.9	37.3	0.36
70/100_unaged	6.5	46.9	23.4	23.2	46.6	0.42
70/100_S30_PAV	4.5	40.9	29.4	25.2	54.6	0.45
50/70_PAV	6.2	38.1	29.4	26.4	55.8	0.48
70/100_PAV	6.2	35.2	34.2	24.4	58.6	0.44

permanent deformation in the service environment (Loeber et al., 1998). Outcomes refer to the average of at least 2 repetitions for each binder, each one consisting in the analysis of 5–10 rods. From the experimental results, no differences emerged in saturates between the plain 70/100 bitumen and the bio-binder considered in unaged conditions, while slightly lower values emerged for the reference one. On the contrary, a slightly reduction and increase in saturates are found in aged conditions for the bio-binder and the reference bitumen, respectively, while stable values were obtained for the 70/100. Lignin introduction causes a lower reduction of aromatics and increase of resins compared to the other binders, while a higher increase of asphaltenes is recognized compared to the plain 70/100, but lower than the 50/70 reference bitumen. Since an intermediate peak (fraction) between resins and asphaltenes resulted after aging, the sum of resins and asphaltenes is also reported in the table, which demonstrate the lower aging susceptibility of 70/100_S30 especially if compared to the reference 50/70. Moreover, the colloidal index (CI) was also calculated as follows in Eq. (4).

$$\mathrm{CI} = \frac{\text{asphaltene} + \text{saturate}}{\text{resin} + \text{aromatic}} \tag{4}$$

In the colloidal model, aromatics and resins represent the dispersing constituents while saturates and asphaltenes are the flocculated constituents. Thus, a lower colloidal index means that the asphaltenes are more peptized by the resins in the oil-based medium (Loeber et al., 1998). However, after aging an increase of asphaltenes is expected, and given the low variability of saturates, the aging effect results in an increase of CI. Indeed, aged conditions experienced higher CI values regardless the investigated binder, however lowest differences appear between unaged and aged conditions for the original 70/100, followed by the bio-binder and finally by the reference 50/70. The lower variability experienced by the 70/100 plain bitumen is given by the almost unchanged values of the flocculated constituents, and the constant sum of the dispersed constituent despite the decrease/increase of the aromatics/resins respectively.

Moreover, lignin contents for the bio-binder in unaged and long-term aged conditions, were determined by solubility test. As expected, the lignin content for the bio-binder before aging was approximately 30% consistently with the designed amount, whereas a significant reduction was found after PAV aging resulting in 22.8%. This experimental evidence could be explained as that the small part of lignin (about 7%) physically and/or chemically interacts with bitumen during the aging test, thus becoming part of asphaltenes. This result could be corroborated by the higher increase of the bio-binder asphaltenes component after aging (≈15%) compared with the asphaltenes variation of plain 70/100.

4.3. *BBR test results*

When exposed to low temperatures asphalt pavements tend to contract, but they are countered by the frictional force offered by the underlying layers. As the temperatures decreases, contraction increases, resulting in an increase of thermal stress and a risk of cracking. Thus, the flexural-creep stiffness (S) and stress relaxation capacity (m) were computed from BBR data to assess the propensity to cracking at low temperature of the investigated binders (Pedro, 2016). Higher m-values indicate a higher binder flexibility, while lower creep stiffness causes fewer thermal stresses to the binder. Thus, a binder with higher m-value and lower creep stiffness would be more desirable to reduce low-temperature cracking.

Fig. 4 shows the S value measured at 60 s (S(60)) and the corresponding m value (m(60)) for the lignin-extended binder and the reference plain bitumen in unaged and long-term aged conditions at each testing temperature. Specifically, temperatures of −12 °C and −18 °C were selected for unaged binders, while −6 °C and −12 °C were selected for aged binders. In the plots, the limiting values of 300 MPa for creep stiffness and 0.300 for creep rate according to the performance grade classification system for asphalt binder (AASHTO, 2016) were also reported.

As expected, the creep stiffness increases with decreasing temperature and increases with the aging, whereas the m value changes oppositely. In unaged conditions, the S(60) value of 70/100_S30 binder is significantly higher than 50/70 binder despite it was designed for having the same consistency (i.e., pen grade) of the reference bitumen (Gaudenzi et al., 2022b), so highlighting the hardening effect induced by the presence of lignin leading to a potential brittle behavior. Moreover, the presence of lignin leads to a slightly reduction of m(60) value. However, if aged conditions are considered, a reduced increase in S(60) but a significant increase in m(60) values were recorded for lignin-extended binder as compared to the reference binder at both −6 °C and −12 °C. From the analysis of plots it can be observed that the temperature of −12 °C is discriminating to satisfy the AASHTO requirements (i.e., $S(60) \leq 300$ MPa and $m(60) > 0.300$) for both investigated binders, however the lignin-extended binder in aged condition within this temperature seems to have a higher capability in absorbing thermal stress and return to the original state following

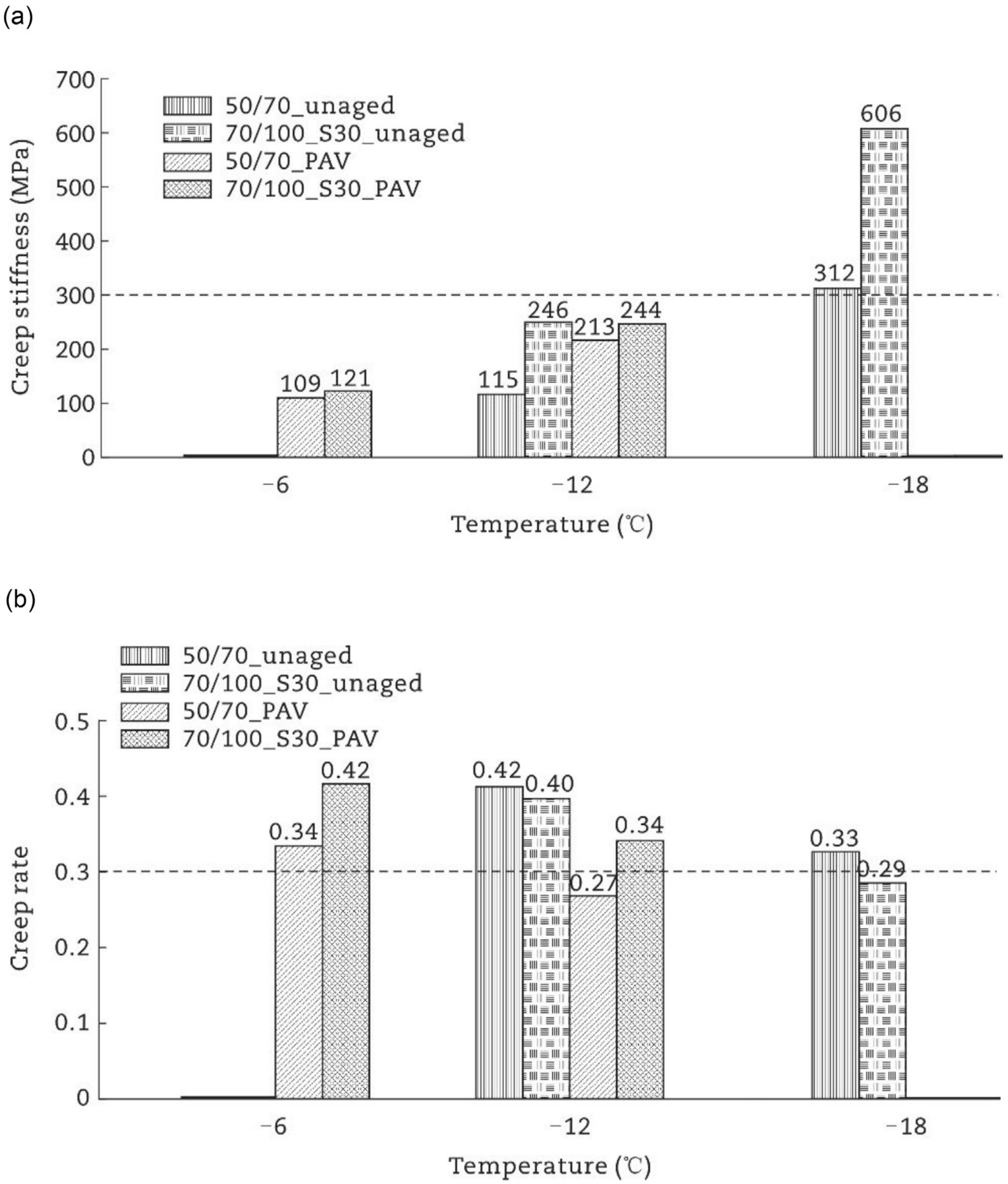

Fig. 4 – BBR test results in terms. (a) Creep stiffness. (b) Creep rate.

exposure to stress despite a comparable creep stiffness to reference binder.

From BBR results derives the calculation of the T_{min} to define the performance grade (PG).

The obtained findings show that a lower temperature is admissible for the lignin-extended binder ($T_{min} = -22$ °C), because of its better ability to combine creep stiffness and creep rate. On the contrary, the reduced stress relaxation capacity of the aged reference bitumen led to an increase of the T_{min} (−16 °C), despite the creep stiffness requirements at −12 °C were satisfied as well.

Moreover, Table 5 shows the critical temperatures, ΔT_c parameter and the T_{min} of PG of each binder. Results indicate that all binders are S-controlled except for 50/70_aged, which is *m*-controlled. In unaged conditions, the stiffness level seems to affect the low-temperature response of both binders, showing comparable value of ΔT_c. As expected, as the aging level increases, ΔT_c tends to negative values because of oxidative phenomenon (Asphalt Institute, 2019). Specifically, the 70/100_S30 binder preserves a proper compromise between stiffness and stress relaxation capability, contrarily to the reference bitumen which becomes predominantly *m*-controlled. Based on the overall analysis and since thermal cracking phenomenon affects asphalt pavements throughout its in-service life, that is as bituminous materials are in aged conditions, it can be highlighted that lignin-extended binder is less prone to the aging associated surface cracking as compared to the reference plain bitumen. These results suggest that the bio-based binder offers better resistance to aging, which could

Table 5 – Critical temperature and ΔT_c parameter for the investigated binders.

Parameter	50/70_unaged	70/100_S30_unaged	50/70_PAV	70/100_S30_PAV
Critical temperature ($S = 300$ MPa) (°C)	−17.6	−12.9	−17.0	−14.7
Critical temperature ($m = 0.300$) (°C)	−20.1	−17.3	−9.3	−15.3
ΔT_c (°C)	2.5	4.3	−7.7	0.5
T_{low} (°C)	–	–	−16	−22

be attributable to the presence of lignin, consistently with other previous studies (Wu et al., 2021; Xu et al., 2021).

4.4. *Frequency sweep results*

Frequency sweep tests allowed the evaluation of the complex modulus (G*) and phase angle for the investigated binders in each aging conditions. Fig. 5 reports the plots of the norm of the complex modulus |G*| as function of the phase angle δ (black diagram) for the lignin-extended binder and the reference plain bitumen at different aging levels. For both binders at each aging condition, a good alignment of data forming continuous curves can be observed, despite the occurrence of some dispersion at higher temperature for the unaged binders. This finding allows to consider both binders thermo-rheologically simple materials, thus proving the general validity of the time-temperature superposition principle (TTSP) (Airey, 2002).

The different trend of data in the black diagram between the two investigated materials, shows the different rheological behavior of bio-binder with respect to the reference bitumen, likely attributable to the presence of the lignin in the binder phase. Indeed, the reference bitumen shows a phase angle at high temperature approaching 90°, which is typical of plain bitumen, whereas the bio-binder shows a plateau region deriving from the decrease of the phase angle as the temperature increases, entailing the progressive predominancy of the elastic component of the complex modulus, similarly to what occurs for polymer modified bitumen. The plateau region is also indicative of lignin present as fillers in the binder. Moreover, as an organic material, bitumen is affected by aging which causes hardening effect resulting in an increase of stiffness and a decrease in phase angle. Thus, a shift of the black diagram curves towards lower phase angles appears as aging level increase (Airey, 2002), indicating a change in composition and structure in the asphalt binder, which seems to be less marked in the lignin-extended bio-binder than in the reference bitumen especially after long-term aging.

The master curves of complex modulus and phase angle of binder in different aging conditions are presented in Fig. 6(a) and (b), respectively. The construction of master curves was performed at the reference temperature of 34 °C and by applying a set of shift factors following the Williams-Landel-Ferry (WLF) model (Williams et al., 1955). The plotted master curves are the average curve of two repetitions.

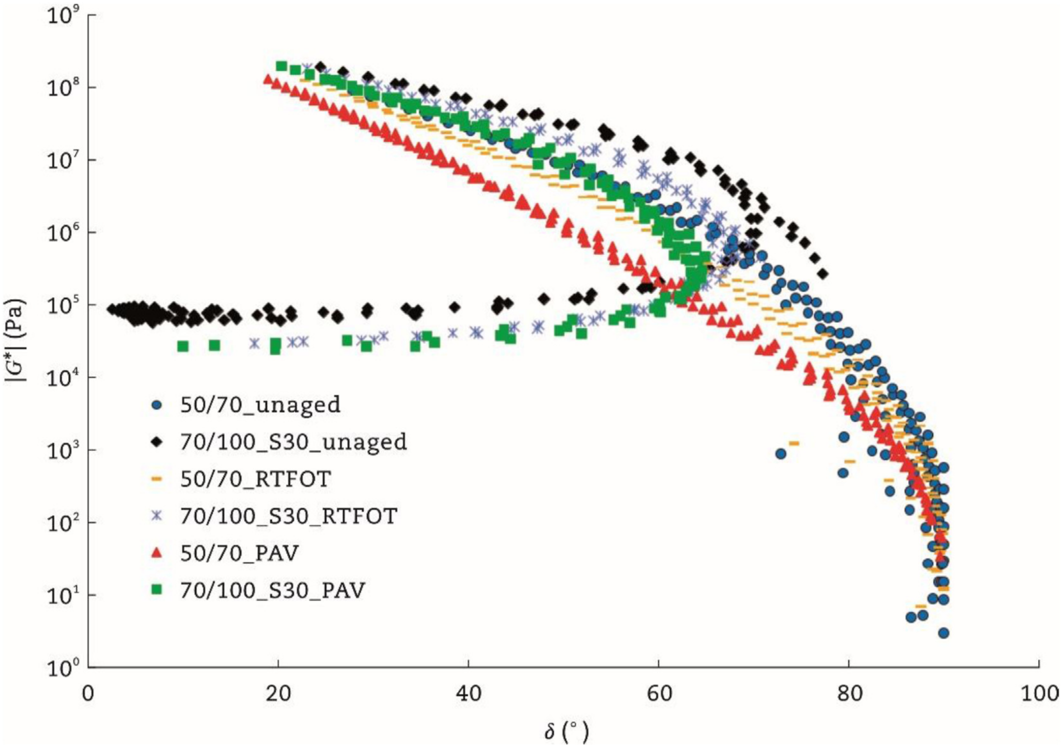

Fig. 5 – Black diagrams of investigated materials in unaged, short- and long-term aged conditions.

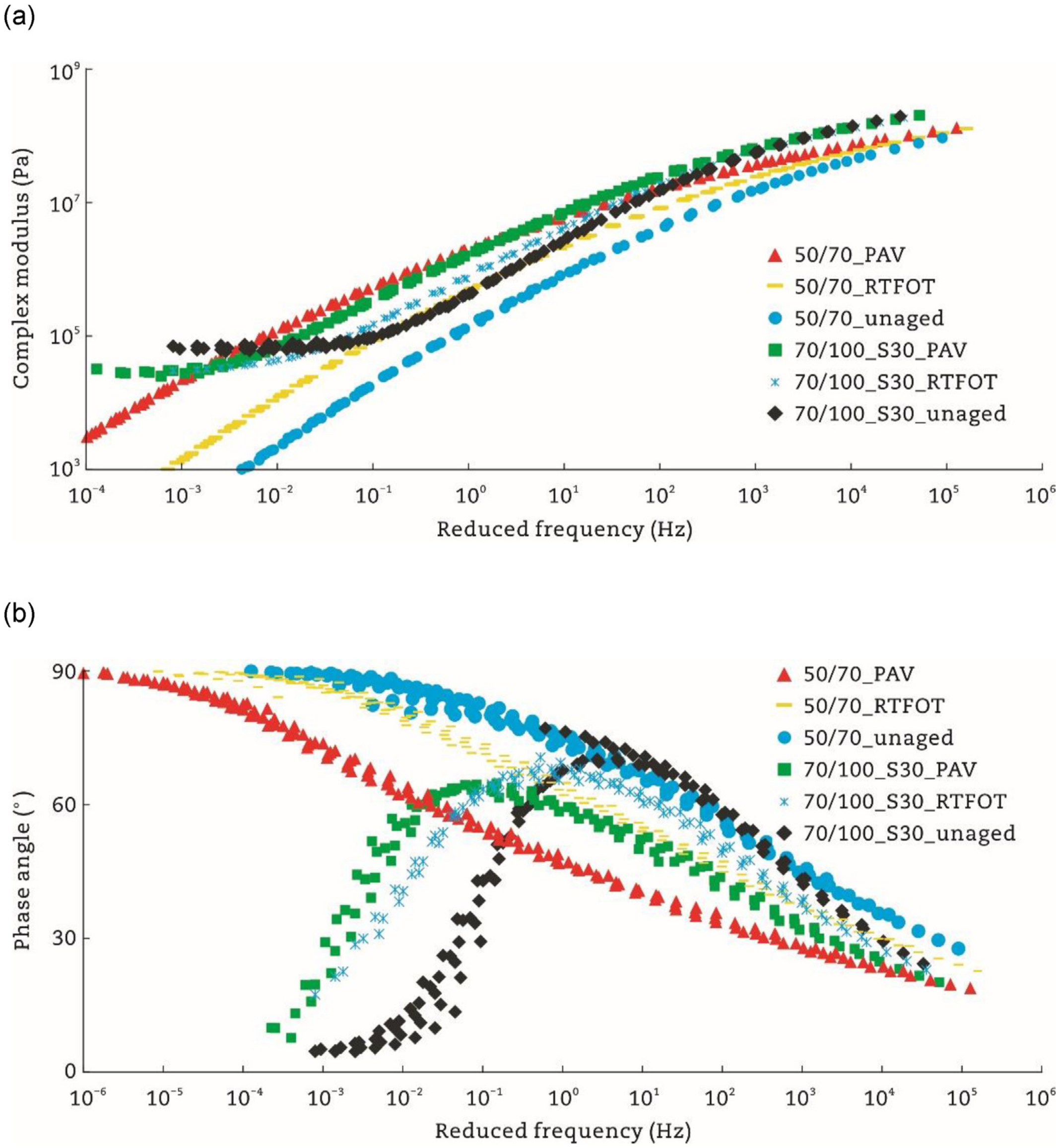

Fig. 6 – Master curves at 34 °C. (a) Complex modulus. (b) Phase angle.

As can be observed, in each aging condition the lignin-extended binder shows a higher stiffness modulus on the overall frequency and temperature domain investigated as compared to the reference bitumen, especially in unaged condition despite both binders was characterized by the same consistency (i.e., penetration value). This finding highlights the global stiffening effect due to the replacement of bitumen with a powdery source (i.e., solid lignin). Moreover, in the low frequency (i.e., high temperatures) range, bio-binder in all aging conditions show a rubbery plateau typical of composite materials (e.g., modified binders or mastics) (Mazzoni et al., 2017), so strengthening the hypothesis according to which lignin behave like a natural bio-polymer which, dispersed within the bituminous matrix, dominates the rheological behavior of the bio-binder. At the same time, also phase angle master curves present a completely different shape compared to the reference bitumen, in agreement with what was found by the Black space. From the analysis of the plots, the aging effects are evident on both binders. Indeed, a general increase of the complex modulus and a decrease of the phase angle with the level of aging can be observed. Specifically, in the high frequency (i.e., low-temperature) region it seems that binders are less sensitive to aging, showing master curves which tend to converge regardless of the aging level. On the contrary, the low frequency (i.e., high temperature) region of the lignin-based binder tend to converge to the same horizontal asymptote value regardless of the aging condition considered, in contrast with results showed by the conventional 50/70 plain bitumen, which shows distinct trends for each of the aging conditions analyzed. Aging effects are more evident throughout intermediate frequency (i.e., intermediate temperature) region for both investigated binders. However, from the plotted results it is clearly visible that the reference 50/70 pen grade bitumen is strongly affected by aging showing master curves significantly distinct, whereas reduced differences can be depicted for the lignin-extended binder, especially in the low frequency (high temperature) region where the three complex modulus master curves tend to overlap each other. This finding can be attributable to the potential antioxidant properties of lignin, even though it is right to point out that the lower content of effective bitumen within the sample as compared to the reference one could contribute to the reduced aging sensitivity of bio-binder (Luo et al., 2019; Pan, 2012), also confirming the filler role of lignin in dominating the rheological response at higher temperature which seems to prevail or hide the aging effects. However, the significant effects of lignin at high temperatures on the rheological response could hide those due to the aging.

Moreover, the same tests returned representative parameters for the determination of the performance grade (PG). The rutting parameter |G*|/sin(δ) and the fatigue parameter |G*|sin(δ) were determined and compared with limit values indicated by the Superpave system (AASHTO, 2016) were further evaluated. The rutting parameter related to the portion of the accumulated, non-recoverable deformation occurring in a pavement was considered by using DSR data evaluated at 10 rad/s and 25 mm plate-plate configuration at high service temperatures (in the current analysis, from 52 °C to 100 °C with a step of 6 °C). High values of |G*|/sin(δ) are recommended since they result in higher permanent deformation resistance (i.e., reduced rutting tendency for asphalt mixture). According to the Superpave method, a minimum value equal to 1 kPa is considered satisfactory for unaged binders, while 2.2 kPa are required for short-term aged conditions (Kennedy et al., 1994). The lowest value between the temperatures which satisfy the two mentioned limits in unaged and short-term aged condition, respectively, represents the most restrictive condition and gives the maximum in-service temperature (T_{max}) that the binder can withstand without showing rutting distress. Fig. 7(a) shows as an example the evolution of parameter |G*|/sin(δ) referred to RTFOT conditions as function of temperature. As can be observed, the rutting parameter increases in presence of lignin, resulting in a shifting of T_{max} to higher temperatures. This result is in accordance with the stiffening effect given by the introduction of powder lignin and the higher viscosity already experienced (Gaudenzi et al., 2022b). In particular, for 50/70 bitumen the unaged condition resulted the most restrictive, giving a T_{high} of 67 °C (64 °C in PG). Whereas, for the lignin-extended binder, the higher temperature of the PG was evaluated in short-term aging condition and was equal to 76 °C, giving a ΔT_{max} of 12 °C. Indeed, for a better understanding of modified binders, further parameters such as the non-recoverable creep compliance should be more representative.

The fatigue parameter |G*|sin(δ) was calculated to evaluate the aptitude to fatigue cracking of material. Specifically, a maximum value of 5000 kPa for the parameter |G*|sin(δ), evaluated on long-term aged binders at the average pavement design temperature, is considered satisfactory. The parameter refers to the DSR configuration corresponding to 8 mm plate specimen and 10 rad/s, and it is related to the contribution of the asphalt binder to the dissipation of energy in a pavement during each loading cycle. As it can be noted in Fig. 7(b), this requirement is met for both binders at a similar temperature around 26 °C, denoting a comparable fatigue performance at intermediate temperatures.

4.5. MSCR

MSCR tests were performed to quantify the non-linear behavior of investigated binders in order to predict their rutting response. Specifically, during testing the amount of residual strain occurring within a specimen due to the stress applied after repeated creep and recovery steps can be directly measured. Hence, the non-recoverable creep compliance J_{nr}, computed by the ratio between accumulated strain and applied stress, was selected as an indicator of rutting resistance of material. Moreover, the analysis of strain evaluated during the recovery steps allowed the computation of the percent recovery parameter R as useful parameter to assess the potential elastic response of binders. For each applied stress and testing temperature, the average of the non-recoverable creep compliance $J_{nr}(\tau, T)$ and the recovery parameter $R(\tau, T)$ are calculated as the mean of J_{nr} and R values evaluated during the last 10 loading cycles, respectively.

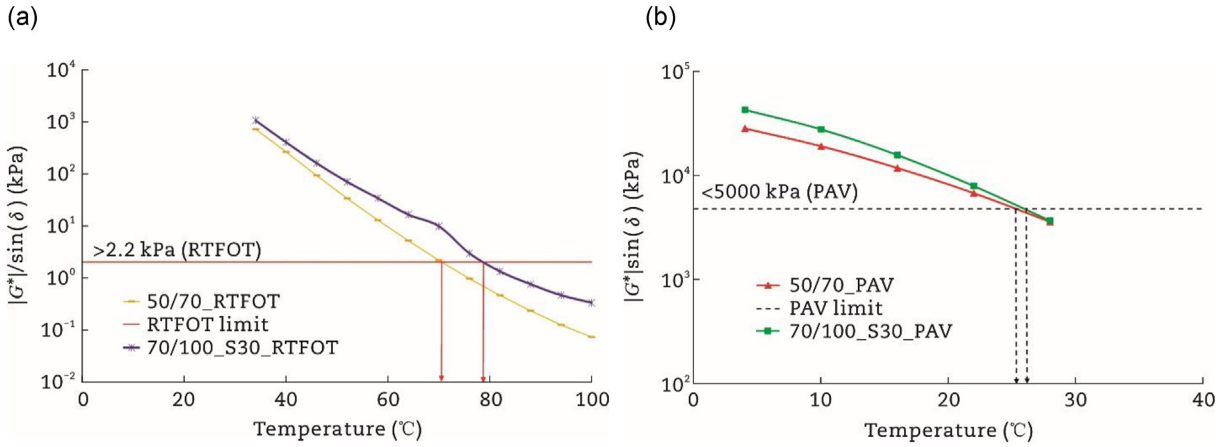

Fig. 7 – Evolution over increasing temperature. (a) Rutting parameter |G*|/sin(δ) in T_{max}. (b) Fatigue parameter |G*|sin(δ) in T_{medium}.

Figs. 8 and 9 show, respectively, the variation of J_{nr} and R-parameter with temperature at both stress levels of 0.1 and 3.2 kPa for the tested binders at the different aging conditions. As expected, J_{nr} increases while R decreases as the stress level and the temperature increases for binders in all testing condition. Indeed, this can be due to more viscous behavior of the bituminous phase at higher temperatures, which results in higher permanent strain and a penalized elastic response of the material under loading. From Fig. 8, it can be observed that the reference plain bitumen shows the greatest J_{nr} values at each temperature, stress level and aging condition performed as compared to lignin-extended binder which gives low J_{nr} values, confirming what found in previous literature works (Arafat et al., 2019; Norgbey et al., 2020; Xu et al., 2021). Moreover, the bio-binders show a reduced temperature sensitivity as lower difference in the J_{nr} trends due to the temperature increase is found with respect to the reference binder, as clearly observed in the short-term aging conditions where a wider temperature set was investigated.

As far as the elastic response is concerned (Fig. 9), it can be noted that the reference plain bitumen shows the lowest values of parameter R regardless of the testing condition. However, a higher stress sensitivity is recognized for the bio-binder due to a significant drop in parameter R with the increase in temperature in unaged and PAV aged condition at stress level of 3.2 kPa, showing a similar response to the reference bitumen in RTFOT aged condition as well. The greater R values showed by the 70/100_S30 at lower stress state indicate the higher binder capability to recover the strain caused by the stress condition applied during the rest period, as compared to the reference bitumen, however this capability of bio-binder is strictly temperature and stress dependent and tends to vanish as the temperature or stress level increases. Thus, these findings support the theory that lignin improves the anti-rutting potential of binder for a specific temperature, stress level and aging state, but it is irrelevant in terms of elastic response under specific testing conditions, so showing rheological effects more similar to those of filler addition.

MSCR test data collected on short-term aged binders were also analyzed following the AASHTO M332 specification (AASHTO, 2018b). In this regard, the parameter J_{nr} and the percent difference in non-recoverable creep compliance $J_{\mathrm{nr,diff}}$ (computed by comparing the J_{nr} measured at the two different stress levels) were used to specify the maximum in-service temperature and the related "traffic category" that

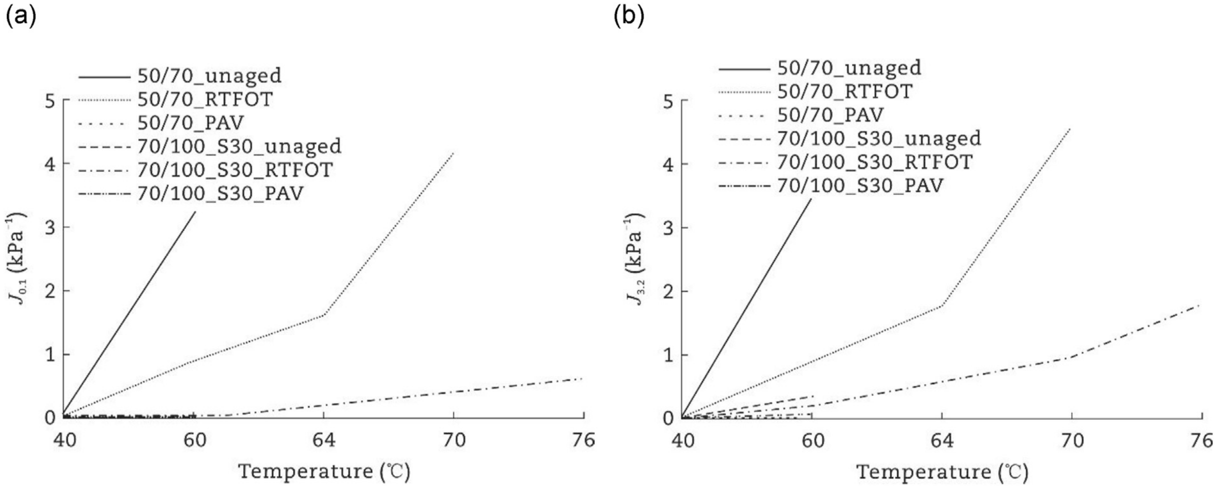

Fig. 8 – MSCR results. (a) Non-recovery at stress level of 0.1 kPa. (b) Non-recovery at stress level of 3.2 kPa.

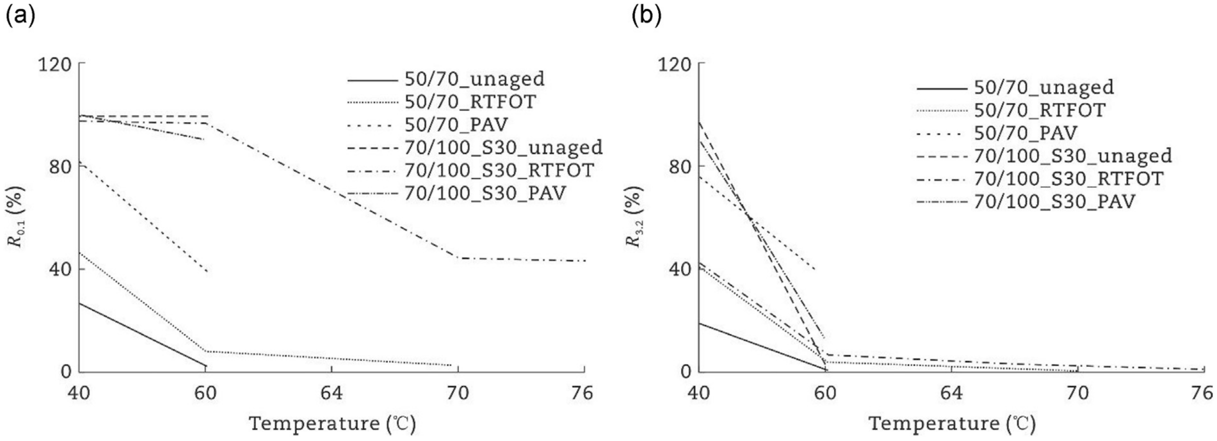

Fig. 9 – MSCR results. (a) Recovery at stress level of 0.1 kPa. (b) Recovery at stress level of 3.2 kPa.

Table 6 – Performance grade of bituminous binders.

Condition			Bituminous binder	
			50/70	70/100_S30
T_{max} (°C)	25 mm DSR plate			
	Unaged binder	$\lvert G^*\rvert/\sin(\delta) \geq 1.00$ kPa	64	>100
	Short-term aged binder	$\lvert G^*\rvert/\sin(\delta) \geq 2.20$ kPa	64	76
	MSCR	$J_{nr,diff} < 75\%$, $J_{nr,3.2} < 2\ kPa^{-1}$	64	76
T_{medium} (°C)	8 mm DSR plate			
	Long-term aged bitumen	$\lvert G^*\rvert\sin(\delta) \leq 5000$ kPa	28	31
T_{min} (°C)	BBR			
	Long-term aged bitumen	300 MPa $\leq S(60) \leq$ 600 MPa	−16	−22
	Long-term aged bitumen	$m(60) > 0.300$		
Performance grade			PG64-16	PG76-22

the material could withstand in relation to its rutting response. Based on this classification criteria, the data analysis shows that at 64 °C the reference plain bitumen meets the requirement on J_{nr} (i.e., <2 kPa) and on $J_{nr,diff}$ (i.e., <75%) with an associated traffic category "H-high traffic" whereas the bio-binder satisfies the same technical requirements and traffic category at higher temperature equal to 76 °C. This finding indicates the effectiveness of lignin to improve the binder PG grade at high temperatures, consistently with what found by the analysis of conventional rutting parameter $\lvert G^*\rvert/\sin(\delta)$.

4.6. *Evolution of the temperature range of performance grade*

Based on the data gathered by rheological tests, i.e., DSR tests (for high temperatures and verification of intermediate temperature) and BBR analysis (for low temperatures), the PG of asphalt binders was determined according to the AASHTO M320 (AASHTO, 2016). Table 6 summarizes the specification setting limits and the corresponding determined temperatures, which represent the thermal domain in which test criteria are met. According to PG classification system, the maximum and minimum in-service temperature are identified, and the PG designation was reported for both investigated binders. Results highlight a wider PG range for the lignin-extended binder. From the frequency sweep tests, the T_{max} limit of lignin-extended binder is governed by the short-term aged conditions, which gave the temperature of 76 °C as result. The same temperature is confirmed by the MSCR investigations, which allow a heavy "H" traffic condition, while the reference plain bitumen is characterized by a lower admissible T_{max} equals to 64 °C, with a consequent ΔT_{max} of 12 °C. At the same time, from the BBR analysis, a lower T_{min} is admissible for the lignin-extended binder, which equals to −22 °C vs. −16 °C of the reference bitumen, with a ΔT_{min} equals to 6 °C. Finally, as concerns fatigue performances, the T_{medium} equals to 28 °C and 31 °C that obtained for the reference and lignin-extended binder respectively, which perfectly satisfy the requirement in fact of $\lvert G^*\rvert\sin(\delta)$.

Based on the obtained results, it is evident that lignin has positive effects from a rheological point of view, leading to the extension of the PG range, with higher T_{max} and lower T_{min} with respect to a conventional 50/70 penetration grade bitumen commonly used in Italy for road applications.

5. Conclusions

This research aims to compare the rheological behavior of a bio-extended binder produced with lignin and a conventional 50/70 pen grade bitumen commonly used in Italy for road application. Specifically, the bio-binder was obtained by partially replace a 70/100 pen grade bitumen with 30% by weight of a powered lignin in order to obtain a binder characterized by a consistency (i.e., penetration grade) comparable to the reference plain bitumen (i.e., 50/70 pen grade). To this end, a laboratory investigation including chemical (FTIR and SARA analysis) and rheological analysis (frequency sweep test, MSCR and BBR tests) was performed on selected binders, considering the aging conditions as well.

Based on the presented results, the following conclusions can be drawn.

- No significant differences in functional groups are observed between the original 70/100 plain bitumen and the lignin-extended binder, thus assuming no chemical interaction between the base binder and lignin.
- SARA analysis highlighted the lower aging sensitivity of the bio-binder. Moreover, solubility test seems to show that a small part of lignin (about 7%) physically and/or chemically interacts with bitumen during the aging test.
- Overall, the lignin-extended binder shows higher stiffness and the occurrence of a rubbery asymptote in the low frequency region, highlighting that the powder lignin dominates the rheological behavior of the bio-binder. Moreover, the master curve analysis shows lower aging effect on the lignin-extended binder with respect to the plain bitumen, reclaiming the potential antioxidant properties of lignin.
- BBR data analysis highlights that the aged lignin-extended binder exhibit a better balance between stiffness and stress relaxation capability, contrarily to the aged reference bitumen which is more suffering in absorbing thermal stress despite an adequate creep stiffness.

- MSCR data analysis shows the reduced susceptibility to permanent deformation as compared to a typical plain bitumen, confirming the stiffening role of the lignin. On the other hand, the elastic response of bio-binder is strictly temperature and stress dependent and tends to vanish as the temperature or stress level increases.
- According to PG classification system, a wider temperature range is found for the lignin-extended binder, indicating an improvement of the performance at low as well as high temperatures.

Overall, the current research study supports the use of lignin as partial replacement of bitumen to produce bio-binder for road applications, so entailing environmental and economic benefits according to the sustainability principles. Specifically, the investigated lignin-extended binder was proved technically suitable as alternative to a conventional plain bitumen commonly used in Italy for road maintenance and construction, with comparable or even improved performances.

Author contributions

Conceptualization: Francesco Canestrari and Xiaohu Lu; writing–original draft preparation: Elena Gaudenzi; writing–review & editing: Fabrizio Cardone; supervision: Xiaohu Lu and Francesco Canestrari.

Conflict of interest

Francesco Canestrari is an associate editor of Journal of Traffic and Transportation Engineering (English Edition) and was not involved in the editorial review or the decision to publish this article. All authors declare that there are no competing interests.

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