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Title

Feasibility of using a lignin-containing waste in asphalt binders

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Abstract

A lot of water waste streams are produced during the production of hardboard panels. This paper analyses the feasibility of using a lignin-containing waste from the hardboard industry in asphalt binders. It would contribute to both waste reduction and decrease of the consumption of asphalt in order to obtain environmental, economic and social benefits. The waste from the hardboard industry was not subjected to any transformation i.e. it was blended directly with the conventional asphalt. Asphalt binder samples blended with 0, 5, 10, 20 and 40% of the waste were aged in a rolling thin-film oven apparatus. Basic characterisation (penetration grade, ring and ball softening point and resilience) as well as advanced characterisation (dynamic viscosity, shear complex modulus and phase angle) were performed. Asphalt binders blended with up to 20% waste can be stored, pumped and handled at hot-mix asphalt facilities. Addition of the waste to asphalt binder increases the viscosity and the shear complex modulus and reduces the phase angle. The waste produces asphalt binders with higher storage modulus and lower loss tangent. The waste enhances fatigue and rutting resistance. Asphalt binder with 20% of waste displays the best potential for use as an extender and as well as an enhancer in asphalt pavements. The research results can offer technical support to value this waste from hardboard production, without the need for subsequent transformations.

Keywords: lignin; asphalt; binder; hardboard; waste; pavement

Statement of novelty

A lignin-containing waste with the potential to be used in asphalt pavements was obtained from the excess water from the wet-forming production process of fibre hardboards.
Asphalt binders blended with the lignin-containing waste were analysed.
The lignin-containing waste can be used as asphalt binder extender.
The lignin-containing waste enhance asphalt binder performance.
The employment of the lignin- containing waste in asphalt binders contribute to minimize waste, since the waste will be valued without the need for subsequent transformations in order to obtain environmental, economic and social benefits.
No published asphalt binder research has been identified for this lignin-containing waste source from the forestry and timber industry.

1. Introduction

12 million m^3 of hardboard panels are produced in the world every year [1]. Hardboard is a composite panel product consisting of lignocellulosic fibers binded under heat and pressure [2]. The hardboard industry utilizes the so-called wet process to form the fibers into mats by using water as the distributing medium [3]. The main disadvantage of the wet process is the discharge of large volumes of waste water streams. Accordingly, the hardboard industry has to invest in effluent treatment which increases the production costs. These facts hinder hardboard development. However, these waste streams are rich in lignin and other organic materials [4].

Asphalt is a petroleum derived material which is employed as binder for mixtures in highway pavements [5]. In the last years, due to the restriction of petroleum, greater price and the undesirable effect on the environment, there has been an unceasing demand to find efficient, cost effective and environment friendly alternative binders to substitute asphalt [6,7].

In this regard, certain chemical similarities exist between lignin and asphalt, as they are both hydrocarbon materials composed mainly of carbon, hydrogen and oxygen. The lignin structure is similar to the resin fraction of an asphalt with large rings joined by alkyl chains [8-10]. Hence, a potentially attractive alternative to hardboard wastewater treatment would be to use the waste rich in lignin as an asphalt extender i.e. as partial substitute of asphalt, to decrease the amount of asphalt binder required in asphalt mixtures.

Despite this potential, no published asphalt binder research has been identified for the use of this lignin-containing waste from the hardboard industry. In contrast, research about the utilization of lignin in asphalt has been preferably oriented towards the use of lignin waste from the paper industry.

Primarily, investigations were conducted with Kraft lignin (sulphate pulping) [11-13], and later with sulfonated lignin (sulphite pulping) [14]. Furthermore, research has been carried out using Organosolv lignin (hot ethanol water mixture process) and Klason lignin (acid-based extraction process) [12,15].

An alternative lignin source for use in asphalt is the steam-exploding process used to convert biomass into ethanol [16]. Another source is bio-refineries that not only produce biofuel [7] but also wastes containing lignin [6,17,18].

Authors have investigated some properties of asphalt when lignin is used as an extender or an enhancer of the asphalt binder properties. In this way, Terrel and Rimsritong [11] and Asukar et al. [14] demonstrated that the addition of lignin to an asphalt binder results in a reduction in penetration values and an increase in the softening point. Sundstrom et al. [16] observed a decrease in ductility values with the addition of lignin, which produces a hardening effect in the asphalt binder that directly increases with the amount of added lignin.

Besides, when lignin is added to asphalt, the binder rheological properties are changed [12-15, 19]. Asphalt binders containing lignin are stiffer than neat binders and exhibit higher rutting resistance [12-14, 19-22]. However, stiffer lignin asphalt binders usually lose some ability to prevent thermal and fatigue cracking [12-14, 19,21-24]. The exception is reported by Batista et al. [22] determining that asphalt binders containing lignin have better performance to thermal cracking at up to −12 °C.

Consequently, from a technical point of view, these results are contradictory: on the one hand, lignin, enhances the rutting resistance of asphalt binders and, on the other hand, it worsens the fatigue resistance of asphalt binders. As is known, rutting and fatigue are the two main causes of failure in flexible road pavements [5].

2. Aims and scope

The main objective of this research is to investigate the feasibility of using a lignin-containing waste from hardboard production as an extender in asphalt binders. The enhancement of the asphalt binder performance is also studied. The waste will be investigated without any transformation i.e. it will be blended directly with the conventional asphalt. Several percentages of the waste will be tested, namely 0% (control mix), 5%, 10%, 20% and 40% by asphalt weight.

In order to demonstrate the viability of using this waste as an extender, it is necessary to investigate the feasibility of the asphalt binder blended with the waste to be stored, pumped and handed at hot-mix asphalt facilities as well as its rutting and fatigue performance. In this sense, to evaluate the performance of the asphalt binders, a basic characterisation test (penetration grade, ring and ball softening point and resilience) as well as an advanced characterisation test (dynamic viscosity, shear complex modulus and phase angle) were performed.

Finally, the employment of the waste in asphalt pavements would contribute to sustainable construction and to minimize waste, since this hardboard production waste, rich in raw materials, will be valued without the need for subsequent transformations, or for leftover waste materials in order to obtain environmental, economic and social benefits.

3. Materials and method

3.1. Materials

3.1.1. Asphalt

A conventional penetration-grade asphalt binder B50/70 (50–70 mm/10 penetration at 25°C and 50–58°C ring and ball softening point) was selected for the research. This asphalt binder is one of the most commonly used in Spanish asphalt pavements.

3.1.2. Hardboard water waste containing lignin

Management of excess water from the hardboard wet-forming process provides the lignin-containing waste. The waste is a viscous, dark brown coloured liquid with a faint odour of caramel (Fig. 1).

Fig. 1 Hardboard plant lignin-containing waste

The origin is 100% natural because the hardboard production process does not require artificial adhesives for the bonding of the fibers. The waste is 56.31% water and 43.69% solid matter. Its main properties are shown in Table 1.

Table 1 Main properties of the waste

Property	Value
pH	3.2
Densitity at 75ºC (kg/m^3)	1,155
Viscosity at 80ºC (mPa.s)	14
Total solids (g/L)	444
Volatile solids (g/L)	385
Sulphates (mg/g)	8
Aluminium (Al) (mg/L)	8,197
Calcium (Ca) (mg/L)	4,600
Magnesium (Mg) (mg/L)	5,500
Sodium (Na) (mg/L)	4,725
Silicon (mg/g)	<3
Vanadium (mg/kg)	<0.5
Chlorine (mg/kg)	6,500

The main components of the dry matter are: 41.46% sugar (mainly xylose, glucose, galactose, rhamnose, arabinose and mannose), 23.39% lignin (16.29% insoluble Klason lignin and 7.10% soluble lignin), 13.3% pectin, 11.8% polyphenols, 9.05% mineral matter and other compounds that appear in low percentages. The high sugar content explains the caramel-like odour, which is particularly noticeable when the waste is blended with asphalt.

3.2. Laboratory testing program

3.2.1. Preparation of asphalt binder

The asphalt binder B50/70 was blended directly with the lignin-containing waste, with 5, 10, 20 and 40% of asphalt weight. The samples were blended using a low speed shear mixer (300 rpm) with a temperature control system. Initially, the 160°C temperature blending resulted in very strong volumetric expansion, which caused bubble formation. For this reason, the 60°C initial temperature blending was gradually increased over 30 min until it reached 160°C, and the bubbles disappeared after a certain time. The samples were blended at 160°C for approximately 60 min.

Following blending completion, the asphalt characterisation (needle penetration, softening point, resilience, dynamic viscosity, and shear complex modulus and phase angle) was assessed under the following four conditions:

1. Neat asphalt binder;
2. Neat asphalt binder after ageing with rolling thin-film oven test (RTFOT);
3. Asphalt binder with 5, 10, 20 and 40% waste by asphalt weight; and
4. Asphalt binder with 5, 10, 20 and 40% waste by asphalt weight following ageing with RTFOT.

Furthermore, the microstructures of the neat asphalt binder and asphalt binders with the waste were examined. Also, the storage stabilities of the neat asphalt binder and asphalt binder with the waste were investigated.

3.2.2. Rolling thin-film oven test

The RTFOT simulates the short-term ageing occurring in the asphalt binder during the mixing process and pavement construction period. The UNE-EN 12607-1 (Determination of the Resistance to Hardening under Influence of Heat and Air. Part 1: RTFOT Method) [25] standard was used in this work for assessing the binder ageing.

In this test, eight glass bottles were loaded with 35 g of asphalt binder (Fig. 2a). The bottles were placed into an oven with a rolling carriage at a speed of 15 rpm for 75 min (Fig. 2b), and the test temperature was 163°C. During the test, the airflow was set to a rate of 4000 ml/min. Six bottles were used to prepare samples for the asphalt binder characterisation (Fig. 2c), and two bottles were used to determine the mass loss. The RTFOTs were performed on the neat binder and asphalt binders with the waste.

Fig. 2 RTFOT: a) bottles loaded with asphalt prior to ageing; b) bottles loaded with asphalt placed in oven; and c) bottles loaded with asphalt following ageing

3.2.3. Basic characterisation

The asphalt binder tests performed consisted of needle penetration, ring and ball (R&B) softening point, and resilience. The penetration test measured the asphalt binder consistency by means of an indentation test and was based on the UNE-EN 1426 standard (Determination of needle penetration) [26]. The penetration value was measured by the distance in tenths of millimetres that a standard needle would penetrate vertically into an asphalt binder sample under standard test conditions. The lower the value of penetration, the harder the bitumen.

The softening point test was conducted to measure the temperature at which the asphalt softens. The standard used for this test is the UNE-EN 1427 (Determination of the softening point ring and ball method) [27].

The resilience test determined the elastic properties of the asphalt binder, expressed as a percentage of its recovery. The standard used for this test is the UNE-EN 13880-3 (Test method for the determination of penetration and recovery (resilience)) [28]. It measured the rebound after a special ball is pushed into the asphalt binder under standard test conditions. Asphalt binder penetration, R&B softening point temperature, and resilience values were obtained for the four conditions indicated in section 3.2.1: (1) Neat asphalt binder; (2) neat asphalt binder after ageing with rolling thin-film oven test (RTFOT); (3) asphalt binder with 5, 10, 20 and 40% waste by asphalt weight; and (4) asphalt binder with 5, 10, 20 and 40% waste by asphalt weight following ageing with RTFOT.

3.2.4. Microstructure

The confocal laser scanning microscopy (CLSM) method used by Bearsley et al. [29] and Handle et al. [30] was employed to observe the microstructure of asphalt samples. CLSM requires minimal sample preparation, which reduces the possibility of producing images that are not representative of the original material.

A spectral confocal microscope, the Nikon A1 R, was used to obtain CLSM images of the asphalt. The asphalt was irradiated with 488 nm wavelength light in order to observe the fluorescence in the 515–545 nm range. The microstructures of the neat asphalt binder and asphalt binders with 10, 20 and 40% waste by asphalt weight were observed. The asphalt sample was placed on a slide and observed at ambient temperature and pressure.

3.2.5. Storage stability

The storage stabilities of the neat asphalt binder and asphalt binders with 5, 10, 20 and 40% waste by asphalt weight were evaluated according to the standard UNE-EN-13399 (Determination of Storage Stability of Modify Bitumen) [31]. In this process, the asphalt binder is stored for a three-day period at 180ºC in an aluminium toothpaste tube. Thereafter, both the softening point test based on UNE-EN 1427 [27] and penetration test based on UNE-EN 1426 [26] are carried out on the binder placed in the top and bottom parts of the tube. In order to define whether an asphalt binder is stable under storage, a maximum difference of 5ºC between the top and bottom R&B temperature is specified [32]. Moreover, a maximum difference of 9×10^{-1} mm between the top and bottom penetration is required [32].

3.2.6. Dynamic viscosity

Dynamic viscosities were obtained at temperatures between 100 and 180°C, by means of a procedure presented by Silva et al. [33]. The standard used is UNE-EN 13302 (Determination of Dynamic Viscosity of Bituminous Binder Using a Rotating Spindle Apparatus) [34].

In the test, a Brookfield rotational viscometer (RV) and a Thermosel temperature control system were used. A torque was applied to a spindle rotating in a special sample container containing the asphalt binder sample. The relative resistance of the spindle to rotation was determined, providing a measure of the asphalt binder dynamic viscosity. Asphalt binder dynamic viscosities were obtained for the four conditions mentioned in section 3.2.1.

3.2.7. Shear complex modulus and phase angle

The shear complex modulus G^* and phase angle δ were obtained using a dynamic shear rheometer (DSR) apparatus. G^* and δ characterize the viscous and the elastic behaviour of asphalt binders. The standard used is UNE-EN 14770 (Determination of Complex Shear Modulus and Phase Angle. Dynamic Shear Rheometer (DSR)) [35]. During the test, different levels of a known oscillatory shear stress were applied to the asphalt binder sample, and the resulting shear strain and time lag were measured. This method was required to guarantee that specimens were tested in the linear region at each temperature and a frequency of 10 rad/s.

Two configurations were used in keeping with the test temperatures. A 25 mm diameter plate and a 1 mm gap was used to conduct the tests at temperatures of 46, 52, 58, 64, 70, 76, 82 and 88°C. At temperatures of 19, 25, 31, 37 and 40°C, the DSR test was performed using an 8 mm diameter plate and a 2 mm gap. Rheological characterisation was carried out for the four conditions mentioned in section 3.2.1.

The shear complex modulus G^* is the ratio between the peak stress and peak strain. The phase angle δ indicates the lag in the stress response compared to the applied strain. For purely elastic materials, the phase angle is zero, while for purely viscous materials, the phase angle is 90°. The shear storage modulus $G^{\prime}=G^*\times\cos(\delta)$ can be obtained from G^* and δ. The storage modulus represents the in-phase component of the complex modulus (elastic portion). Moreover, the shear loss modulus $G^{\prime\prime}=G^*\times\sin(\delta)$ can be obtained. The loss modulus represents the out-of-phase component of the complex modulus (viscous portion). Finally, the loss tangent can be obtained simply as the tangent of the phase angle, or as the ratio of the loss to storage modulus: $\tan(\delta)=G^{\prime\prime}/G^{\prime}$. This parameter indicates the asphalt binder relative behaviour in terms of the elastic and viscous portions. Goodrich [36,37] revealed that the $\tan(\delta)$ values of RTFOT aged asphalt binders correlate strongly with the fatigue and permanent behaviour of hot asphalt mixes. Goodrich [36,37] demonstrated that asphalt binders with a lower loss tangent exhibit superior fatigue and rutting resistance. That is, because $\tan(\delta)$ is equal to the $G^{\prime\prime}/G^{\prime}$ ratio, a decrease in this ratio indicates that the asphalt binder has a higher storage modulus (elastic portion of shear complex modulus), and therefore improved elastic properties.

4. Results and discussion

4.1. RTFOT ageing

The purposes of this procedure are: a) provide an aged bitumen that can be used for additional testing of physical and rheological properties and b) determine the mass quantity of volatiles lost from the bitumen during the test to assess its aging. As stated earlier, the tested asphalt binders are elaborated with 0, 5, 10, 20 and 40% waste by asphalt weight.

Fig. 3 illustrates the mass loss results obtained using the RTFOT. The mass loss increased with the blended waste percentage. As recommended by the Asphalt Institute [5], the mass loss does not exceed 1%.

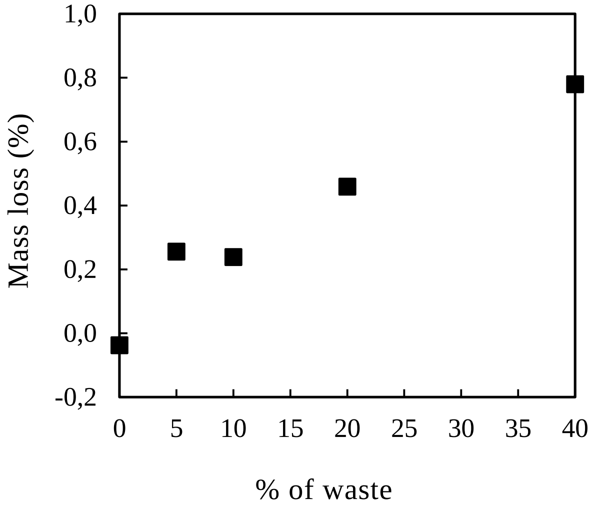

Fig. 3 Mass loss following RTOFT ageing

Therefore, excessive ageing is not expected from volatilisation during the mixing and placement of the hot-mix asphalt. The neat binder sample unexpectedly experienced a mass increase, which was possibly owing to certain oxidation and dehydrogenation reactions. Another possible explanation is an increase in the asphaltenes (components with higher molecular weight) proportion induced during ageing [38].

4.2. Asphalt binder basic characterisation

In this section are determined: a) the asphalt binder consistency by means of the penetration test, b) the temperature at which the asphalt softens by means of the softening point test and c) the elastic properties of the asphalt binder by means of the resilience test. As stated earlier, the asphalt binders tested are elaborated with 0, 5, 10, 20 and 40% waste by asphalt weight.

Fig. 4 illustrates the results of the penetration tests (Fig. 4a), ring and ball (R&B) softening point tests (Fig. 4b) and resilience tests (Fig. 4c) carried out on the studied samples. In Fig. 4a, as expected, the asphalt samples without RTOFT ageing exhibit higher penetration values that the samples with RTOFT ageing. The penetration values of the asphalt samples with and without RTOFT ageing tend to decrease when the waste percentage increases; therefore, the amount of blended waste affects the penetration values. When additional waste, the asphalt with and without RTFOT ageing becomes harder.

In Fig. 4b, the asphalt samples without RTOFT ageing exhibit lower R&B temperatures than those with RTOFT ageing. In both cases, the R&B temperature increases when the waste percentage increases. Increasing the amount of waste reduces the tendency of the asphalt binder to flow.

In Fig. 4c, the asphalt samples without RTOFT ageing exhibit lower resilience values than those with RTOFT ageing, indicating that the samples with RTOFT ageing are more elastic. In both cases, the resilience increases when the waste percentage increases; increasing the amount of waste increases the asphalt binder elasticity.

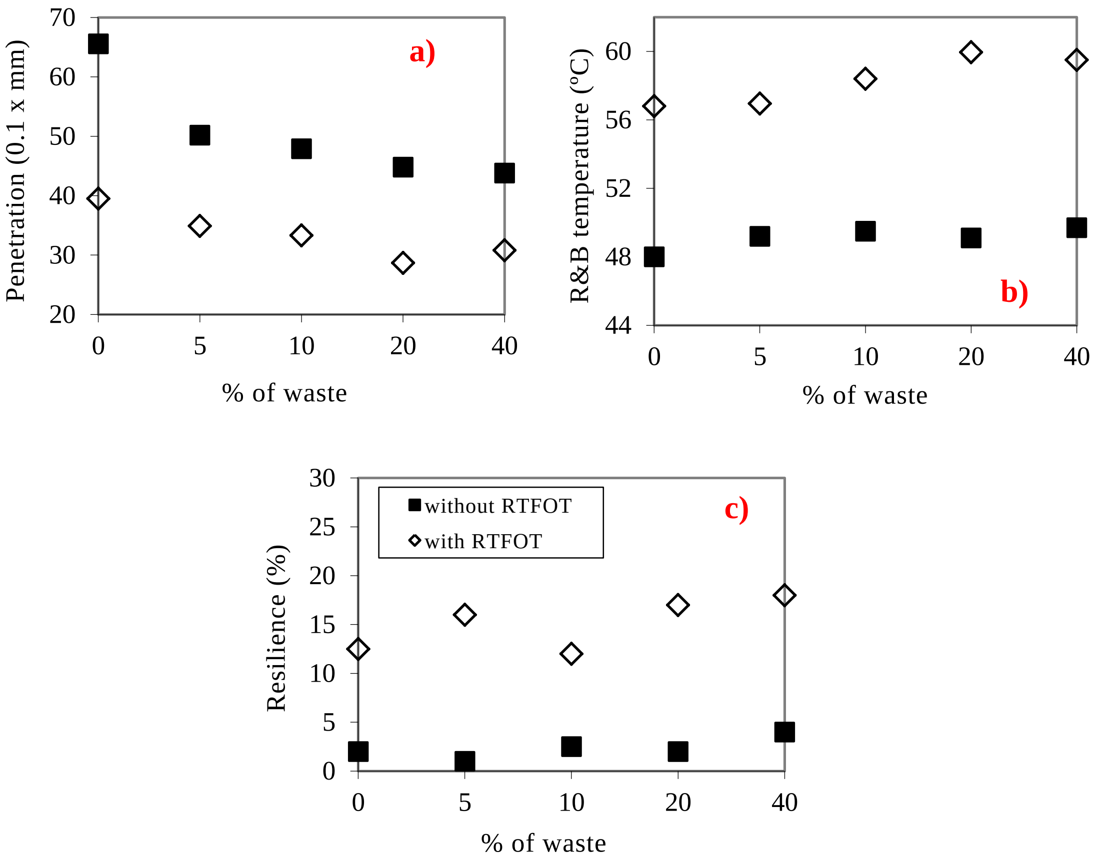

Fig. 4 Asphalt binder basic characterisation: a) penetration tests; b) R&B softening point tests; and c) resilience tests

4.3. CLSM microstructure images

In this section is examined the microstructure (morphology) of asphalt samples with 0, 10, 20 and 40% waste by asphalt weight.

Fig. 5 illustrates the microstructures of the neat asphalt binder and asphalt binders with 10, 20 and 40% waste. Fluorescing regions can clearly be seen in each asphalt sample CLSM image. In Fig. 5a (neat asphalt), the fluorescence is attributed to the asphaltene structure forming a disperse phase in the continuous maltene matrix (green colour) [29].

In Fig. 5b (10% waste), alongside the fluorescing regions certain dark particles are visible, attributed to the waste particles. In Fig. 5c (20% waste) many more waste dark particles are distributed across the binder. In Fig. 5d, larger waste dark particles with a rounded form exist. In this specific case, certain smaller dark particles have coalesced, forming large rounded particles. In all of the figures, the waste (dark colour) is always dispersed in the asphalt matrix (green colour). Therefore, the asphalt binders with waste binders never form a continuous, rich waste phase; in contrast, a continuous asphalt-rich phase is always formed.

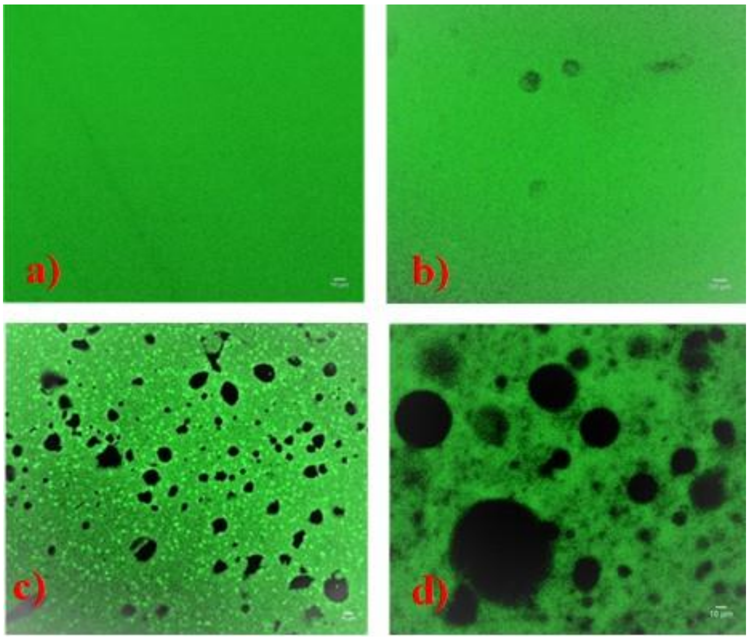

Fig. 5 CLSM images: a) 0% waste; b) 10% waste; c) 20% waste; d) 40% waste

4.4. Storage stability

In this section is examined the storage stability of asphalt samples with 0, 5, 10, 20 and 40% waste by asphalt weight.

Table 2 displays the storage stability results. In the bottom and top parts of the tube, the softening point temperature values increase with the waste amount. The differences between the softening point top and bottom temperatures are lower than the permitted maximum difference of 5ºC. An exception is the 40% waste, indicating that a certain amount of waste decantation is produced.

However, in the top and bottom part, there is a tendency of decreasing penetration values with an increased waste amount. The differences between the top and bottom penetration are lower than the permitted maximum difference of 9×10^{-1} mm, although an exception is observed for the 40% waste.

Table 2 Storage stability results in terms of softening point and penetration tests

Test	% waste 0	 5	 10	 20	 40
Top, softening point (ºC)	48.2	48.7	49.4	50.4	49.5
Bottom, softening point (ºC)	48.3	47.8	50.9	49.9	63.8
Difference in softening point (ºC)	0.1	-0.9	1.5	-0.5	14.3
Top, penetration (0.1 mm)	67.6	49.8	45.8	46.3	46.2
Bottom, penetration (0.1 mm)	66.9	48.2	45.6	46.1	34.8
Difference in penetration (0.1 mm)	0.7	1.6	0.2	0.2	11.4

Therefore, stability problems are not expected, owing to waste phase separation during storage of the asphalt binders with 5, 10 and 20% waste. In contrast, the asphalt binder with 40% waste is not suitable for storage. This result is related to the microstructure observed in Fig. 5d. With 40% waste, the smaller particles coalesce, forming large rounded particles that decant when the binder is stored at high temperatures, thereby resulting in a loss of stability.

4.5. Asphalt dynamic viscosities

In this section are defined the dynamic viscosities at high temperatures of asphalt binders with 0, 5, 10, 20 and 40% waste by asphalt weight. The dynamic viscosities at high temperatures determine the thermal susceptibility and the resistance to flow of the samples. These provide some assurance that asphalt binders can be pumped and handled at the hot mix facility.

Fig. 6 a) and Fig. 6 b) illustrate the dynamic viscosity of the neat asphalt binder (B) and asphalt binders blended with 5% (B5), 10% (B10), 20% (B20) and 40% (B40) waste. Increasing the temperature produces a decrease in the viscosity. However, when the waste amount increases, the viscosities do not exhibit a clear tendency. Between 100°C and 110°C, addition of the waste produces an increase in the dynamic viscosities, except for the B20 asphalt binder. When the waste is added, the dynamic viscosities do not increase between 110°C and 180°C, except for the case of asphalt binder B20.

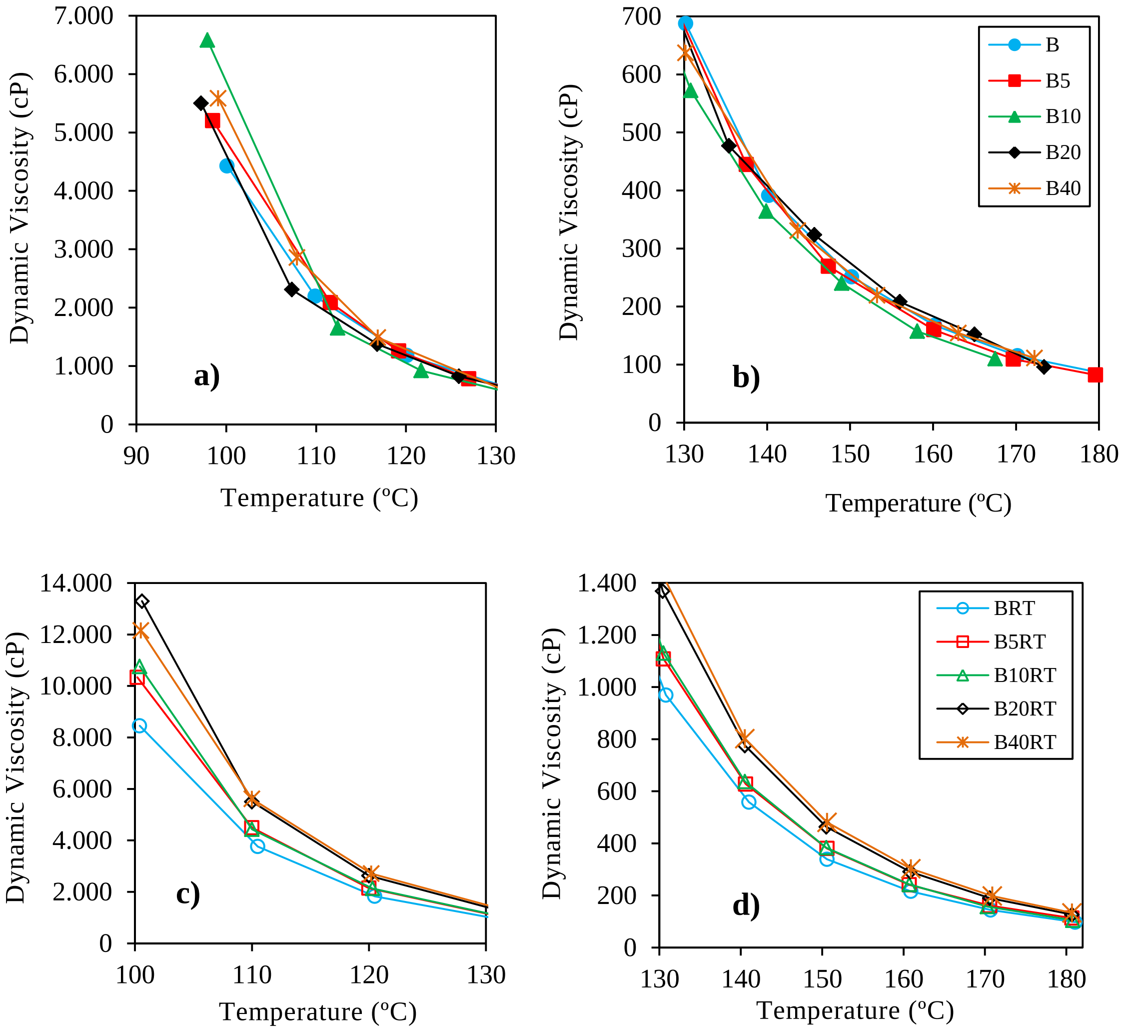

Fig. 6 Dynamic rotational viscosity at high temperatures: a) Unaged binder samples temperature ranging from 90 ºC to 130 ºC; b) Unaged binder samples temperature ranging from 130 ºC to 180 ºC; c) RTFOT aged binder samples temperature ranging from 100 ºC to 130 ºC; d) RTFOT aged binder samples temperature ranging from 130 ºC to 180 ºC.

Fig. 6 c) and Fig. 6 d) illustrate the dynamic viscosity of the RTFOT aged neat asphalt binder (BRT) and RTFOT aged asphalt binders with 5% (B5RT), 10% (B10RT), 20% (B20RT) and 40% (B40RT) waste. Following RTFOT ageing, when the waste amount increases, the viscosity values exhibit a clear tendency. The addition of waste to the neat binder produces an increase in the dynamic viscosity, which is related to the amount of waste added. In Fig. 6 c) and Fig. 6 d), a higher amount of waste results in greater viscosity. The results in Fig. 6 c) and Fig. 6 d) differ from those observed in Fig. a) and Fig. 6 b), possibly because the samples with waste have lost certain unstable or volatile components following RTFOT ageing.

Fig. 7 illustrates the dynamic viscosity at 135°C for aged and unaged asphalt binder samples. According to the Asphalt Institute [5], a maximum viscosity of 3000 cP at 135°C is required for effective mixability and workability of an asphalt binder. For the unaged samples, the viscosity varies between 545 cP (B binder) and 500 cP (B40 binder); for aged samples, the viscosity varies between 800 cP (BRT binder) and 1145 cP (B40RT binder). Therefore, a viscosity of 3000 cP is not exceeded at 135°C, and the asphalt binders can therefore be pumped and handled at the hot-mixing facility [5].

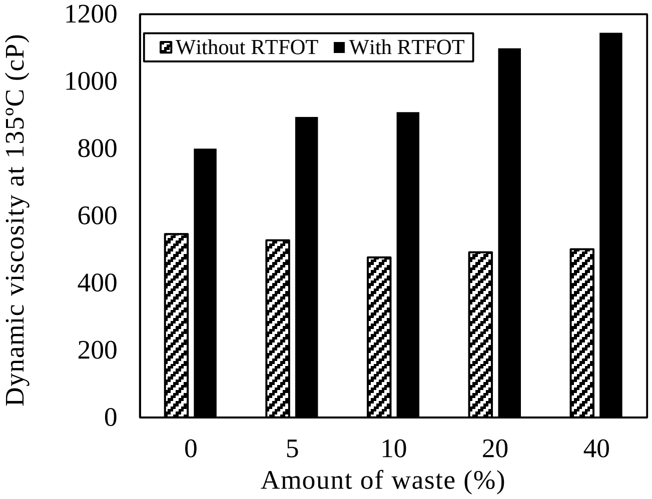

Fig. 7 Dynamic rotational viscosities at 135°C

In Fig. 7, the unaged samples exhibit similar dynamic viscosities; there is a possible slight tendency of decreasing viscosity with an increase in the waste amount, because a certain unstable component is present in the samples. However, this effect is beneficial as it is possible to reduce the asphalt binder mixing temperature slightly in order to manufacture hot-mix asphalt. The RTFOT aged asphalt binder samples exhibit different viscosities and the tendency is increased viscosity with an increase in the waste, possibly because the unstable component is removed following RTFOT aging.

4.6 Shear complex modulus and phase angle

In this section are analysed the rheology characteristics ($G^*$and δ) of asphalt binders with 0, 5, 10, 20 and 40% waste by asphalt weight. For that end, isochronal plots are used. An isochronal plot is a curve in a graph representing the behaviour of a system at a constant frequency. In this case, the curves of G^*or δ versus the temperature at constant frequencies are the isochrones. As we stated earlier, G^* and δ are used to characterize both the viscous and the elastic behaviour of asphalt binders. The viscous and the elastic behaviour at high and intermediate temperatures predict the permanent deformation and fatigue cracking of asphalt binders.

Fig. 8 a) and Fig. 8 b) display the isochronal plots of the shear complex modulus (G^*) in the unaged asphalt binder samples. In Fig. 8 a), G^* decreases with the addition of waste at 19°C and 25°C; above 31°C, G^* increases with the addition of waste. The exception is for asphalt binder B10 at 31°C. In Fig. 8 b), between 46°C and 88°C, there is an increase in G^* with an increase in the amount of waste. Between 46°C and 88°C, 40% of the waste produces the maximum G^* (Fig. 8 b).

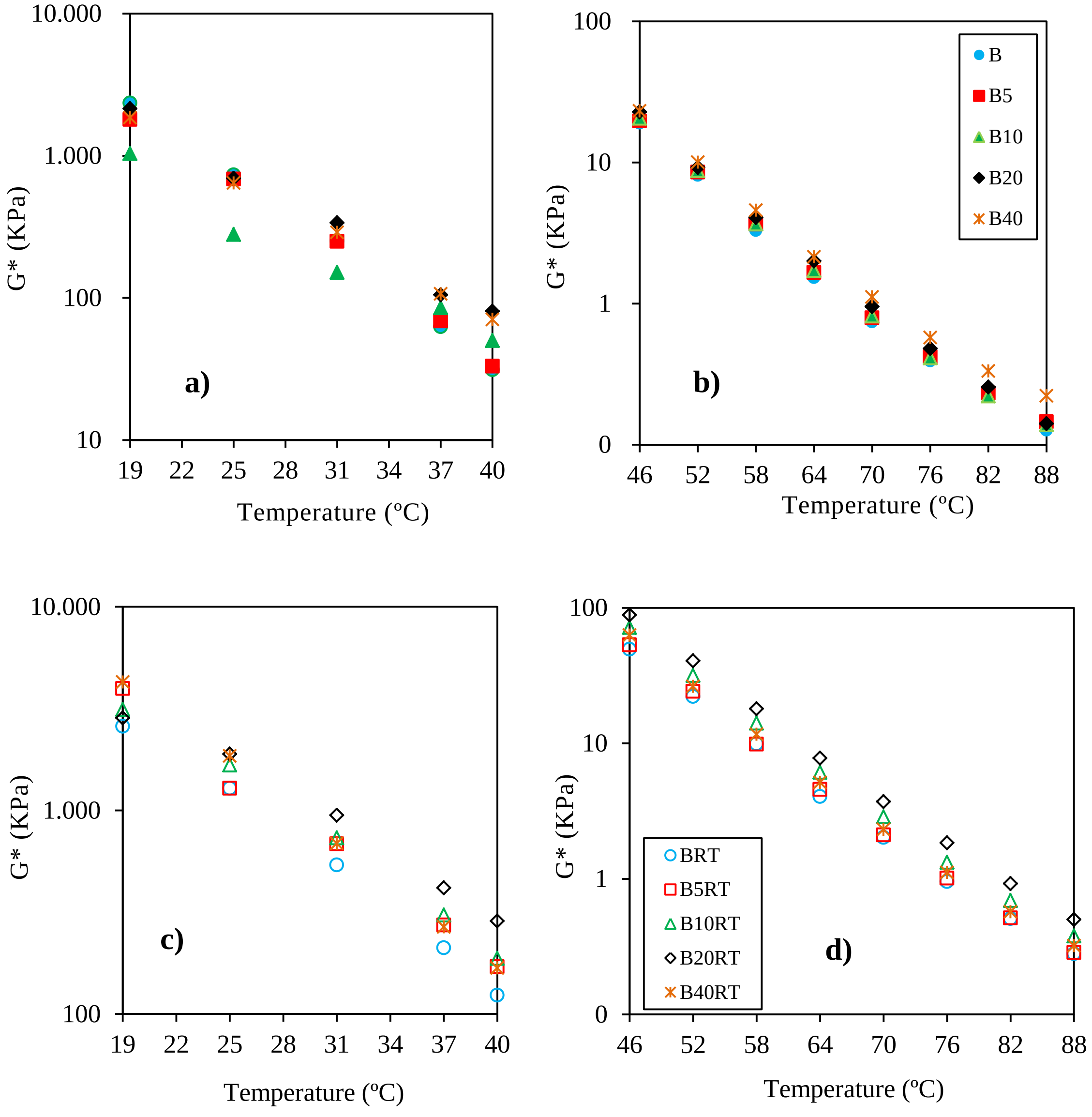

Fig. 8 Isochronal plots of shear complex modulus: a) Unaged asphalt binder samples 8 mm plate; b) Unaged asphalt binder samples 25 mm plate; c) RTFOT aged asphalt binder samples 8 mm plate; d) RTFOT aged asphalt

Fig. 8 c) and Fig. 8 d) display the isochronal plots of the shear complex modulus (G^*) for RTFOT aged asphalt binder samples. When Fig. 8 c) and Fig. 8 d) is compared to Fig. 8 a) and Fig. 8 b), it can be observed that the asphalt binders following RTFOT ageing exhibit a higher shear complex modulus than the unaged asphalt binder samples. In Fig. 8 c) and Fig. 8 d), G^* increases with the waste over the entire temperature range. The waste produces a visibly stiffer asphalt binder. In the aged asphalt binders, 20% of the waste produces the maximum G^* between 31°C and 88°C (Fig. 8 d).

In a road pavement, when an asphalt binder reaches high temperatures, stiffening creates a positive effect, as high G* values are required for rutting resistance. However, at intermediate temperatures, after several years of service, strong stiffening may produce fatigue cracking of the asphalt pavement [5].

Fig. 9 a) and Fig. 9 b) display the isochronal plots of the phase angle (δ) for the unaged asphalt binder samples. In Fig. 9 a) and Fig. 9 b), the phase angles of the asphalt binders exhibit the following increasing and decreasing trend with increasing temperature:

I. Phase angle increase trend: the phase angle is primarily affected by the asphalt-rich matrix at lower temperatures; as the temperature increases, the binder begins to flow and becomes more viscous.

II. Phase angle decrease trend: the waste particles predominantly influence the decreasing phase angle, because these particles are significantly stiffer than the asphalt-rich matrix.

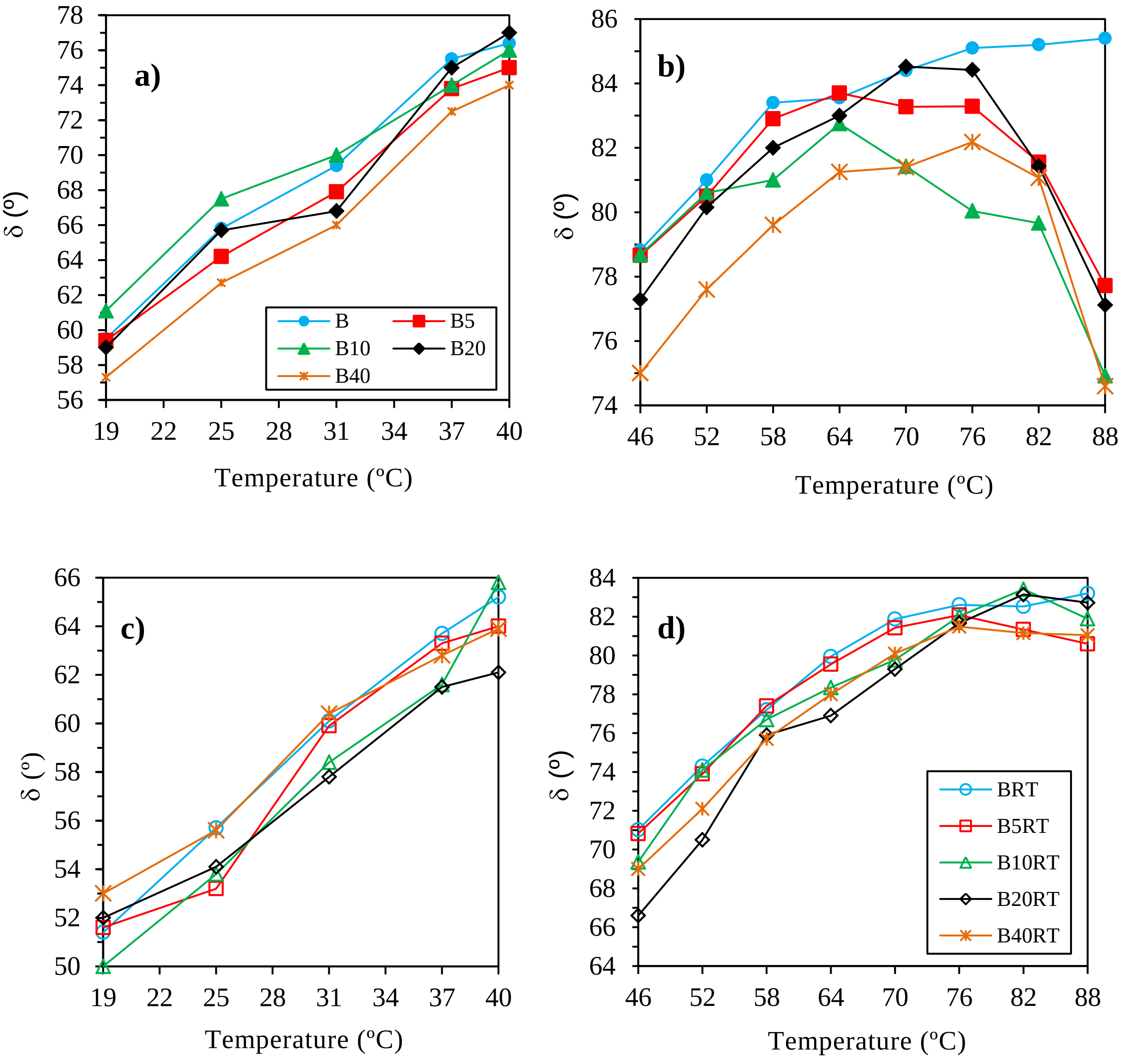

Fig. 9 Isochronal plots of phase angle: a) Unaged asphalt binder samples 8 mm plate; b) Unaged asphalt binder samples 25 mm plate; c) RTFOT aged asphalt binder samples 8 mm plate; d) RTFOT aged asphalt binder samples 25 mm plate

The phase angle of the neat asphalt binder (B) without waste particles exhibits only an increasing trend approaching 86°C (Fig. 9 b), indicating predominantly viscous behaviour. Therefore, the waste improves the elastic behaviour (response) of the binders compared to the neat asphalt binder. These results are similar to those revealed in the phase angle isochronal plots of asphalt concrete mixes [37,39] and phase angle isochronal plots of polymer-modified asphalt [40,41].

Fig. 9 c) and Fig. 9 d) display the isochronal plots of the phase angle (δ) for RTFOT aged asphalt binder samples. In this case, a less accentuated increasing and decreasing trend is observed with increasing temperature. For the most part, the RTFOT aged asphalt binder samples are more elastic than the unaged samples, because they have lower δ values. In Fig. 9 c) and Fig. 9 d), δ decreases with the amount of waste at all temperatures except for 82°C.

Therefore, the waste produces asphalt binders that are more elastic than the neat asphalt binder. In service road pavements, asphalt binders will recover significantly more deformation from an applied load. The phase angle results agree with the resilience test results (Fig. 3c).

Following RTFOT ageing at intermediate temperatures (19, 25, 31, 37 and 40°C), an increase in the waste amount increases the shear complex modulus G^* and decreases the phase angle δ. This is a positive effect, as long as the G^* values following several years of pavement service remain sufficiently low to avoid fatigue cracking [5]. However, following RTFOT ageing, the waste increases the shear complex modulus G^* and decreases the phase angle δ. This is a positive effect, as RTFOT aged asphalt binders with high G^* values and low δ values are stiff and elastic binders. These are desirable qualities for rutting resistance at high temperatures [5].

Finally, the stiffening effect of the lignin-containing waste in the asphalt binder can be applied conjointly with warm-mix asphalt (WMA) and half-warm-mix asphalt (HWMA) technologies [42,43]. Therefore, it would be possible to apply asphalt mixtures at lower production temperatures, without a decrease in the final stiffness.

4.7. Loss tangent and storage modulus

In this section are defined the loss tangent and storage modulus of aged asphalt binders with 0, 5, 10, 20 and 40% waste by asphalt weight. For that end, isochronal plots curves of tan(δ) and G' versus temperature are represented. As stated earlier, the loss tangent and the storage modulus are used to obtain the elastic and the viscous portion of the shear complex modulus.

Fig. 10 a) and Fig. 10 b) illustrate the loss tangent versus intermediate and high temperatures in RTFOT aged asphalt binders. The figures indicate a prevailing viscous behaviour over the entire tested temperature interval (tan(δ) >1). When the temperature rises, tan(δ) increases, because the loss modulus G'' (viscous portion) increases with respect to G' (elastic portion). From 76°C, when the temperature rises, tan(δ) decreases in asphalt binders B5RT and B40RT, and from 82°C, tan(δ) decreases in binders B10RT and B20RT (Fig. 10 b).

In Fig. 10 a), at 19°C only B10RT binder has a lower tan(δ) value than the neat BRT asphalt binder, while binders B5RT and B20RT have similar values. Between 25°C and 37°C, B5RT, B10RT and B20RT0 have lower tan(δ) values than the BRT neat asphalt binder. At 31°C, BRT40 has a slightly higher tan(δ) value than BRT, and at 40°C, B10RT has a higher tan(δ) value than BRT. At 19°C, the addition of 5 and 20% waste produces a very small fatigue worsening, which is slightly higher with the addition of 40%. In contrast, between 25°C and 40°C, the addition of 5, 10, 20 and 40% waste improves the asphalt binder fatigue performance. The inexplicable exception to this behaviour is B10RT at 40°C, while binder B20RT exhibits the best fatigue behaviour at 31, 37 and 40°C.

In Fig. 10 b), between 46°C and 76°C, tan(δ) is lower in asphalt binders B5RT, B10RT, B20RT and B40RT than in neat asphalt binder BRT. At 82°C, tan(δ) is lower in asphalt binders B5RT and B40RT, and at 88°C, tan(δ) is higher in asphalt binder BRT. Therefore, addition of the waste to the neat asphalt binder improves rutting resistance, except at 82°C.

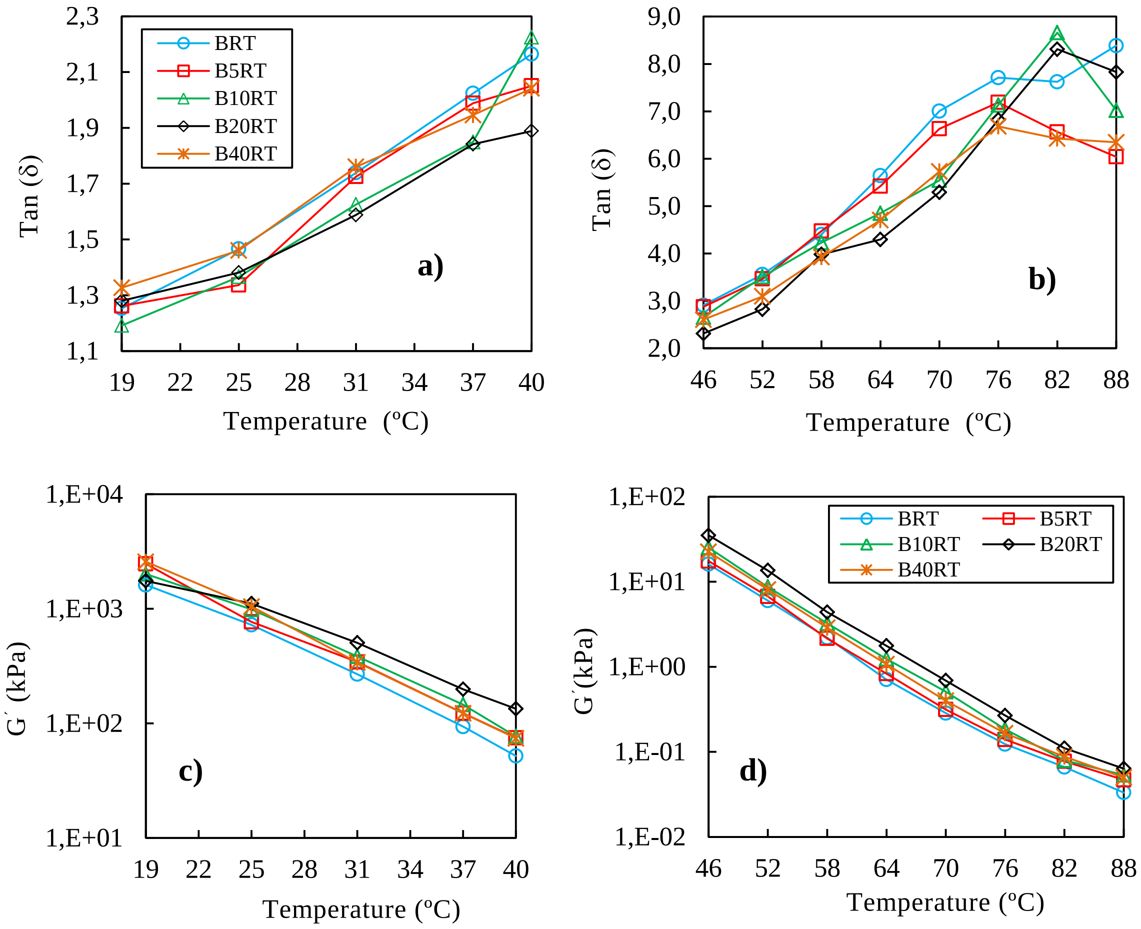

Fig. 10 a) Loss tangent versus temperature in RTFOT aged asphalt binders samples 8 mm plate b) Loss tangent versus temperature 25 mm plate; c) Storage modulus versus temperature in in RTFOT aged asphalt binder samples 8 mm plate; d) Storage modulus versus temperature in in RTFOT aged asphalt binder samples 25 mm plate

Fig. 10 c) and Fig 10 d) display the storage modulus versus intermediate and high temperatures in RTFOT aged asphalt binders. Fig. 10 c) indicates that the storage modulus (G´) increases with an increase in waste content in the entire intermediate temperature range. The addition of waste produces a more elastic asphalt binder, favouring improved fatigue performance, at intermediate temperatures.

Fig. 10 d) illustrates the storage modulus (G') versus temperature between 46 and 88°C. The figure indicates that the storage modulus (G') increases with addition of the waste. Therefore, addition of the waste produces a more elastic and less viscous asphalt binder, improving rutting resistance, at high temperatures. The B20RT asphalt binder has a higher storage modulus and presumably offers the best rutting resistance.

4.8. Superpave rutting resistance evaluation

The Superpave methodology, developed as part of the Strategic Highway Research Program (SHRP) in the USA, evaluates rutting resistance by means of the ratio $G^*/\sin(\delta)$ [5] for higher temperatures. Higher $G^*/\sin(\delta)$ values indicate stiff elastic asphalt binders, and consequently, improved rutting resistance at high temperatures.

It has been established that Superpave specifications require two joint conditions in order to address rutting (at a determined high temperature) [5]:

- A minimum $G^*/\sin(\delta)$ value of 1 kPa is required in the unaged asphalt binder (original asphalt binder).
- A minimum $G^*/\sin(\delta)$ value of 2.2 kPa is required in the RTFOT aged asphalt binder (asphalt binder following mixing process and pavement construction period).

Tables 3 and 4 display the $G^*/\sin(\delta)$ values versus temperatures in unaged and RTFOT aged binders, respectively. In Table 3, the unaged binders B, B5, B10 and B20 resist rutting until 64°C, while the B40 binder resists rutting until 70°C. For 76, 82 and 88°C, all $G^*/\sin(\delta)$ binder values are below 1 kPa; therefore, the asphalt binders do not resist rutting at these temperatures. Table 3 indicates that a greater waste content increase $G^*/\sin(\delta)$ improves the rutting resistance of the binders.

Table 3 Rutting resistance in unaged asphalt binder samples in terms of $G^*/\sin(\delta)$ values

T (ºC)	**B**	**B5**	**B10**	**B20**	**B40**
46	19.35	20.06	20.99	23.41	24.02
52	8.19	8.65	8.86	9.35	10.28
58	3.32	3.68	3.71	4.11	4.66
64	1.53	1.67	1.73	2.02	2.17
70	0.75	0.80	0.82	0.95	1.13
76	0.39	0.42	0.42	0.48	0.58
82	0.22	0.23	0.23	0.26	0.34
88	0.13	0.15	0.14	0.14	0.23

Table 4 indicates that the aged BRT and B5RT binders resist rutting until 64°C, while the B10RT, B20RT and B40RT binders resist rutting until 70°C. For 76, 82 and 88°C, all $G^*/\sin(\delta)$ binder values are below 2.2 kPa; therefore, the binders do not resist rutting at these high temperatures. A continuous increase in rutting resistance ($G^*/\sin(\delta)$ is produced in neat asphalt binder BRT and binders B5RT, B10RT and B20RT with an increased waste amount. The B40RT asphalt binder decreases rutting resistance with respect to asphalt binder B20RT.

Table 4 Rutting resistance in RTFOT aged asphalt binder samples in terms of $G^*/\sin(\delta)$ values

T (ºC)	**BRT**	**B5RT**	**B10RT**	**B20RT**	**B40RT**
46	52.27	56.31	76.47	96.31	67.23
52	23.01	25.17	32.97	43.09	27.62
58	10.15	10.11	14.47	18.56	12.04
64	4.11	4.65	6.25	7.99	5.27
70	2.04	2.13	2.92	3.78	2.36
76	0.96	1.02	1.34	1.86	1.13
82	0.51	0.52	0.70	0.93	0.58
88	0.28	0.29	0.38	0.51	0.33

According to Superpave specifications, the optimum blended waste content for rutting resistance is 20%. As expected, this result agrees with those observed in Fig. 10 (d), where the maximum storage modulus is demonstrated for binder B20RT.

5. Conclusions

This study has investigated the feasibility for using asphalt binders blended with a lignin-containing waste from hardboard production. Based on the results achieved in the research, the following conclusions can be drawn.

- Asphalt binders blended with 5, 10 and 20% of lignin-containing waste can be stored, pumped and handled at hot-mixing facilities. Also, they do not age excessively, owing to volatilisation during hot-mix asphalt mixing and placement.

- In general, the incorporation of the waste offers two beneficial effects: at high temperatures, it increases G^* and decreases δ, enhancing rutting resistance; and at intermediate temperatures, it decreases δ, enhancing stiff asphalt binder fatigue resistance. This is in agreement with the decreasing $\tan(\delta)$ and increasing G' values, as studies have proven that this improves rutting resistance at high temperatures and fatigue resistance at intermediate temperatures.

- According to the Superpave rutting evaluation carried out in this study, the incorporation of the lignin-containing waste enhances asphalt binder rutting resistance. A higher waste content results in greater rutting resistance up to 20% waste. Therefore, asphalt binder with 20% of waste have shown the best rutting resistance.

- Consequently, the lignin-containing waste has the feasibility to be used as an extender and as well as an enhancer in asphalt binders of up to 20% waste.

- Directly adding 20% water waste from hardboard production into asphalt binders is a simple and efficient method to contribute to sustainable construction, decreasing the amount of asphalt binder required in asphalt mixtures. It is also a simple and effective method to minimize waste, since this waste from hardboard production will be valued without the need for subsequent transformations.

In this paper the lignin-containing waste analysed had a high water content. Further investigation will be considered to separate the water from the solid matter. Then the dry solid matter will be mixed with bitumen during the samples preparation.

Finally, it must be added that the present research was a first step within the development of asphalt binders with the lignin-containing waste from the production of hardboard. The results at laboratory level were encouraging and showed that the partial substitution of asphalt by the waste has a strong industrial potential. However, further investigation at full-scale is needed in order to accurately assess aspects, such as the actual on-site mechanical performance and the environmental, economic and social feasibility.

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