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Restoration of physical properties on an aged crumb rubber modified bitumen adding a bio-based recycling agent

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ABSTRACT

Reclaimed asphalt pavement (RAP) is a popular practice in the pavement industry due to environmental and economic benefits. But, RAP must be processed and treated before being reused in new pavements for preventing premature failures. A widely adopted strategy consists of adding recycling agents, and specifically rejuvenators, to mitigate the aging of the RAP binder. From the circular economy perspective, also crumb rubber (CR) from end-of-life tyres have been used to prepare binders with enhanced properties. The actual possibility of using the RAP derived from removed and/or reprocessed pavements containing CR particles (CR-RAP) for the formulation of new suitable mixtures is still a little-explored topic. Moreover, the available rejuvenators tailored on RAP have not yet evidence of effectiveness in producing asphalt mixtures containing CR-RAP. For this purpose, unaged, aged and rejuvenated blends with and without CR were studied through a rheological characterisation to evaluate the potential restoration of their physical properties when a bio-based recycling agent is added. The results of viscosity, frequency sweep, creep-recovery and multiple stress creep recovery (MSCR) tests have highlighted on the one hand the significant contribution of CR in reducing the accelerated aging effects of the binder, on the hand the effectiveness of the rejuvenator in restoring, more clearly at low temperatures, the rheological behaviour of the binders (above all for CRMB) similar to the unaged condition.

1. Introduction

Asphalt pavements deteriorate after some years of use and, therefore, they must be removed from the road. The so-called reclaimed asphalt pavement (RAP) is the term given to the removed and/or reprocessed pavement materials containing bitumen and aggregates. After the milling or extraction, the RAP is processed using a series of operations which includes crushing, screening, conveying and stacking [1]. RAP is adopted in the construction of new pavements as our society is demanding a more efficient and economic use of energy and natural resources due to growing concerns over global warming and climate change emergency [2,3]. The main environmental benefits of reusing RAP is the decrease in the amount of waste construction materials, the reduction in energy consumption

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and emissions in the production of raw materials (both binder and aggregates) and the savings related to the extraction and transportation of virgin materials [4,5]. On the other hand, there are also economic advantages: costs are reduced especially those related to asphalt binders, which is the biggest cost item in the production of asphalt mixtures. It has been estimated that by using 100% RAP the costs can be reduced by 50–70% [6,7]. Due to the desire to find a more sustainable way of paving, the pavement sector is developing and implementing new technologies to improve and maximise the recycling of pavements. Italian experiences on the primary road network have scientifically evaluated in-plant experimental productions for high RAP contents with particularly satisfactory outcomes of the detected performances [8,9].

However, RAP must be processed and treated before being reused in the construction of a new pavement. One of the most difficult challenges is the use of increased amounts of recycled materials [10]. It is known that the recycled binders are significantly stiffer than the virgin binders as they are highly aged and this does not allow to incorporate a large amount of RAP [11], as this might cause premature distresses in the pavement such as low temperature cracking and fatigue failure [12–14]. The stiffness of the RAP binder depends on the original binder, the climatic conditions to which the pavement has been exposed and the period between the road construction and the pavement reclaiming. Also, RAP binder has high viscosity and oxidation compared to a virgin binder [15]. Nonetheless, recycling agents, and specifically rejuvenators, have been widely adopted to overcome these issues and to mitigate the problems associated with the use of aged binders [16–19]. This is a common practice to restore RAP binders' properties (physical, rheological, chemical) to a condition similar to that of a virgin binder [20–23]. When a recycled agent is added, it is expected that the molecular mobility improves as some proportions of lighter oils that were lost are restored and that some agglomerations caused by aging can be returned [24,25].

From the circular economy perspective, crumb rubber (CR) from end-of-life tyres has been implanted in the past decades showing that asphalt mixtures properties can be improved while overcoming the used tyres disposal problem, reducing the use of raw materials, extending pavement service life and achieving other environmental advantages [26,27]. In the last two decades, asphalt mixes made using binders modified using high amounts of CR have become a widespread practice, so much that many national specifications contemplate a systematic use of these materials [28]. Although the recycling of the end-of-life pavements containing CR is of extreme and current interest, it is still a little-explored topic. Thus, the actual possibility of using the RAP derived from removed and/or reprocessed pavements containing CR particles (CR-RAP) for the formulation of new suitable mixtures should be further investigated [29,30]. The presence of rubber, which is characterised by a complex composition and forms a network structure within the binder matrix, affects the aging process of the bitumen. Although several studies have been conducted on the aging mechanisms of the crumb rubber modified bitumen (CRMB), the effects of the CR degradation on the rheological properties remain unclear [31–33]. Moreover, the existing rejuvenators designed for RAP have not yet evidence of effectiveness in producing asphalt mixtures containing CR-RAP, lacking a specific characterisation of the influence of the long-term aging phenomena on CRMB and a complete knowledge of the rejuvenating mechanism on these rubber materials [34,35].

Hence, the dual objective of this study was on the one hand to understand the effects induced by accelerated aging procedures on the rheological properties of a CRM bitumen (unmodified binder was used as a reference) and on the other hand to evaluate the potential and the effectiveness of a bio-based recycling agent, which is not specifically formulated for the rejuvenation of CRMBs, on a oxidised binder containing CR particles. This kind of recycling agents offers environmental advantages, showing at the same a good affinity with the crumb rubber so much that rejuvenators enriched with CR are being recently developed [36,37]. The experimental set up involved the analysis of the behaviour of the unaged, aged and rejuvenated blends, adopting different rheological performance-related test methods. For this purpose, viscosity, frequency sweep and creep-recovery (single and multiple) tests were performed using a dynamic shear rheometer.

2. Materials and methods

To study the effectiveness of the rejuvenator on the performance of bituminous binders, artificially aged binders rather than extracted and recovered from RAP were considered. This choice was dictated by conceptual and practical reasons. Uncertainties about real characteristics of the original binder used in the pavement, i.e., possible presence of polymers or other modifiers were removed and several drawbacks related to the extraction and recovery processes were overcome: the processes could affect the recovered binder properties, operational difficulties occur to completely remove the residues of solvents after extraction and some methods heavily depend on the skill of the operator for consistent results [38].

Therefore, the experimental investigation was organised starting from a virgin unmodified binder (B) and a crumb rubber modified bitumen (CRMB), which were subsequently aged and then rejuvenated. The same bitumen, specifically the same production batch, was used for all the analysed blends. The base binder for preparing all blends was a 70/100 penetration grade bitumen ($\text{Pen}_{25\ ^\circ\text{C}}$=84 0.1 mm; R&B =46.4 °C). The CRMB was produced by adding to the bitumen B 20% wt. CR powder (0–0.8 mm grain size), which consists of a balanced mix of processed automobile and truck scrap tyres. A thermogravimetric analysis (TGA) of the recycled rubber, which was performed using the TGA/SDTA 851e thermobalance (Mettler Toledo, USA), revealed the presence of polymers (57.3%), carbon black (32.1%), ash (6.0%) and a mix of plasticiser and additives (4.6%). The modified binder was obtained by directly blending the CR to the preheated binder and by stirring in a laboratory mixer the ingredients for 60 min at 4000 rpm, maintaining the temperature at 180 °C.

These two reference binders were artificially aged using the rolling thin-film oven (RTFO) procedure, in which elevated temperatures reproduce the manufacturing and placement aging, and the pressure-aging vessel (PAV) test, which uses heat and pressure to simulate in-service aging over a 7–10 year period. Specifically, two subsets of samples were created: short-term (STA) and long-term aged (LTA) products. In the first case, the EN 12607–1 standard procedure (163 °C for 75 min) was selected. A modified RTFO protocol proposed by Bahia et al. [39], which involves the insertion of a steel rod (127 mm long by 6.4 mm in diameter) inside the glass bottles to create shearing forces to uniformly spread the high viscosity binder film, was used for the CRMB. Besides, LTA binders underwent a secondary aging cycle: RTFO aged samples were exposed for 20 h to elevated temperatures under pressurised conditions (90 °C and 2.1 MPa) according to EN 14769 standard.

The LTA binders were finally rejuvenated using a commercial bio-based recycling agent (blend of vegetal fatty acids and esters). It is an odour free, heat-resistant and storage stable liquid ($\rho_{@20°C}$=0.92 g/cm^3 and $\eta_{@20°C}$<100 mPa·s), which can reconstitute the chemical composition of the oxidised bitumen and enhances the dispersion process between the aged binder and the new bitumen. This recycling agent, which is formulated to be suitable also for polymer modified binders, has a surfactant property that improves the binder-aggregate chemical affinity and increases the asphalt mixture workability during production and laying processes. The rejuvenated blends were prepared by adding the recycling agent to the preheated aged binders and mixing them with a high shear mixer (Silverson, UK) at a high temperature (160–170 °C) for 120 s to produce homogeneous products, which were stored at ambient temperature for 96 h before testing to allow the complete rejuvenator diffusion. The dosage of the recycling agent was established according to the producer recommendations, i.e. 0.15% by weight of the RAP content. Considering a hypothetical recycled asphalt mixture consisting of 70% RAP and assuming a 4.7% RAP binder content, the amount of the recycling agent was set to 3.2% by weight of the binder. Table 1 provides an overview of the prepared and analysed binders with their labels.

Physica MCR 101 (Anton Paar, Austria) dynamic shear rheometer (DSR) was used to measure some physical properties of the binders and verify the actual effectiveness of the rejuvenator agent. First, a controlled shear rate (CSR) rotational test was used to measure the temperature-dependent shear viscosity (η(T)), defined as the ratio between the shear stress (τ) and the corresponding shear rate ($\dot{\gamma}$), presetting a constant $\dot{\gamma}$ up to the achievement of the steady-state condition. The test setup was a 25 mm-diameter parallel plates geometry with a gap of 1 mm (2 mm for CRMBs), in a temperature range from 80 °C to 160 °C. A shear rate of 6.8 s^{-1} was selected between 80 and 140 °C, of 10 s^{-1} at 150 °C and of 12 s^{-1} at 160 °C.

Primary emphasis was given to the analysis of the linear viscoelastic (LVE) behaviour of the unaged, aged and rejuvenated blends, adopting different rheological performance-related test methods. Frequency sweep tests were used to describe the time-dependent behaviour of the binders using a sinusoidal (oscillatory) loading mode, in terms of complex shear modulus (G*) and phase angle (δ), which are considered predictors of rutting and fatigue cracking in asphalt pavements. Specifically, G* , i.e., the ratio of the applied shear stress (τ) to the resulting shear strain (γ), can be defined as the sample's total resistance to deformation when repeatedly sheared, whereas phase angle (δ) is the lag between τ and γ in harmonic oscillation. These tests were performed in a controlled shear stress (CSS) mode, applying a fixed shear stress amplitude within the LVE region for each temperature value, which was previously established through stress amplitude sweep tests, while the angular frequency (ω) changed in a logarithmic way between 1 and 100 rad/s. Frequency sweeps were completed from -10 to 80 ºC, at 10 ºC intervals, using the parallel plate measuring system: 8 mm-diameter parallel plates and a sample height (test gap) of 2 mm at low and intermediate temperatures ($-10 \leq T \leq 30$ °C), while 25 mm-diameter plates and a test gap of 1 mm for higher temperatures.

Creep and recovery test protocol was applied to evaluate the binders' potential for permanent deformations. This method is used to determine the presence of recoverable (γ_e) and non-recoverable (γ_v) deformations in bitumen using two shear stress steps, i.e., stress and rest phases. Specifically, single and multiple stress creep and recovery (MSCR) testing were considered. In both cases, a 25 mm parallel plate geometry with a 1 mm gap setting was selected. In the single creep-recovery test, the sample was subjected to a constant shear load (τ) for 300 s, then allowed to recover, at zero load, for 600 s. During the recovery period the shear strain (γ) was continuously monitored. Tests were performed at 25 and 40 °C setting the τ equal to 100 and 5 Pa, respectively.

CRMBs were also tested following the MSCR test (EN 16659). This procedure, which is specifically designed for modified bitumens, gives information about the rutting performance of the binders. MSCR test, which was performed at 60 °C, started with the application of a low stress ($\tau = 0.1$ kPa) for 10 creep/recovery cycles then the stress was increased to 3.2 kPa and repeated for additional 10 cycles. Each cycle is composed of a stress phase of 1 s and a rest phase of 9 s. The presence of elastic response is determined by measuring the percent recovery (%R), that is the recovered strain during the rest phase of a cycle, and the non-recoverable creep compliance (J_{nr}), that is the residual strain after a creep and recovery cycle divided by the stress applied.

3. Results

3.1. Shear viscosity

The temperature dependent viscosity curves (Fig. 1a and b) show the classical trend of binder viscosity decrease with temperature and increase with aging. The aging effect results in an upward vertical shift of the curves, proportional to the aging intensity: starting from the top (higher η values), LTA, STA and unaged curves are recognised. Moreover, CRMBs resulted to be more viscous than unmodified B binders, registering 3–9 times higher η values depending on temperature and aging configurations. This trend is mainly

Table 1
Unaged, aged and rejuvenated binders.

Binder	Unaged	Aged		Rejuvenated
		RTFO	RTFO+PAV	RTFO+PAV+RJ
70/100	B	B_STA	B_LTA	B_RJ
Crumb rubber modified	CRMB	CRMB_STA	CRMB_LTA	CRMB_RJ

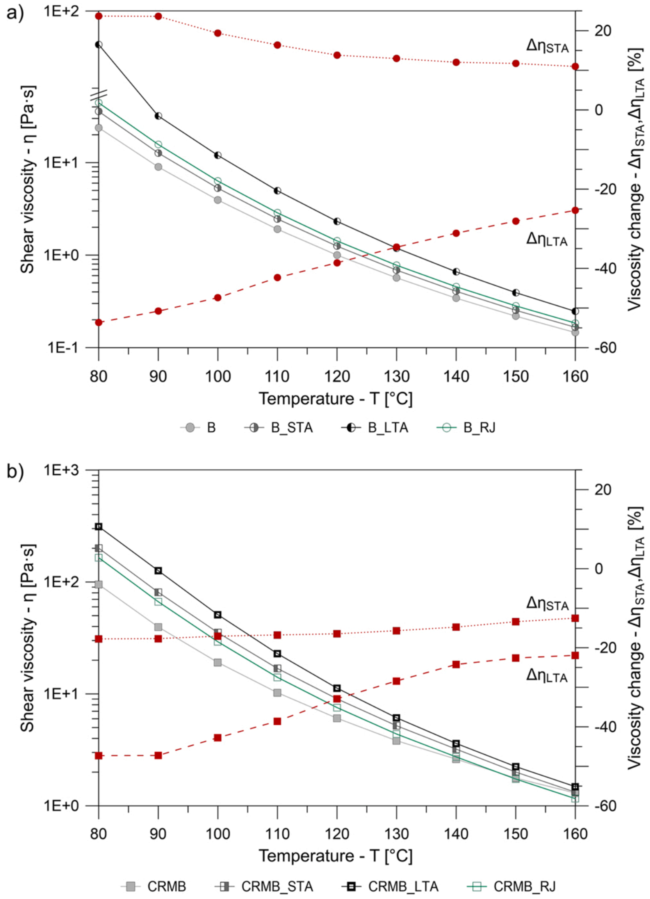

Fig. 1. Shear viscosity vs temperature for binder of B-series (a) and CRMB-series (b).

attributable to the absorption of the lighter components of bitumen by the CR [27]. The viscosity aging index ($\eta_{AI}=\eta_{aged}/\eta_{unaged}$) revealed that each accelerated aging method differently affected the two binder types: specifically, the combination of RTFO+PAV increased more significantly the viscosity of B. The addition of the rejuvenator substantially lowered the viscosity of both B- and CRM-binders, reaching for temperatures above 140 °C η values very close to and, in some cases, lower than those of the unaged configuration. The percentage change of viscosity, calculated with respect to the STA ($\Delta\eta_{STA} = \frac{\eta_{RJ}-\eta_{STA}}{\eta_{STA}}$) and LTA ($\Delta\eta_{LTA} = \frac{\eta_{RJ}-\eta_{LTA}}{\eta_{LTA}}$) conditions, are reported in Fig. 1a and b on the secondary Y axis. The way the indices were defined, a positive value stands for higher viscosity of the rejuvenated condition compared to the aged one. Fig. 1a shows a marked temperature dependent $\Delta\eta_{LTA}$ for B (−54% @80 °C to −25%@160 °C), but this viscosity decrease did not allow to recover the initial values and even those measured after the RTFO test. Instead, the addition of the rejuvenator in CRMBs led to a reduction in viscosity, which turns out to be lower than those recorded after the RTFO+PAV ($-47\%<\Delta\eta_{LTA}<-22\%$) and RTFO ($-18\%<\Delta\eta_{STA}<-12\%$) procedures.

3.2. Frequency sweep

The analysis of the viscoelastic quantities in the LVE region of unaged, aged and rejuvenated binders started from the frequency sweep test data. STA and LTA processes resulted in a stiffer (G* increase) and more elastic (δ reduction) response of materials. These trends were more noticeable at medium and high temperatures. Complex modulus isothermal curves were combined in a single plot and shifted with respect to a reference temperature, in this case 20 °C, using the Williams, Landel and Ferry (WLF) model [40] to form a continuous curve according to the time-temperature superposition (TTS) principle. The master curves (Fig. 2a and b) revealed an upward vertical shift from the unaged binder trend following the accelerated aging methods, whereas a downward vertical shift is evident for B_RJ and CRMB_RJ with respect to the long-term aged curve. The effect of rejuvenator turns out to be more significant at high frequencies (low temperatures) independently of the binder type. Although the rejuvenated G* -curve lies above that of the corresponding virgin binder at low and medium frequencies (high and medium temperatures), a crossover is observed around the ω_r = 10 rad/s. Moreover, a greater reduction in complex modulus is found when the agent is added to the CRMB: the rejuvenated G* -curve (CRMB_RJ) stands below the CRMB_LTA one over the whole angular frequency spectrum.

An analytical evaluation of the aging following RTFO and RTFO+PAV procedures, as well as the rejuvenator efficacy, required the

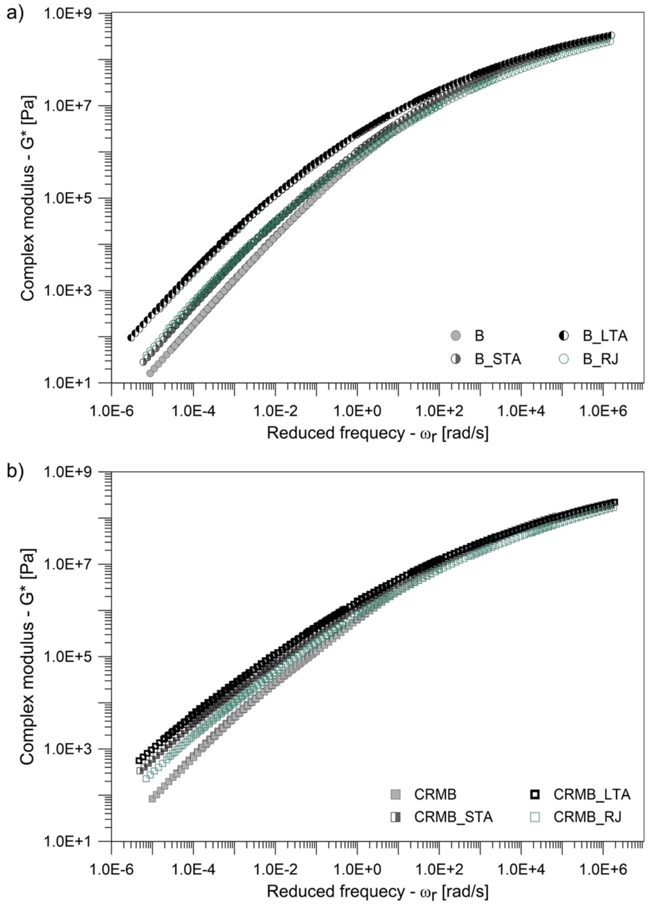

Fig. 2. Master curve for binder of B-series (a) and CRMB-series (b) at 20 °C.

determination of aging indices. As proposed by other authors [41,42], complex modulus aging index ($G^*_{AI}=G^*_{aged}/G^*_{unaged}$) and phase angle aging index ($\delta_{AI}=\delta_{aged}/\delta_{unaged}$) were calculated. The way the indices were defined, $G^*_{AI}> 1$ describes a material stiffening, whereas $\delta_{AI}< 1$ implies a more elastic response. In addition, although the fatigue ($G^*\cdot\sin\delta$) and rutting ($G^*/\sin\delta$) factors introduced in the SHRP programs in the past decades have poor correlation with the real performance of binders [43,44], the assessment of their variation (expressed in term of aging index) after aging and rejuvenator processes appeared to be relevant parameters to consider. The G* and δ values measured at loading frequency of 10 rad/s were used to calculate the aging indices, which were plotted versus temperature in Figs. 3 and 4. These indices allowed to highlight and quantify the trends already emerged from the master curve

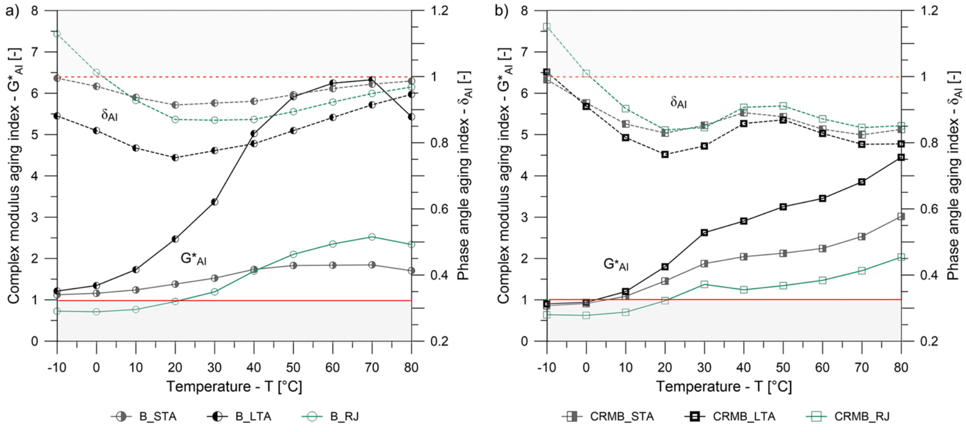

Fig. 3. Complex modulus (Y1) and phase angle (Y2) aging indices vs temperature.

reading, namely, the aging thermo-dependence and the high effectiveness of the rejuvenator in restoring, more clearly at low temperatures, the rheological behaviour of the binders (above all for CRMB) similar to the unaged condition. On the one hand, the binders aging is more evident in the viscoelastic behaviour at medium-high temperatures (> 30 °C); on the other hand, the effectiveness of the rejuvenator is evidenced by the values of the aging indices close to unit and significantly lower than those registered after the RTFO+PAV procedures. Another remarkable aspect is related to the limitation of aging effects for CRMB_LTA, whereby all aging indices were about 30–50% lower than those of B_LTA. The accelerated aging methods always involve for unmodified bitumens an oxidising action that implies an increase in the asphaltene fraction, and finally, in the consistency of the binder itself [15]. This phenomenon implies a complex modulus increase and a phase angle reduction; which inevitably define aging indices much higher than unit (vice versa for δ_{AI}). By way of example, considering the temperature of 60 °C, B_LTA binder registered values of $G^*_{AI,}$ $(G^*\cdot\sin\delta)_{AI}$ and $(G^*/\sin\delta)_{AI}$ greater than 6. But, a beneficial partial boosting of ductility resources is observed for CRMB_STA and CRMB_LTA at low temperatures: all aging indices including the G* quantity resulted to be less than unit below 10 °C. This behaviour denotes that the accelerated aging procedures (thin film, high temperatures and high pressures) led to the CR degradation, generating a more homogeneous binder matrix and enhancing the interaction between the rubber particles and the bitumen, i.e., desired effects during the production of CRMB in plant [45]. This observed behaviour is in line with the findings of other studies, which attributed the improved aging resistance to the presence of polymeric rubber chains, antioxidant agents and carbon black in CRM binders [46,47].

3.3. Creep-recovery and multiple stress creep recovery test

Some peculiar viscoelastic features of the binders, as well as of the aging and rejuvenation processes, were analysed using the creep and creep recovery curves, in addition to the frequency sweep data. By way of example, Fig. 5 depicts the time-dependent strain function $\gamma(t)$ of B and CRMB at 25 °C ($\tau = 100$ Pa) for all the considered conditions. First, a different deformation response between unmodified and CRM bitumen is clearly identified. Moreover, as also evidenced by the data shown in Table 2, which summarises the measured values of maximum strain at the end of the stress phase (γ_{max}), the recovered (γ_e) and un-recovered (γ_v) strain and the elastic recovery ($\%R=100\cdot(\gamma_e/\gamma_{max})$), the reported trends are extremely thermo-dependent. The graphical representation of $\gamma(t)$ for CRMB (25 and 40 °C) and B at 25 °C describes the classic viscoelastic behaviour in which an increasing deformation is observed as long as the load is applied ($0 \leq t \leq 300$ s), distinguishing between a purely elastic deformation (γ_1) occurring immediately after the beginning of the test, a delayed viscoelastic deformation (γ_2) followed by a purely viscous one (γ_3) after reaching the steady state [48,49]. In the rest phase ($300 \leq t \leq 900$ s), a significant part of deformation is recovered (%R=33 @25 °C and 27 @40 °C). Instead, B exhibits an almost "viscous" behaviour at 40 °C, showing a continuous increasing deformation in the creep phase and a very limited elastic recovery (%R=2 @40 °C) after the loading removal (Table 2). Almost all deformation energy is presumably dissipated in internal friction or spent to deform the liquid matrix of the bitumen, mainly producing a non-reversible flow [50]. Besides, B registered a higher γ_{max} value than CRMB in all aging conditions (Table 2). Indeed, the CR acts on the one hand as a partial inhibitor of the deformation phenomena, making the binder stiffer and, simultaneously, as an elasticity enhancer by increasing the extent of γ_e, improving the resistance to permanent deformation (rutting). However, regardless of the binder type, the aging processes significantly affected the response under shear creep and recovery: marked changes in both parameters, i.e., γ_{max} reduction and %R increase, were especially visible in B_LTA and CRMB_LTA. These findings are completely in line with what has already emerged from the frequency sweep tests, in which the accelerated aging methods have transformed the binders in stiffer (G* increase) and more elastic (δ reduction) materials. The addition of the rejuvenator reduced this aging effect, making the rejuvenated binders' behaviour closer to that registered after the RTFOT method (STA condition). But unlike B_RJ in which the shear strain curve stands between those of B_STA and B_LTA, the CRMB_RJ showed consistently higher values (and lower %R values) than CRMB_STA, approaching therefore the behaviour of the unaged CRMB.

The increase of the stress level and of the number of load applications in the creep-recovery procedure highlight the activation of the CR network inside the binder, capturing not only the stiffening effect but also the delayed elastic one. Starting from the data

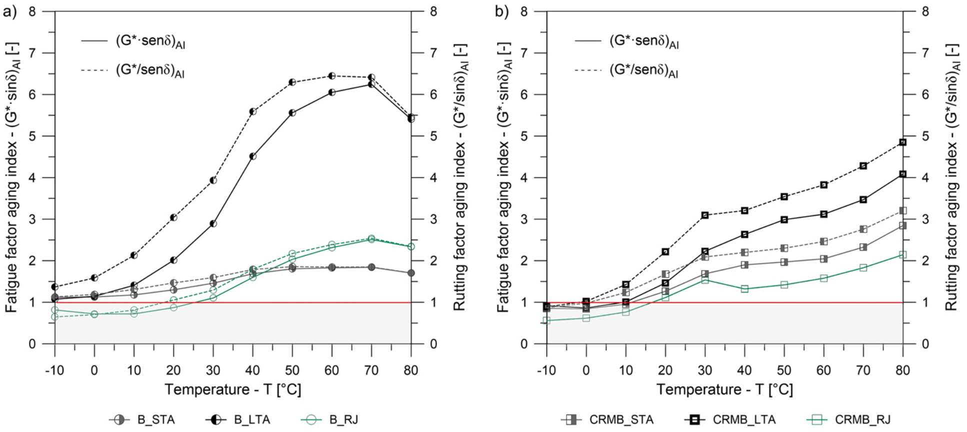

Fig. 4. Fatigue factor (Y1) and rutting factor (Y2) aging indices vs temperature.

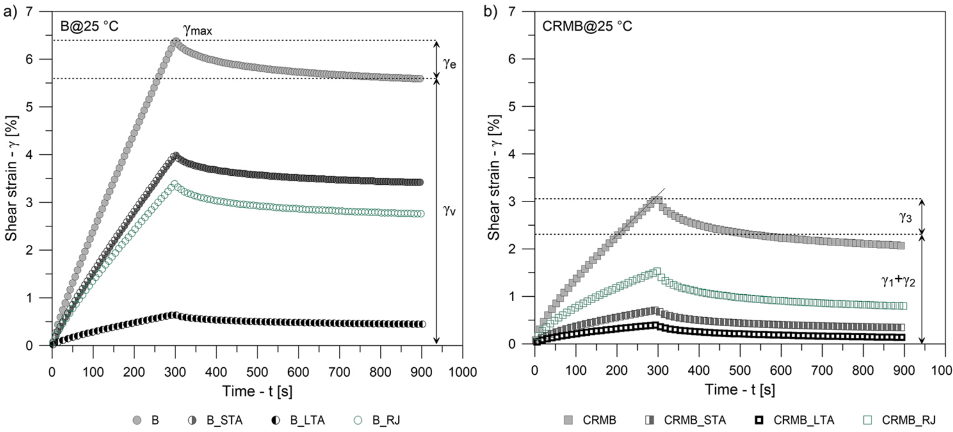

Fig. 5. Creep and creep-recovery curves γ(t) of B-series (a) and CRMB-series (b) at 25 °C.

Table 2
Strain parameters measured in creep and recovery test.

Binder	T = 25 °C; τ = 100 Pa				T = 40 °C; τ = 5 Pa			
	γ_{max}	γ_e	γ_v	%R	γ_{max}	γ_e	γ_v	%R
B	6.42	5.59	0.83	12.9	10.03	9.83	0.20	2.0
B_STA	4.01	3.42	0.59	14.7	5.92	5.77	0.15	2.5
B_LTA	0.66	0.45	0.21	31.4	0.89	0.79	0.10	11.3
B_RJ	3.41	2.76	0.65	19.1	4.45	4.20	0.15	3.4
CRMB	3.09	2.07	1.02	33.0	2.31	1.69	0.62	26.8
CRMB_STA	0.72	0.35	0.37	52.0	0.39	0.21	0.18	46.2
CRMB_LTA	0.41	0.14	0.27	65.8	0.25	0.11	0.14	54.6
CRMB_RJ	1.53	0.80	0.73	47.9	0.86	0.55	0.31	36.2

measured during the MSCR test, the same parameters introduced in the creep and recovery test were calculated for each of the 10 cycles for both loading condition (0.1 and 3.2 kPa) and the average values of %R ($\%R_{@0.1}$ and $\%R_{@3.2}$) were determined. In addition to the evaluation of the elastic response, non-recoverable creep compliance ($J_{nr}= \gamma_v/\tau$), i.e., an indicator of the resistance of bituminous binders to permanent deformation under repeated load, was also defined. Again, values were calculated for all cycles, and the average of the 10 cycles for each load condition was determined ($J_{nr@0.1}$ and $J_{nr@3.2}$). To better highlight the behaviour of the material, the first 6 cycles in the loading condition of 0.1 kPa were diagrammed in the Fig. 6. Table 3 reports all the %R and J_{nr} values referred to the 4 binders of the CRMB-series. The trends in the curves and the values of the parameters confirm what emerged from the creep-recovery test, even for higher temperature (60 °C) and creep stress (0.1 and 3.2 kPa). An increase of aging intensity corresponded to a significant enhancement of elastic recovery accompanied by a reduction of non-recoverable compliance, denoting the ability of CRM aged binders

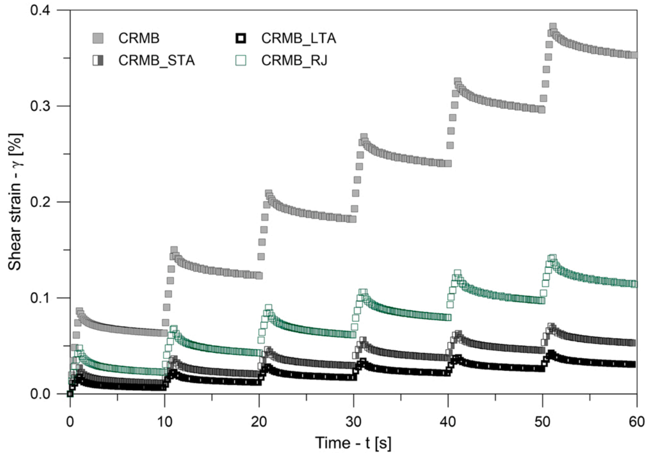

Fig. 6. Creep-recovery curve after 6 consecutive cycles (T = 60 °C and τ = 0.1 kPa).

Table 3
Average %R and J_{nr} calculated from MSCR data.

Binder	T = 60 °C; τ = 0.1 kPa		T = 60 °C; τ = 3.2 kPa	
	$\%R_{@0.1}$	$J_{nr@0.1}$	$\%R_{@3.2}$	$J_{nr@3.2}$
CRMB	35.9 ± 3.3	0.56 ± 0.03	12.2 ± 1.4	0.89 ± 0.01
CRMB_STA	73.7 ± 6.3	0.07 ± 0.02	57.3 ± 1.1	0.12 ± 0.01
CRMB_LTA	77.8 ± 6.1	0.04 ± 0.01	70.4 ± 3.2	0.05 ± 0.01
CRMB_RJ	64.4 ± 4.6	0.16 ± 0.03	46.0 ± 0.7	0.27 ± 0.01

to resist rutting. Moreover, the addition of the rejuvenator makes the behaviour of CRMB_RJ close to that of CRMB_STA but with significantly lower %R and almost double J_{nr} values.

4. Conclusions

Reclaimed asphalt pavement that contains crumb rubber particles (CR-RAP) can be a promising solution for the production of sustainable asphalt mixtures, providing benefits in terms of material, cost and environmental savings. The effectiveness of adding rejuvenators to RAP mixes containing CR is still an understudied topic. Hence, the aim of this study was to understand the effects induced by accelerated aging procedures on a unmodified and a CRM bitumen and to evaluate the potential restoration of their physical properties resulting from the addition of a bio-based recycling agent.

The tests performed using a dynamic shear rheometer (DSR) have highlighted some macroscopic trends, which may suggest some explanation on the aging mechanisms of CRMBs and on their rejuvenation.

- The addition of the rejuvenator significantly lowered the viscosity of the unmodified bitumen and the CRMB. However, while for the B-binders the viscosity decrease did not allow to recover the initial values (including those measured after the RTFO test), the addition of the bio-based recycling agent in the CRM-binders decreased the viscosity below the unaged CRMB for temperatures above 140 °C.
- According to the frequency sweep tests, the rejuvenator was more effective for the CRM-binders compared to the B-binders, above all at low and medium temperatures. Furthermore, the CRMB_RJ master curve (G^* vs ω_r) lied not only below the CRMB_STA but even below the CRMB at high frequency (low temperature), which is in accordance with the viscosity results.
- Complex modulus and phase angle aging indices, as well as fatigue and rutting aging indices were calculated to analytically quantify the aging mechanisms and the rejuvenator efficacy. These indices clearly showed the aging thermo-dependence and the high effectiveness of the rejuvenator in restoring, more clearly at low temperatures, the rheological behaviour of the binders (above all for CRMB) similar to the unaged condition. Another relevant aspect is the mitigation of aging for CRMB_LTA, presenting aging indices about 30–50% lower than those of B_LTA
- According to the creep-recovery and multiple stress creep recovery tests, a different deformation response (strains and elastic recovery) of between the B and CRM binders was clearly identified, reporting an extremely thermo-dependent trends. Also, regardless of the binder type, the aging processes significantly affected the response under shear creep and recovery and the addition of the rejuvenator reduced the aging effect. In particular, the rejuvenated CRM binder approached the behaviour of the unaged CRM binder, but registering higher elastic recovery (%R) and lower non-recoverable creep compliance (J_{nr}) regardless of the temperature and the stress level.

Finally, this study shows that the addition of the bio-based recycling agent, despite not being specifically formulated for crumb rubber modified binders, was able to mitigate the aging of CRMB, even more than for the unmodified bitumen, almost restoring its rheological properties similar to the unaged condition. Hence, the recycling of the end-of-life pavements containing CR seems to be not only an option in the design of new asphalt mixtures, but possibly an even better solution in terms of mechanical behaviour and savings compared to a conventional RAP. Obviously, the findings of this research represent a piece of the knowledge on the CRMBs' aging and rejuvenation processes. Further investigation including the analysis of different rejuvenator type and content, as well as the study of the behaviour in the asphalt mixtures scale are necessary for the generalisation of the obtained results.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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