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Investigating the “circular propensity” of road bio-binders: effectiveness in hot recycling of reclaimed asphalt and recyclability potential

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Abstract

In pavement engineering, the use of bio-binders and reclaimed asphalt (RA) promotes the principles of sustainability and circular economy, without penalizing or even improving the performance. In this regard, this study focuses on the “circular propensity” of bio-binders obtained by partially replacing a conventional bitumen with a bio-oil generated as a residue by the wood and paper industries. Specifically, the objectives are: 1) to assess the effectiveness of bio-binders in the hot recycling of traditional RA and 2) to evaluate, in a long-term perspective, their recyclability potential. For this purpose, two severely aged binders (one “RAP” binder recovered from reclaimed asphalt and one laboratory-produced “Bio-RAP” binder) and two fresh binders (one bio-binder and one bitumen) are blended to reproduce four hot recycled binders. The mechanical behaviour and the aging susceptibility of these blends are compared to those of a control virgin bitumen. The experimental investigation includes conventional tests, rheological testing and modelling (modified CAM model) as well as chemical analysis (Fourier transform infrared spectroscopy). The main results indicate that the hot recycling of reclaimed bio-asphalt (bio-RA) may lead to mixtures less susceptible to cracking as compared to the recycling of conventional RA, as well as the use of bio-binders in the hot recycling of conventional RA may be beneficial in terms of cracking. Even though the blends with the bio-binder are characterized by a lower aging rate, the permanent deformation behaviour of all the recycled blends studied is comparable in unaged and short-term aged conditions, i.e. the circumstances under which rutting is usually a concern. Finally, the recycled blends show significantly lower aging susceptibility than the control bitumen. Overall, these results suggest that the bio-binders studied are effective in the hot recycling of RA and 100% recyclable, and their use in asphalt pavements can lead to significant technical and environmental benefits.

Key Words: Pavement engineering; asphalt; bio-binders; recycling; sustainability; circular economy.

1. Introduction

The current environmental issues have highlighted the worldwide need for a cleaner production, inspired by the principles of sustainability and circular economy. In fact, a sustainable and circular-economy-based production would allow to meet the needs of the present without compromising the ability of future generations to meet their own needs, by minimizing resources and energy consumption, wastes and emissions (Brundtland, 1987; Geissdoerfer et al., 2017).

Generally speaking, the construction sector is responsible for significant environmental impact, as it implies the consumption of natural resources (mostly non-renewable), causes emissions and produces wastes (Ghisellini et al., 2018; Hendrickson and Horvath, 2000; Kucukvar and Tatari, 2013). Therefore, it is truly necessary for the sector to undergo important changes in a relatively short time, such as the adoption of new construction technologies and innovative materials, aimed at promoting sustainability and circular economy without penalizing the performance.

Within this framework, the use of bio-binders is attracting more and more interest in pavement engineering (Ingrassia et al., 2019a). In general, bio-binders can be defined as binders in which petroleum-based bitumen is partially replaced with renewable bio-oils, which typically derive from residues or by-products like waste wood (Yang et al., 2014), non-edible vegetable biomass (Raouf and Williams, 2010), waste cooking oil (Sun et al., 2016) or animal manure (Fini et al., 2011). Previous studies have shown that, beside remarkable environmental benefits (Samieadel et al., 2018a), the use of bio-oils as partial replacement of bitumen may also provide important technical benefits such as reduced aging susceptibility (Hosseinnezhad et al., 2019; Ingrassia et al., 2019b), improved adhesion with aggregates (Bearsley and Haverkamp, 2007; Ingrassia et al., 2019c) and increased moisture damage resistance (Bearsley and Haverkamp, 2007; Ingrassia et al., 2019c), thanks to the bio-oils' chemical peculiarities.

At the same time, the progress in hot recycling technologies is allowing the production of new hot asphalt mixtures containing extremely high percentages of reclaimed asphalt (RA) without negatively affecting the performance (Zaumanis and Mallick, 2015; Zaumanis et al., 2014a, 2014b). RA, which derives from the milling of asphalt pavements at the end of their service life, is a very valuable material and, in hot recycling, the aged bitumen can be reactivated, at least partially (Lo Presti et al., 2019). The reuse of RA promotes the principles of sustainability and circular economy, by reducing the consumption of new non-renewable raw materials and the disposal of wastes, exactly as in the case of bio-binders.

The combined use of bio-binders and RA, both composed of used/end-of-life materials, may potentially result in sustainable asphalt mixtures with superior performance. Some bio-oils have been already studied as possible rejuvenators for RA (Behnood, 2019; Cavalli et al., 2018; Gong et al., 2016; Kowalski et al., 2017). However, to date, there are still uncertainties on the most suitable rejuvenator addition location in the asphalt plant, and some ad-hoc modification to the plant may also be necessary (Behnood, 2019; Lu et al., 2019; Zaumanis et al., 2019). A different employment for bio-oils in hot recycling might be to produce and store the bio-binder, by pre-blending bitumen and bio-oil. In this way, the issue of introducing the bio-based material in the production process could be easily overcome, provided that the stored bio-binder does not have problems in terms of storage stability, which is generally true (Abdullahi Ahmad et al., 2017; He et al., 2019; Ingrassia et al., 2019d), except in the presence of modifiers like polymers or crumb rubber.

At the same time, to ensure a wide market acceptance of bio-binders, it is essential – in a long-term perspective – to verify also if asphalt mixtures containing bio-binders can be 100% recycled at the end of their service life, as normally happens for traditional mixtures (Ingrassia et al., 2019a). Nevertheless, to date, no study has focused on the recyclability of bio-asphalt mixtures.

In this regard, this study has two main objectives: 1) to assess the effectiveness of bio-binders in the hot recycling of bitumen recovered from a typical RA, and 2) to evaluate, in a long-term perspective, the recyclability potential of bio-binders. For this purpose, two severely aged binders (one "RAP" binder recovered from reclaimed asphalt and one laboratory-produced "Bio-RAP" binder) and two fresh binders (one bio-binder and one bitumen) were blended to simulate four binders resulting from the hot recycling of asphalt. In order to investigate also their aging susceptibility, the blends were short-term and long-term aged in the laboratory. All binders were subjected to mechanical tests (including conventional tests as well as rheological testing and modelling) and chemical analysis (Fourier transform infrared spectroscopy).

2. Materials and methods

2.1 Base binders and control bitumen

In order to reproduce in the laboratory the binder resulting from the hot recycling of asphalt, two severely aged binders were considered. The first one, coded as "RAP", was recovered according to EN 12697-3 (2018) from a typical reclaimed asphalt, taken from a milled pavement at the end of its service life (more than 20 years). The second one, coded as "Bio-RAP", was artificially aged in the laboratory by subjecting the bio-binder A (described below) to one rolling thin film oven test (RTFOT) (EN 12607-1, 2015) and two consecutive pressure aging vessel (PAV) conditionings (EN 14769, 2013) (i.e. 40 hours PAV). Indeed, according to several studies (Bowers et al., 2014; Samieadel et al., 2018b; Zadshir et al., 2018), such procedure allows to prepare a reliable artificial

RA in the laboratory. This approach was necessary for producing RA deriving from a bio-binder (bio-RA), because no asphalt pavement containing bio-binders had been in service for a sufficiently long period of time when this study was carried out. It is worth noting that the RAP binder was harder, stiffer and more elastic than the Bio-RAP binder (Table 1 and Figure 1). These differences were properly considered in the materials preparation.

Moreover, the study included two different fresh binders to be combined with RAP and Bio-RAP: a bio-binder "A" composed of 10% by weight of a wood-based bio-oil, and a bitumen "B" having physical and mechanical properties similar to A (Table 1 and Figure 1), both characterized by a penetration of 120 dmm. As for the bio-oil used, it is a by-product of the wood and paper industries, and its physical and chemical properties can be found elsewhere (Ingrassia et al., 2019d).

In addition, a bitumen, coded as "Control" and having intermediate mechanical properties between the aged binders (RAP and Bio-RAP) and the fresh ones (A and B) (Table 1 and Figure 1), was added to the investigation as a commonly used virgin bitumen, with the aim of comparing its mechanical behaviour and aging susceptibility with those of the recycled blends.

Table 1 – Penetration and softening point of the base binders and the Control bitumen

Binder	Penetration [dmm]	Softening point [°C]
RAP	19	65.8
Bio-RAP	27	61.6
A	120	45.3
B	120	45.0
Control	55	52.4

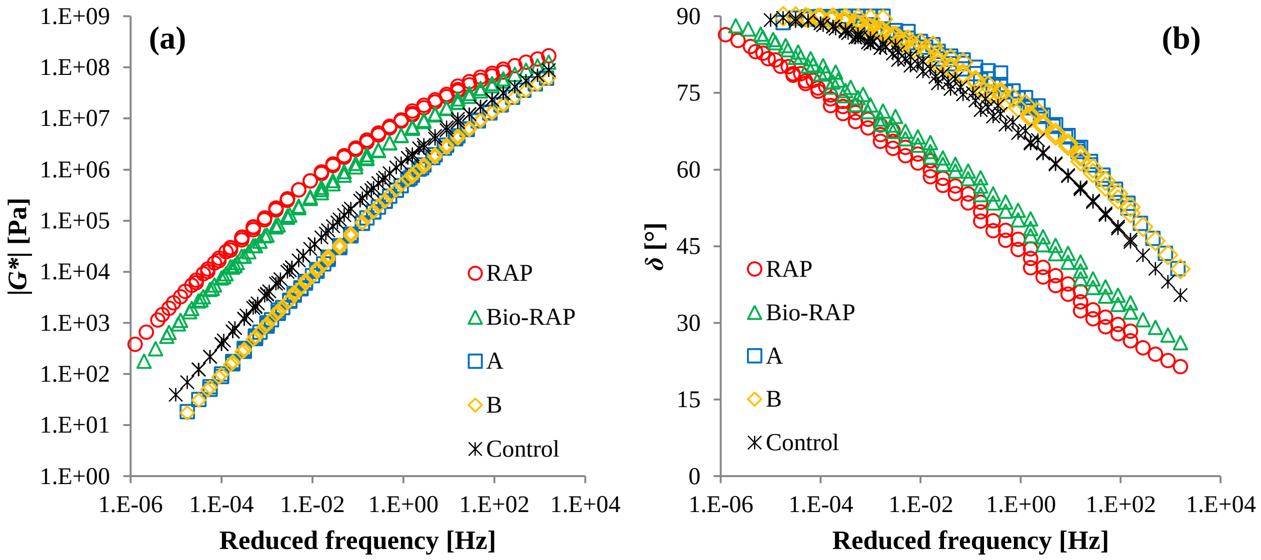


Figure 1 – Master curves of (a) complex modulus and (b) phase angle for the base binders and the Control bitumen, at a reference temperature of 20 °C

2.2 Recycled blends

The base binders (RAP, Bio-RAP, A and B) were combined according to appropriate proportions to prepare four blends (Table 2) having physical-mechanical properties similar to each other and comparable to the Control bitumen (see Section 3). After pre-heating the base binders at 160 °C (similarly to the temperatures usually adopted for hot recycling), the blends were produced by means of a laboratory mixer, considering a mixing speed of 500 rpm for 10 min.

A brief description of the recycled blends is provided in Table 2. Among the blends produced, B+RAP can be considered a sort of reference, as it represents the binder of a typical hot recycled asphalt mixture. The blend A+bio-RAP allows to evaluate, in a long-term perspective, the actual bio-binders' recyclability potential, whereas the blend A+RAP is useful to assess the effectiveness of bio-binders in hot recycling of bitumen. Finally, the blend B+bio-RAP represents an intermediate situation, opposite to that of A+RAP.

Table 2 – Recycled blends produced

Blend	Composition (by weight)	Description
A+RAP	71% A + 29% RAP	Recycled binder formed by a severely aged bitumen blended with a fresh bio-binder
B+RAP	71% B + 29% RAP	Recycled binder in which the aged and the fresh binders are composed of 100% bitumen
A+bio-RAP	62% A + 38% Bio-RAP	Recycled binder in which the aged and the fresh binders are bio-based (both contain 10% by weight of wood bio-oil)
B+bio-RAP	62% B + 38% Bio-RAP	Recycled binder formed by a severely aged bio-binder blended with a fresh bitumen

It is worth noting that the blends studied (Table 2) reproduce the binders of recycled asphalt mixtures with high reclaimed asphalt content. Indeed, the correspondence between the composition of the blends shown in Table 2 and the RA content in the recycled mixture can be evaluated through Equation (1) (Jiménez del Barco Carrión et al., 2017).

$$RA\ in\ the\ recycled\ mixture\ (\%) = 100 \cdot \frac{RVB \cdot DB\ content}{RAb\ content \cdot DOB} \tag{1}$$

where *RVB* is the replaced virgin binder, i.e. the percentage of fresh binder in the recycled mixture that is replaced by the RA binder (29% for A+RAP and B+RAP and 38% for A+bio-RAP and B+bio-RAP); *DB content* is the designed binder content in the recycled mixture, given by fresh binder plus reactivated RA binder (fixed equal to a typical value of 5% in the calculation); *RAb content* is the binder content in the RA (fixed equal to a typical value of 4% in the calculation); *DOB* is the degree of blending between the RA binder and the fresh binder (assumed equal to 70%, i.e. partial blending, according to Jiménez del Barco Carrión et al., 2017; Shirodkar et al., 2011; Stimilli et al., 2015). Consequently, the blends A+RAP and B+RAP represent mixtures containing about 50% RA by weight, while the blends A+bio-RAP and B+bio-RAP are representative of mixtures containing about 70% RA by weight.

The four blends (Table 2) were short-term (RTFOT) and long-term (PAV) aged, in order to reproduce the binder properties immediately after laying of hot recycled asphalt and after 10-15 year-service life of hot recycled asphalt, respectively.

Therefore, nineteen binders were studied overall, including the four base binders, the four blends at three different aging levels as well as the Control bitumen at three aging levels.

2.3 Mechanical tests

The mechanical properties of the binders were investigated through conventional and rheological tests.

The conventional properties, i.e. penetration and softening point, were assessed according to EN 1426 (2015) and EN 1427 (2015), respectively.

The rheological properties were studied through frequency sweep tests at different temperatures with a dynamic shear rheometer (DSR) in plate-plate configuration using 8 mm and 25 mm geometry, according to EN 14770 (2012). The norm of the complex modulus $|G^*|$ and the phase angle δ were determined at temperatures ranging from 0 to 80 °C with a step of 10 °C, increasing the frequency from 1 to 100 rad/s (i.e. from 0.159 to 15.9 Hz) with a fixed logarithmic step. All tests were carried out at a low shear strain of 0.05%, in order to study the behaviour of the binders in the linear viscoelastic (LVE) domain. At least two specimens were tested for each binder.

The master curves of $|G^*|$ and δ were developed by shifting the experimental data in the frequency domain at 20 °C, which was chosen as reference temperature (T_{ref}). The temperature-dependency of the shift factors was modelled according to the Williams-Landel-Ferry (WLF) law (Williams et al., 1955), as in Equation (2).

$$\log a_T(T) = -\frac{C_1\left(T - T_{ref}\right)}{C_2 + \left(T - T_{ref}\right)} \tag{2}$$

where $a_T(T)$ is the shift factor at temperature T, while C_1 and C_2 are empirical constants.

The complex modulus master curve was then modelled through the modified Christensen-Anderson-Marasteanu (CAM) model (Bahia et al., 2001), defined as in Equation (3) in the case of bituminous binders.

$$|\,G^*(f)| = \frac{G_g}{\left[1+\left(\frac{f_c}{f}\right)^k\right]^{\frac{m_e}{k}}} \tag{3}$$

where f is the reduced frequency, G_g is the glassy modulus (i.e. the value of the complex modulus for $f \to \infty$), f_c is the crossover frequency (i.e. the frequency at which the storage modulus G_1 and the loss modulus G_2 are approximately equal), k and m_e are dimensionless shape parameters. The distance between G_g and $|G^*(f_c)|$ is the so-called rheological index R, whose expression is as in Equation (4) in the case of bituminous binders.

$$R = \frac{m_e}{k}\log 2 \tag{4}$$

In the modelling procedure, G_g was fixed equal to 10^9 Pa based on literature suggestions (Anderson et al., 1994), whereas the parameters f_c, k and m_e (and consequently R) were determined by minimizing the error between model and experimental data, with the aim of achieving the best fitting.

In addition, the rheological behaviour at typical high service temperatures (60, 70 and 80 °C) was further investigated in terms of permanent deformation resistance through multiple stress creep and recovery (MSCR) tests, performed with the DSR with the 25 mm plate-plate geometry, according to EN 16659 (2015). At each testing temperature, two stress levels were considered, 0.1 and 3.2 kPa. The single stress level involved the application of 10 creep and recovery cycles, each one consisting in 1 s of loading and 9 s of recovery (without any load). The non-recoverable creep compliance J_{nr} and the percent strain recovery $\%R$ were determined for each binder, by testing at least two specimens.

2.4 Chemical analysis

The chemical characteristics of the binders were investigated through Fourier transform infrared (FTIR) spectroscopy in transmission mode (Marsac et al., 2014). The specimens were prepared by dissolving the binder in chloroform ($CHCl_3$). This solution was spread on sodium chloride (NaCl) plates and then the solvent was evaporated, in order to have a very thin binder film to be analysed. The transmittance/absorbance was evaluated at ambient temperature for wavenumbers between 500 and 4000 cm^{-1} with a resolution of 4 cm^{-1}, and each spectrum was obtained as the average of 16 consecutive scans on the specimen. For each binder, at least three specimens were tested and then the average spectrum was determined. The spectral analysis was carried out with an integration method consisting in the calculation of the areas under the most significant peaks by means of a tangential approach (Hofko et al., 2017).

3. Results and analyses

3.1 Conventional properties

The penetration and the softening point data are summarized in Table 3. It can be observed that the conventional properties of the four recycled blends are very similar at all aging levels. In terms of penetration and softening point, the Control bitumen seems slightly harder than the recycled blends in unaged conditions, but this difference tends to become smaller as aging increases. In general, the results in Table 3 indicate that the recycled blends and the Control bitumen are characterized by similar grade, which makes their comparison reasonable. The effect of aging emerges, as expected, as a penetration decrease and softening point increase due to the binder hardening.

Nevertheless, it is worth noting that a more reliable comparison between the binders in terms of mechanical behaviour as well as aging susceptibility can be made by considering the results of the rheological and chemical tests, which are presented in the following Sections.

Table 3 – Penetration and softening point of the recycled blends and the Control bitumen

Aging level	Binder	Penetration [dmm]	Softening point [°C]
Unaged	A+RAP	64	50.1
	B+RAP	65	50.2
	A+bio-RAP	60	50.4
	B+bio-RAP	61	50.2
	Control	55	52.4
RTFOT	A+RAP	47	55.7
	B+RAP	43	54.8
	A+bio-RAP	45	54.7
	B+bio-RAP	47	55.4
	Control	35	55.7
PAV	A+RAP	25	61.6
	B+RAP	25	62.5
	A+bio-RAP	24	62.2
	B+bio-RAP	28	62.9
	Control	25	63.0

3.2 Master curves

Figures 2, 3 and 4 show the master curves of complex modulus $|G^*|$ and phase angle δ (experimental data) for the recycled blends and the Control bitumen in unaged, short-term aged and long-term aged conditions, respectively. In general, the rheological behaviour of the binders is comparable at all aging levels. From the comparison between Figures 2, 3 and 4, it can be observed that for all binders the effect of aging results in increased complex modulus and decreased phase angle, meaning that the stiffness and the elasticity of the binders increase with aging (as expected). It is also important to notice that the Control bitumen seems the binder that undergoes the most significant rheological changes due to the aging process ($|G^*|$ increase and δ reduction). This finding is in contrast with what emerged from the conventional tests, suggesting that the latter may not be sufficiently reliable to examine the mechanical behaviour and aging susceptibility of bituminous binders. It should be noted also that all binders are thermo-rheologically simple (including the ones that contain the bio-oil), as perfectly continuous $|G^*|$ and δ master curves can be obtained by using the same shift factors (Yusoff et al., 2011).

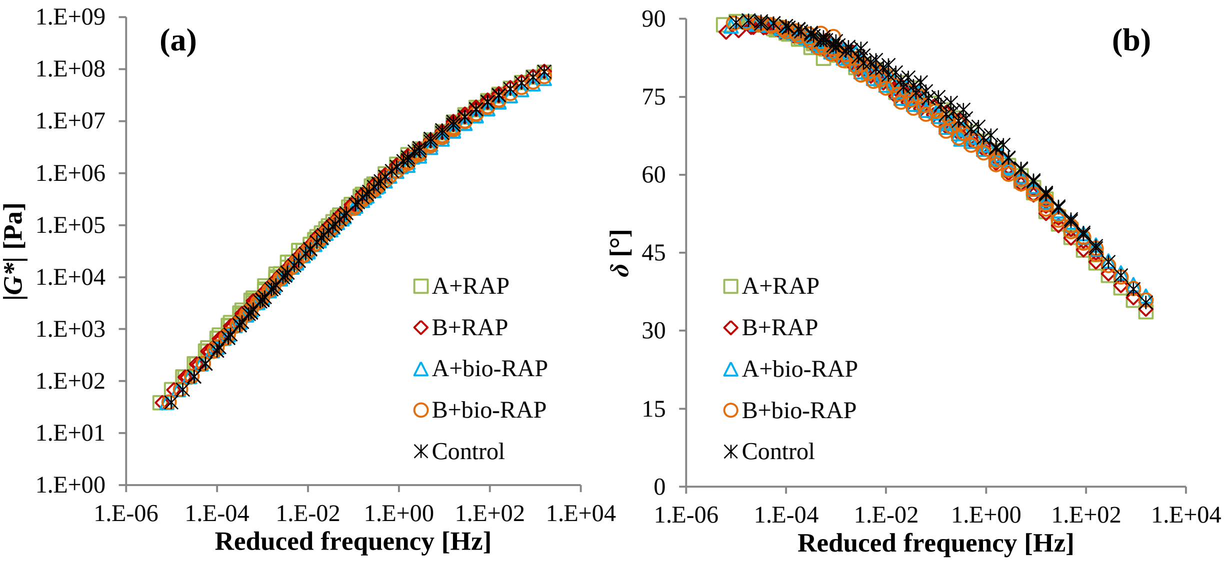


Figure 2 – Master curves of (a) complex modulus and (b) phase angle for the unaged recycled blends and Control bitumen, at a reference temperature of 20 °C

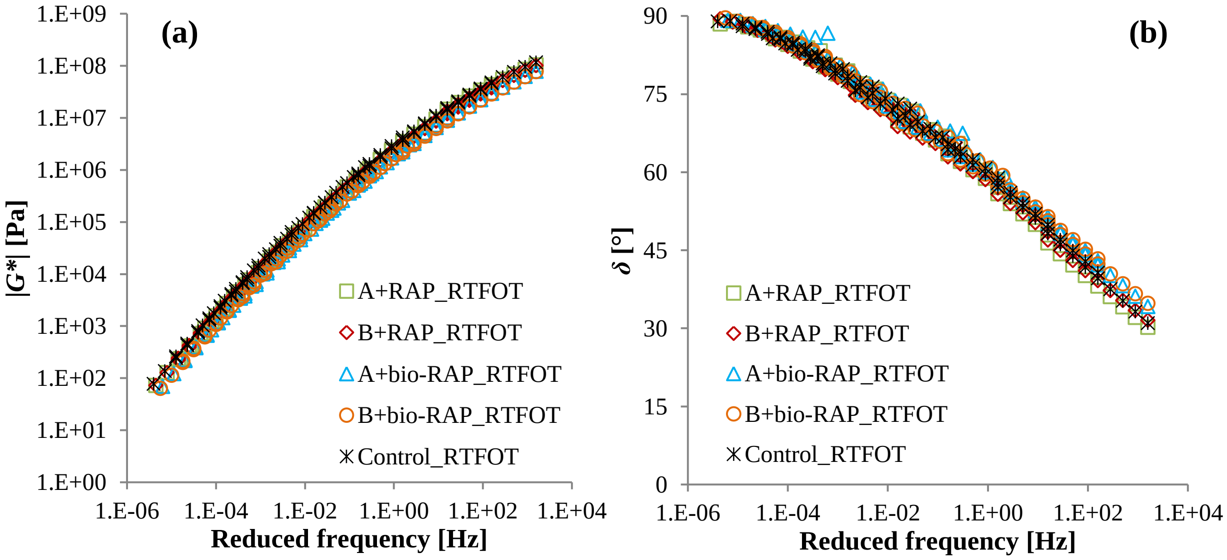


Figure 3 – Master curves of (a) complex modulus and (b) phase angle for the short-term aged recycled blends and Control bitumen, at a reference temperature of 20 °C

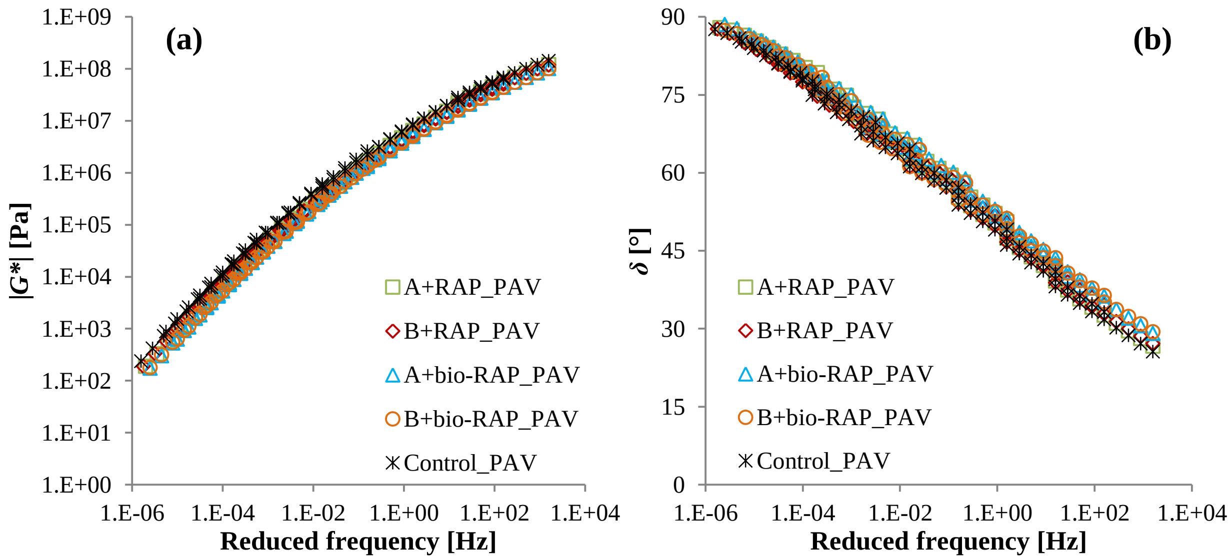


Figure 4 – Master curves of (a) complex modulus and (b) phase angle for the long-term aged recycled blends and Control bitumen, at a reference temperature of 20 °C

In order to better compare the rheological behaviour of the binders, the parameters deriving from the modelling of the complex modulus master curves can be considered. As an example of the model fitting to the experimental data, Figure 5 shows the $|G^*|$ master curve for A+bio-RAP at different aging levels (the other binders are not shown for brevity). The figure depicts more clearly the increase of $|G^*|$ due to aging.

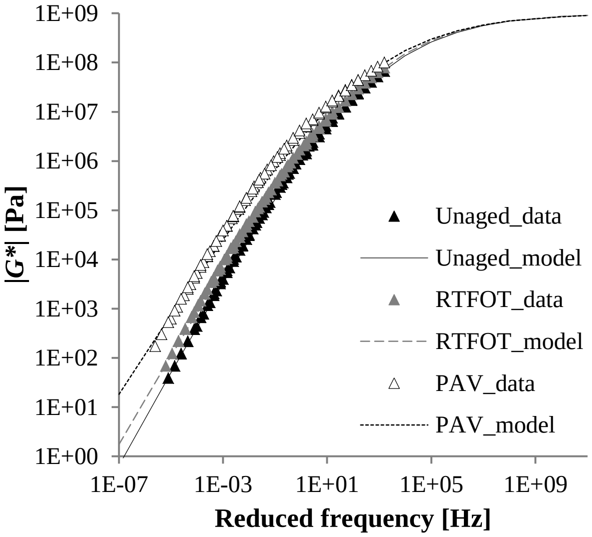


Figure 5 – Complex modulus master curve at 20 °C for A+bio-RAP at different aging levels (experimental data and modified CAM model)

Table 4 summarizes the modified CAM and WLF parameters obtained from the modelling. In terms of modified CAM parameters, aging causes a general reduction of f_c (which does not emerge, however, after RTFOT) and m_e as well as a slight decrease of k. Consequently, R decreases with aging as well. Based on the physical meaning of these parameters (Bahia et al., 2001), the reduction of the crossover frequency f_c indicates greater elastic component in the binder behaviour, while the decrement of the rheological index R denotes a more rapid transition from the elastic to the viscous behaviour (in fact, a hypothetical R value equal to zero would indicate a sudden transition between perfectly elastic and purely viscous behaviour). As for the WLF parameters, greater C_1 and C_2 values are observed for higher aging levels, suggesting a reduced molecular mobility. All these aging-related trends are in line with previous findings by Mazzoni et al. (2018), who studied the influence of different rejuvenators on the aging of hot recycled asphalt binders. It is worth noting that the variation of the parameters due to aging is more evident in the case of the Control bitumen, denoting a possible greater aging susceptibility (in accordance with the observations made from the comparison between Figures 2, 3 and 4), once again in contrast with the outcomes of the conventional tests.

Since the RAP binder is stiffer and more elastic than the Bio-RAP binder (Figure 1), it exhibits lower f_c, m_e and R as well as higher C_1 and C_2 than Bio-RAP. Conversely, the parameters are almost the same for the bio-binder A and the conventional bitumen B, as their rheological behaviour is practically identical (Figure 1). When the same aging level is considered, in general, all the recycled blends and the Control bitumen exhibit comparable modified CAM and WLF parameters, confirming the similarity of the master curves noticed from Figures 2, 3 and 4. More in detail, for the blends containing Bio-RAP as aged binder, R tends to be higher, while C_1 and C_2 tend to be lower, as compared to the other blends and the Control bitumen. Finally, f_c seems to be mainly affected by the long-term aging rather than by the material type.

Table 4 – Modified CAM and WLF parameters

Binder	f_c [Hz]	k	m_e	R	C_1	C_2
RAP	200	0.21	0.75	1.08	39	382
Bio-RAP	300	0.21	0.79	1.14	31	307
A	400	0.22	1.06	1.48	16	181
B	400	0.21	1.06	1.51	15	171
A+RAP	350	0.22	0.95	1.33	22	228
A+RAP_RTFOT	350	0.21	0.89	1.28	24	248
A+RAP_PAV	250	0.20	0.80	1.20	32	323
B+RAP	350	0.22	0.95	1.30	21	220
B+RAP_RTFOT	350	0.21	0.89	1.25	24	248
B+RAP_PAV	250	0.20	0.80	1.20	35	348
A+bio-RAP	350	0.20	0.96	1.44	20	215
A+bio-RAP_RTFOT	350	0.20	0.92	1.38	22	239
A+bio-RAP_PAV	250	0.19	0.82	1.30	30	303
B+bio-RAP	350	0.21	0.96	1.41	19	206
B+bio-RAP_RTFOT	350	0.20	0.90	1.35	23	242
B+bio-RAP_PAV	250	0.19	0.82	1.30	30	303
Control	350	0.22	0.97	1.33	19	203
Control_RTFOT	350	0.21	0.89	1.20	25	257
Control_PAV	250	0.21	0.78	1.11	35	344

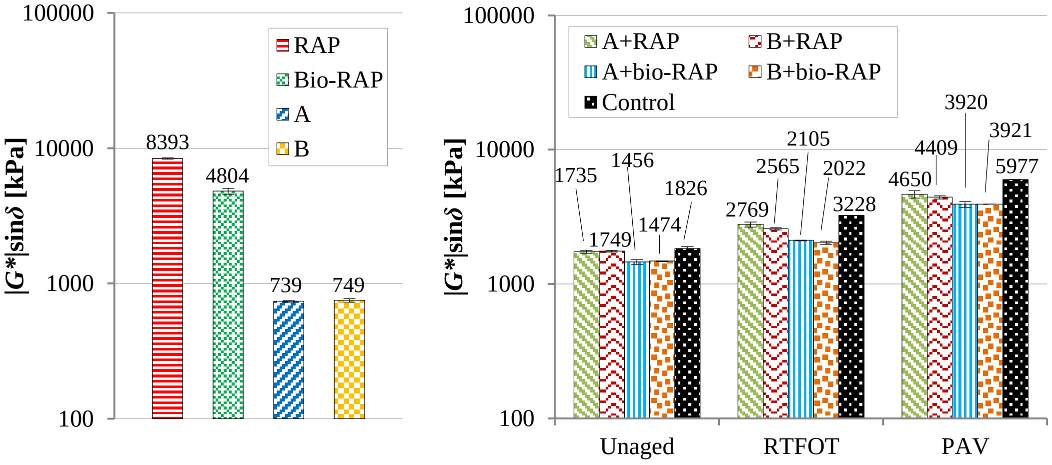
3.3 Fatigue parameter

In order to evaluate the fatigue behaviour of the binders, the rheological data were elaborated to determine the Superpave fatigue parameter $|G^*|\sin\delta$ introduced by the Strategic Highway Research Program (SHRP) (Kennedy et al., 1994). The parameter was calculated at the reference frequency of 10 rad/s (1.59 Hz) and at 20 °C, which was selected as a representative intermediate service temperature (Figure 6). In general, low values of $|G^*|\sin\delta$ indicate a good resistance of the binder to fatigue cracking, and a maximum value of 5000 kPa is set for $|G^*|\sin\delta$ after PAV aging.

From Figure 6a, it can be observed that the fresh binders A and B exhibit almost identical values of $|G^*|\sin\delta$, which are relatively low due to their high penetration grade (see Table 1). Moreover, as expected, the RAP binder shows a higher $|G^*|\sin\delta$ value as compared to the Bio-RAP binder.

As for the comparison between the recycled blends, Figure 6b shows that, at all aging levels, the blends containing Bio-RAP as aged binder are characterized by lower values of $|G^*|\sin\delta$ with respect to the blends containing RAP. Instead, the effect of the fresh binder type (A or B) is not very evident. Moreover, the Control bitumen exhibits higher $|G^*|\sin\delta$ values than the recycled blends, especially after aging, suggesting a possible faster aging rate (it is the only binder that does not meet the limit of 5000 kPa after PAV). These observations are generally in line with the results of rheological testing and modelling presented in Section 3.2.

The meaning of these results in terms of performance is discussed more in depth after the analysis of the aging susceptibility of the binders in Section 3.6.



(a) **(b)**

Figure 6 – Fatigue parameter at 20 °C and 10 rad/s for (a) base binders, (b) recycled blends and Control bitumen at different aging levels

3.4 Non-recoverable creep compliance and percent strain recovery

The values of the non-recoverable creep compliance (J_{nr}) obtained at 70 °C and 3.2 kPa are shown in Figure 7, whereas Figure 8 displays the values of the percent strain recovery (*%R*) calculated at the same temperature and stress level.

Firstly, it can be noted that – as expected – aging causes a reduction of J_{nr} and a simultaneous increase of *%R*. In fact, as already discussed in Section 3.2, the binder becomes stiffer and more elastic with aging, thus exhibiting lower deformability and higher recovery capability. In general, lower J_{nr} and higher *%R* indicate a greater resistance against permanent deformations.

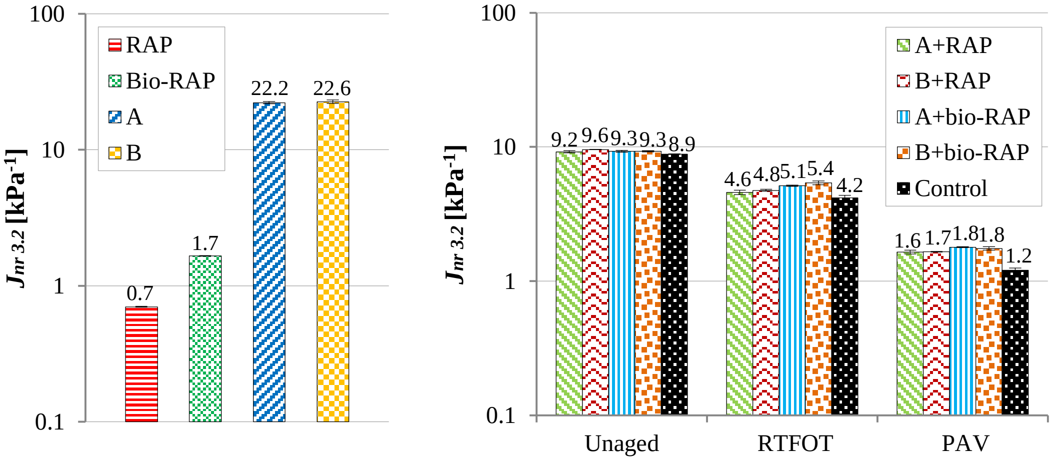
As for the base binders (Figures 7a and 8a), the bio-binder A and the bitumen B are characterized by identical values of J_{nr} and *%R* (equal to zero), further confirming that their mechanical behaviour is very similar (Table 1 and Figure 1). Instead, the RAP binder exhibits lower J_{nr} and higher *%R* as compared to the Bio-RAP binder, in line with what observed in terms of conventional properties and master curves (Table 1 and Figure 1).

Figures 7b and 8b show that all the recycled blends and the Control bitumen have comparable J_{nr} and *%R* values in unaged conditions. Some differences between the binders emerge with aging, once more suggesting that the binders may have dissimilar aging susceptibility. Specifically, after aging, the blends containing Bio-RAP tend to exhibit the highest J_{nr} values together with the lowest

%R, while the Control bitumen, on the contrary, is characterized by the lowest J_{nr} values and the highest *%R*. The effect of the fresh binder type in the blend can be mainly observed in the percent strain recovery after PAV (Figure 8b). In fact, the blends containing A (A+RAP, A+bio-RAP) present smaller *%R* values than the corresponding blends containing B (B+RAP, B+bio-RAP). All these observations are consistent with the results presented in Sections 3.2 and 3.3.

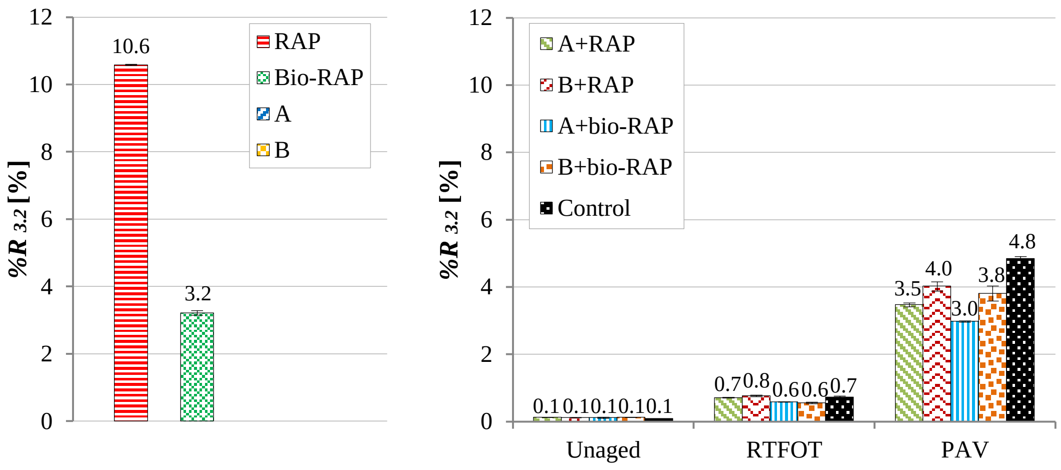
Analogous observations can be made at the other temperatures (60 and 80 °C) and stress levels (0.1 kPa) investigated (not shown for brevity).

The implications of these outcomes in terms of performance are further discussed in Section 3.6, after the in-depth analysis of the aging susceptibility of the binders.



(a) **(b)**

Figure 7 – Non-recoverable creep compliance at 70 °C and 3.2 kPa for (a) base binders, (b) recycled blends and Control bitumen at different aging levels



(a) **(b)**

Figure 8 – Percent strain recovery at 70 °C and 3.2 kPa for (a) base binders, (b) recycled blends and Control bitumen at different aging levels

3.5 FTIR spectra

The absorbance spectra of the base binders are shown in Figure 9, where the main peaks are highlighted. From the spectra of binders A and Bio-RAP, it can be observed that the bio-oil modification is identified by the peak at 1735 cm^{-1}, which is absent in the spectrum of conventional bitumen (e.g. binders B and RAP) and corresponds to the C=O stretch of the bio-oil's esters, as already shown in previous studies (Ingrassia et al., 2019b, 2019d). Moreover, for all the binders, the effect of aging emerges as the formation of new bonds at about 1700 and 1030 cm^{-1} (see the comparison between bio-based binders A and Bio-RAP and between conventional bitumens B and RAP), corresponding respectively to the C=O stretch of carbonyl functional groups and the S=O stretch of sulfoxides. It is well known that the C=O and S=O bonds formed during aging at these wavenumbers are responsible for significant physical and rheological changes undergone by bituminous binders (Petersen, 2009; Petersen and Glaser, 2011). However, a previous study (Ingrassia et al., 2019b) highlighted that, for the same type of bio-binders, aging can imply also the formation of new compounds that show absorption at 1735 cm^{-1}.

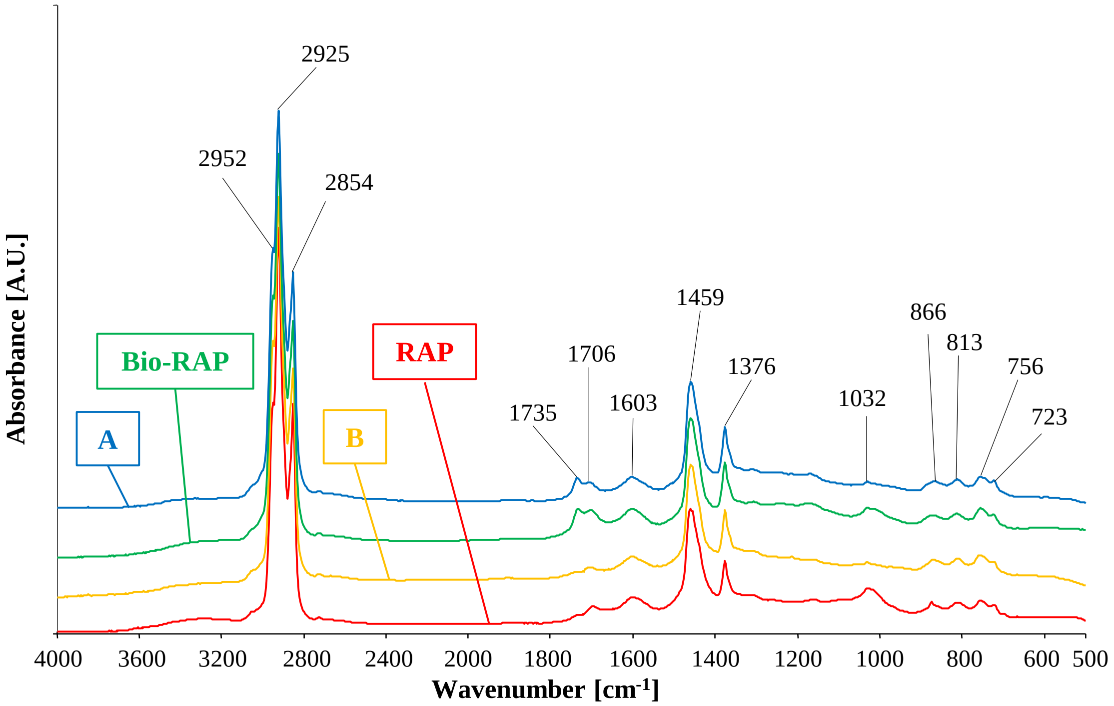


Figure 9 – FTIR spectra of the base binders

No additional peaks were observed in the spectra of the recycled blends, indicating that in all cases only a physical blending occurred between the binders, without any chemical reaction (not even in the presence of the bio-binder). Therefore, in the quantitative analysis, the area between 1679 and 1753 cm^{-1} (A_{CO}) and the area between 972 and 1063 cm^{-1} (A_{SO}) were calculated. It should be pointed out that both peaks at 1700 and 1735 cm^{-1} were included in the calculation of A_{CO} because of the peculiar shape of the bio-binder spectrum in this wavenumber range (see Figure 9), which does not allow to consider these peaks as completely distinct and independent from each other. In order to minimise the influence of specimen thickness (or infrared radiation path length), these areas were normalised as in Equations (5) and (6), where A_{ref} is the reference area between 1330 and 1518 cm^{-1} including the peaks at 1376 and 1459 cm^{-1}, which correspond to the CH_3 and CH_2 bend of the aliphatic groups (that are supposed not to be affected by oxidation) (Marsac et al., 2014).

$$I_{CO} = \frac{A_{CO}}{A_{ref}} \tag{5}$$

$$I_{SO} = \frac{A_{SO}}{A_{ref}} \tag{6}$$

The indices I_{CO} and I_{SO} calculated for the recycled blends and the Control bitumen are shown in Figures 10a and 10b, respectively, whereas their sum is shown in Figure 10c. It should be noted that, in Figure 10, each graph presents a different scale on the y-axis to emphasize the differences between the binders.

As can be seen from Figure 10a, the index I_{CO} increases with aging for all binders, and such increment seems to be almost linear. Moreover, it can be noted that the blends containing the bio-oil exhibit higher I_{CO} values due to the peak at 1735 cm^{-1}. Indeed, the highest values are shown by A+bio-RAP, followed by A+RAP and B+bio-RAP, whereas B+RAP and the Control bitumen exhibit significantly lower I_{CO} values.

Conversely, the index I_{SO} is almost unchanged after RTFOT, while it markedly increases after long-term aging (PAV) (Figure 10b). In general, higher I_{SO} values are observed in the presence of RAP (A+RAP and B+RAP), as this binder was recovered from the reclaimed asphalt deriving from a pavement which was in service for more than 20 years (see Section 2.1). Furthermore, since the Control bitumen is entirely virgin (unlike the recycled blends), it exhibits the lowest I_{SO} values, especially in unaged and short-term aged conditions.

Finally, the sum of I_{CO} and I_{SO} seems to be a reliable chemical aging indicator, as it increases with aging for all binders (Figure 10c). Specifically, this increment is relatively small after RTFOT, whereas it becomes larger after PAV, which seems consistent with the severity of the mechanical changes typically undergone by bituminous binders after short-term and long-term aging. A similar FTIR index was effectively used in a previous study to compare the aging susceptibility of this type of bio-binders and conventional bitumens (Ingrassia et al., 2019b).

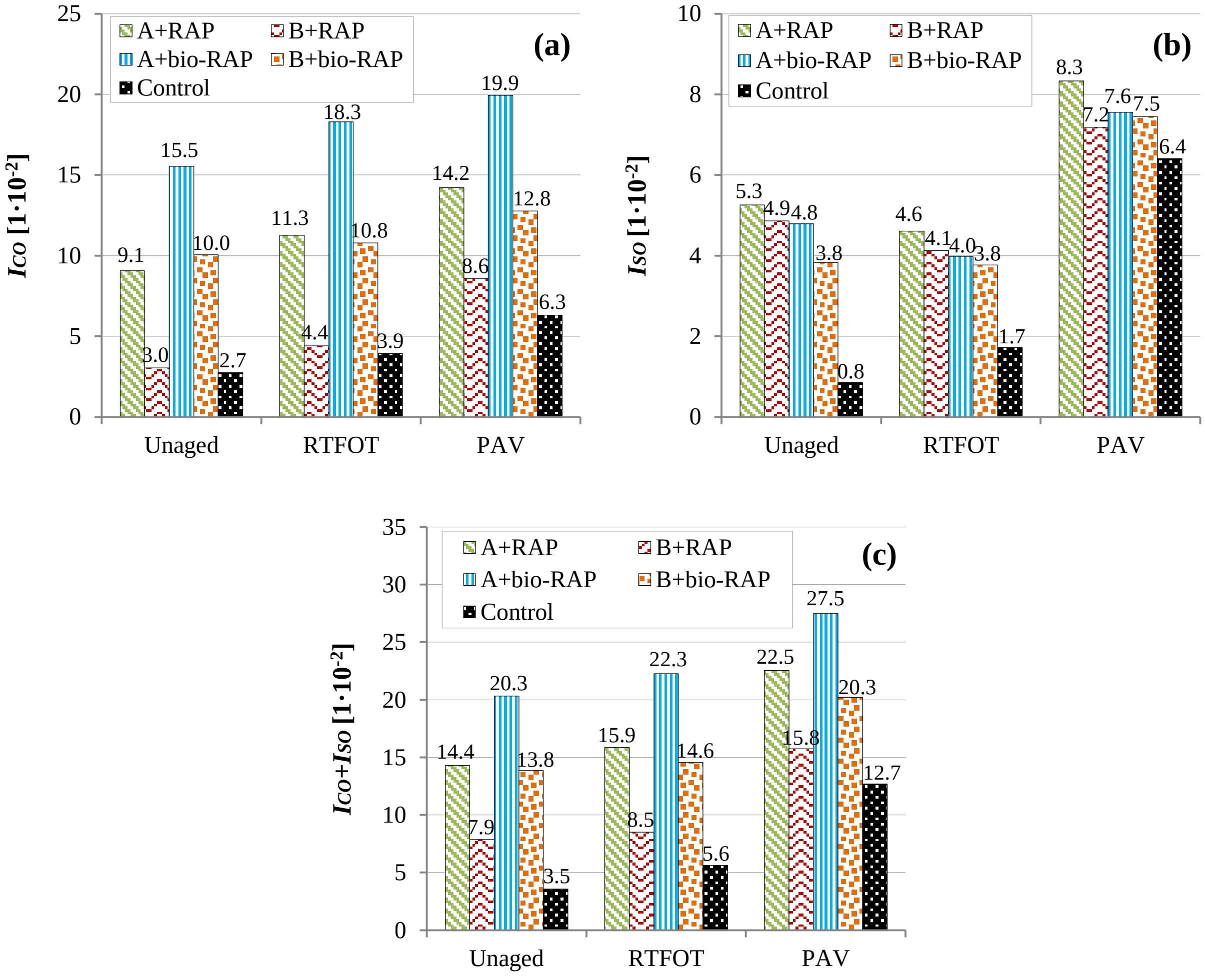


Figure 10 – FTIR indices: (a) I_{CO}, (b) I_{SO}, (c) I_{CO}+I_{SO}

3.6 Aging susceptibility

In order to compare the aging susceptibility of the binders, one chemical and one mechanical parameter strictly related to the oxidation of the binders were considered.

From the chemical point of view, it is generally known that the FTIR spectrum is a sort of chemical fingerprint of the material (Smith, 2011). In Section 3.5, it has been shown that, for the binders studied, aging causes the formation of additional compounds at 1700, 1735 and 1030 cm^{-1}, quantified through the sum of the indices I_{CO} and I_{SO}.

Therefore, the chemical aging index AI_{FTIR}, defined as the ratio between I_{CO}+I_{SO} for the aged binder and I_{CO}+I_{SO} for the unaged binder (Equation (7)), was considered.

$$AI_{FTIR} = \frac{(I_{CO} + I_{SO})_{aged}}{(I_{CO} + I_{SO})_{unaged}} \quad (7)$$

From the mechanical standpoint, instead, different parameters are considered in literature to study the aging susceptibility of bituminous binders. Some of them are based on the results of conventional test (e.g. the retained penetration and the softening point increment), while others are more performance-based (i.e. calculated from the results of rheological testing and/or modelling). In general, a reliable mechanical aging index should be unambiguously related to the evolution of the binder oxidation. In Sections 3.2, 3.3 and 3.4, it has been shown that the modified CAM parameters m_e and R and the WLF parameters C_1 and C_2 exhibit a clear trend with aging, but their variation due to aging is relatively small as compared to that of the fatigue parameter $|G^*|\sin\delta$, the non-recoverable creep compliance J_{nr} and the percent strain recovery $\%R$. Moreover, it is worth pointing out that $|G^*|\sin\delta$, J_{nr} and $\%R$ are determined directly from rheological tests, whereas the modified CAM and WLF parameters are the results of a modelling procedure that might be affected by a certain degree of inaccuracy. In addition, the effect of oxidative aging can be more easily quantified at very low frequencies/very high temperatures (Rad et al., 2018). Therefore, based on these considerations, J_{nr} was considered as the most appropriate mechanical parameter to be used in the analysis of the aging susceptibility of the binders studied. Specifically, the values of J_{nr} determined at the highest stress level (3.2 kPa) and 70 °C were considered. This temperature was chosen among those investigated (60, 70 and 80 °C) because the recycled blends and the Control bitumen should have an upper performance grade (PG) close to 70 °C.

Since the non-recoverable creep compliance decreases with aging, the mechanical aging index AI_{MSCR} was defined as the ratio between J_{nr} of the unaged binder and J_{nr} of the aged binder (Equation (8)), in order to have AI values higher than 1 (as in the case of AI_{FTIR}).

$$AI_{MSCR} = \frac{(J_{nr}@70°C, 3.2\ kPa)_{unaged}}{(J_{nr}@70°C, 3.2\ kPa)_{aged}} \tag{8}$$

The values of AI_{FTIR} and AI_{MSCR} obtained are shown in Figures 11a and 11b, respectively. Firstly, it can be observed that, after short-term aging, no big differences emerge between the binders (except for AI_{FTIR} for the Control bitumen), probably because this aging condition is not severe enough to assess the aging susceptibility of the binders or all the studied binders are highly aging-resistant in RTFOT. On the contrary, after long-term aging, there is a clear distinction between the binders, which are ranked as follows (from the highest to the lowest aging susceptibility): Control bitumen, B+RAP, A+RAP, B+bio-RAP, A+bio-RAP. It should be noted that this ranking is observed both in terms of AI_{FTIR} and AI_{MSCR} after PAV.

These findings indicate that the aging susceptibility of the recycled blends is significantly lower than that of the Control bitumen, analogously to previous results by Mazzoni et al. (2018). As a possible explanation of this phenomenon, it should be considered that the recycled blends already contain a certain amount of severely aged binder, and the aging rate tends to be faster when the binder is virgin while it progressively slows down as aging develops (Luo et al., 2018). Moreover, the blends containing Bio-RAP as aged binder (A+bio-RAP and B+bio-RAP) have lower aging susceptibility than the blends containing RAP (A+RAP and B+RAP). At the same time, the blends containing the bio-binder A as fresh binder (A+RAP and A+bio-RAP) tend to exhibit lower aging susceptibility than the blends containing the conventional bitumen B (B+RAP, B+bio-RAP). These results are fully in agreement with previous findings by Ingrassia et al. (2019b), who observed that this type of bio-binders is less affected by aging as compared to conventional bitumens with similar penetration grade.

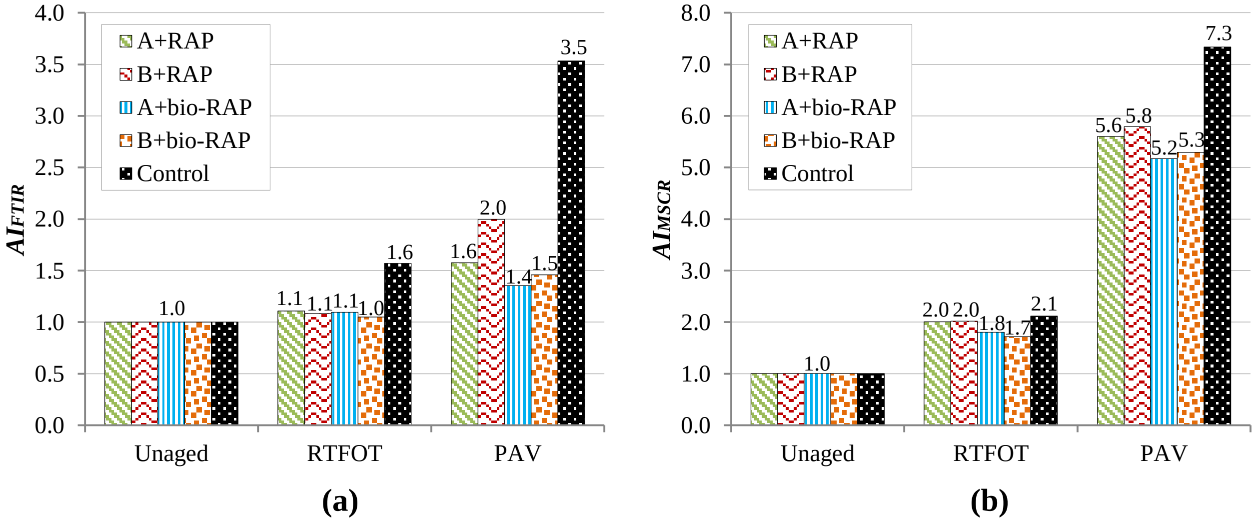


Figure 11 – Aging index based on (a) FTIR results, (b) MSCR results

Based on these outcomes and on the results provided in Sections 3.3 and 3.4, the hot recycling of bio-RA may lead to asphalt mixtures less susceptible to cracking as compared to the recycling of conventional RA, as well as the use of bio-binders in the hot recycling of conventional RA may be beneficial in terms of cracking, thanks to a lower aging rate of the binder. On the other hand, the reduced aging undergone by the blends with the bio-binder might imply a higher rutting tendency for the resulting asphalt mixture. Nevertheless, it should be emphasized that rutting is usually a concern when the binder is unaged or short-term aged, and under these circumstances the behaviour of all the recycled blends studied is broadly the same. Moreover, the results obtained suggest that mixtures containing reclaimed asphalt may be characterized by a lower aging rate as compared to virgin asphalt mixtures (with possible consequences in terms of mechanical behaviour).

In Figure 12, three pairs of values (AI_{FTIR}, AI_{MSCR}) are plotted for every binder. The pair at (1,1) is the origin of the graph, as it represents the unaged condition ("zero" condition) for all binders, intermediate (AI_{FTIR}, AI_{MSCR}) values represent the short-term aged condition, whereas the highest (AI_{FTIR}, AI_{MSCR}) values correspond to the long-term aged condition. The figure demonstrates that, if the binders are examined separately, there is an excellent linear relationship between AI_{MSCR} and AI_{FTIR} (R^2 very close to 1), indicating that the chemical and mechanical parameters considered as aging indicators ($I_{CO}+I_{SO}$ and J_{nr}) are effectively correlated with the binder oxidation.

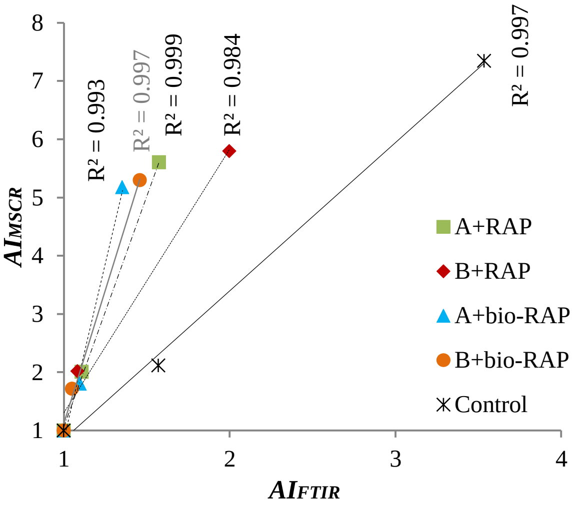


Figure 12 – Correlation between AI_{MSCR} and AI_{FTIR}

4. Conclusions

The objectives of this study were 1) to assess the effectiveness of bio-binders in the hot recycling of bitumen from typical reclaimed asphalt (RA), and 2) to evaluate the recyclability potential of bio-binders. Two severely aged binders (one "RAP" binder recovered from reclaimed asphalt and one laboratory-produced "Bio-RAP" binder) and two fresh binders (one bio-binder and one bitumen) were properly blended to simulate four binders resulting from the hot recycling of asphalt. The blends were then short-term and long-term aged in order to investigate their aging susceptibility. The mechanical behaviour and the aging susceptibility of the recycled blends were also compared to those of a common virgin bitumen (Control). The experimental investigation included mechanical tests (conventional tests as well as rheological testing and modelling) and chemical analysis (Fourier transform infrared spectroscopy).

The main conclusions can be summarized as follows:

- FTIR analysis highlights that, for all the binders tested (with and without the bio-binder), oxidative aging can be tracked from the chemical point of view by considering the formation of C=O bonds at 1700 and 1735 cm^{-1} plus S=O bonds at 1030 cm^{-1}.
- From the mechanical point of view, the aging susceptibility of the binders studied can be assessed through the evolution of the non-recoverable creep compliance, which is linearly correlated with the chemical oxidation for each binder.
- The recycled blends containing the bio-binder undergo lower aging than the corresponding blends containing the conventional bitumen.
- The hot recycling of reclaimed bio-asphalt (bio-RA) may lead to mixtures less susceptible to cracking as compared to the recycling of conventional RA, as well as the use of bio-binders in the hot recycling of conventional RA may be beneficial in terms of cracking, thanks to a lower aging rate of the binder.
- The reduced aging undergone by the blends with the bio-binder might imply a higher rutting tendency for the resulting asphalt mixture. However, the behaviour of all the recycled blends studied is generally comparable in unaged and short-term aged conditions, i.e. when rutting is usually a concern.
- The recycled blends show significantly lower aging susceptibility than the Control bitumen (virgin), probably because they already contain a certain percentage of severely aged binder.

Overall, these results suggest that bio-binders can be considered effective in the hot recycling of RA and 100% recyclable, and their use in asphalt pavements can lead to significant technical as well as environmental benefits. However, the results of this investigation should be integrated also with the study of other aspects, such as low-temperature performance and water sensitivity.

Future work will focus on the performance-based characterization of bio-asphalt mixtures and corresponding traditional asphalt mixtures.

Conflict of interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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