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Investigating the effects of rejuvenating agents by means of Reclaimed Asphalt Model System

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Abstract

The experimental study described in the paper investigated the effectiveness of rejuvenating agents used in reclaimed asphalt pavements by means of a novel methodological approach. Such approach is based on mechanical testing of Reclaimed Asphalt Model Systems (RAMS), constituted by a single-size reclaimed asphalt compacted to a high value of voids in mineral aggregates. The rationale behind the methodology is that the response of RAMS under external loadings is mainly dependent upon the bonding of the binder phase, thus allowing the direct assessment of the effect of employed rejuvenator on the aged binder of reclaimed asphalt. The investigation was conducted on a set of rejuvenators of different types, including two vegetable oils and a paraffinic hydrocarbon oil. The experimental program involved using standardized test procedures for the determination of the so-called dynamic modulus, the fatigue failure properties and the indirect tensile strength of the systems. The sensitivity to ageing effects of rejuvenating products was also investigated.
Although the results obtained from the study do not allow definitive conclusions to be drawn on RAMS, they confirm that the use of proposed approach have good potentialities for the assessment and ranking of rejuvenating agents.

Keywords
Reclaimed asphalt, Rejuvenators, Model systems, Dynamic modulus, Fatigue failure, Indirect tensile strength, Ageing

1 Introduction

The growing need for sustainable solutions in the transportation infrastructure sector has considerably increased the use of Reclaimed Asphalt (RA) for the construction and maintenance of road pavements [1]. Recycling of RA leads to several environmental benefits, including reduced depletion of non-renewable resources (virgin aggregates and bituminous binder), greenhouse emissions, generation of waste and landfill disposals [2-5]. The less demand of raw quarry materials and the associated lower extraction costs offer benefits in economic terms, too [6].
Notwithstanding the environmental and economic advantages associated to usage of RA, the production of asphalt mixtures containing RA poses several issues related to RA homogeneity,

retained humidity of RA, method of introducing RA in the mixer, control of mixing time and temperature [7-10].

The addition of RA has also a significant impact on the mechanical performance of final mixture. It is beneficial in the short term against rutting, while in the long term it may increase cracking susceptibility of the material with consequent risk of premature fatigue failure and low-temperature rupture of pavement structure [11-13]. This is mainly due to the presence of aged binder, that usually make asphalt mixture containing RA more stiff and fragile compared to conventional ones [14-16].

Chemical ageing of asphalt binder is caused by the combination of several phenomena that produce irreversible changes in molecular structure of the material. Such phenomena include oxidation reactions, polymerization and evaporation of lighter components [17]. It has been demonstrated that susceptibility of a given binder to chemical ageing depends upon its crude source and manufacturing process [18]. To prevent loss of fatigue endurance and thermal cracking resistance of mixtures containing significant percentage of RA, rejuvenating agents (RE) are commonly used in mix production [19]. The RE is intended to restore (or partially restore) the physical and chemical properties of aged binder by rebalancing the relative proportion of maltene fractions with respect to other components and by making the RA binder effectively available for blending [20].

Various materials have been used as RE in asphalt mixtures, including vegetable oils, waste oils, bio-oils, refined crude oil products [21-26]. Secondary artificial antioxidants, like dilauryl thiodipropionate, have been also recently investigated as regenerator of RA binders [27].

A possible drawback associated to the usage of RE consists in excessive softening of the binder phase that may results in severe rutting problems [28]. The use of suitable RE product and dosage is then of paramount importance to prevent such kind of problems and to make RE effective in achieving the performance targets of the mixture. This depends on many factors involved in the mix design of the material, including the source of RA, the compatibility between RE and RA binder, the way RE is added to other mix components, the characteristics of virgin bitumen [29].

The effectiveness of RE and related effects can be assessed at the binder scale, by analysing the rheological and chemical properties of binder extracted from RA, or at the mixture scale, by performing mechanical tests on mixtures containing RA [30]. At the binder scale, full activation of RA binder is generally assumed even thought this could not be the real case since some of the RA binder may still stay inherent [31]. Moreover, when the analysis is carried out on the extracted binder, some concerns remain about the possible damage of RA binder due to solvents adopted in the extraction process [32-33]. At the mixture scale, the effects of RE product may be partially superposed to those of other mix-dependant factors, including aggregate interlock and bulk volume effects. This may hamper the assessment of the actual potential of rejuvenating agent in enhancing the performance of the whole asphalt mixture [22, 32, 34]. Moreover, laboratory testing may be very time consuming, especially in the case in which more RE products are tested in combination with different types of mixtures and/or RA sources.

An alternative approach for the evaluation of the effectiveness of RE products has been recently proposed by Dalmazzo et al. [35]. Such an approach relies on the concept of Reclaimed Asphalt Model System (RAMS), made of single-sized RA material properly selected to obtain a skeleton structure characterized by a very high volume of internal voids. The rationale behind this method is that the response of RAMS under external loadings is mainly dependent upon the bonding of the binder phase. This was deemed allowing the direct assessment of the effect of considered RE on the actual binder of RA, without the need of extracting this last one from reclaimed asphalt or subjecting virgin binder to artificial aging.

Moving from the preliminary findings of previous work, the experimental study described in this paper investigated a set of RE products by means of RAMS approach.

The objective of the study was to assess and compare the effectiveness of selected products in restoring the characteristics of aged binder contained in RA. The evaluation of the effects of aging on rejuvenated binder was also an objective of the study.

At the same time, the investigation aimed to confirm the efficacy of the proposed methodology in being used as a simple and straightforward tool for discriminating and ranking RE potential, overcoming the limitations of other aforementioned techniques.
Coherently with the objectives and aims of the study, the laboratory experimentation entailed using well established standardized test procedures to be carried with ordinary laboratory equipment. These procedures included the test methods for determining the so-called dynamic modulus, fatigue failure and indirect tensile strength of the model systems.

2 Materials and methods

2.1 Base materials

The RA employed in the investigation derived from the demolition and milling of existing pavement located on a motorway section in northern Italy. It was sampled from a local plant where reclaimed materials were processed through a crushing unit and subsequently screened into different fractions. For the purposes of this study, a single-sized 5/8 RA fraction was used to prepare the model systems. This choice was dictated by the need of reducing the presence of clusters and fine particles, to attain uniform lithic skeletons coated by a controlled volume of asphalt binder [35]. The selected fraction was obtained by sieving from a 20RA0/14 stockpile, in accordance with EN 13108-8 [36].
Representative 5/8 RA samples were then subjected to a preliminary laboratory characterization consisting in the determination of binder content, density and particles gradation. Binder content was determined via ignition method, following the procedure indicated by EN 12697-39 [37]. Binder content values were expressed in terms of percentage by the weight of RA (B_{RA}) and percentage by the weight of aggregates (B_{AGG}). Determination of density included the maximum density of RA particles (ρ_{mv}), measured according to EN 12697-5 [38], and that of aggregates recovered from RA ignition (ρ_p), measured according to EN 1097-6 - Annex H [39]. Sieve analysis for the determination of particles gradation was carried out according to EN 12697-2 [40] for RA, and according to EN 933-1 [41] for recovered aggregates. The results obtained from testing are reported in the following Tables 1 and 2.

Table 1. Characteristics of RA and aggregate particles recovered from ignition.

	B_{RA} (%)	B_{Agg} (%)	ρ_{mv} (Mg/m³)	ρ_p (Mg/m³)
RA 5/8	4.3	4.5	2.579	2.74

Table 2. Sieve-size distribution of RA and recovered aggregates.

Diameter (mm)	Passing (%)	
	5/8 RA	Aggregates recovered from 5/8 RA
10	100	100
8	99.9	100
6.3	64.8	77.9
5.6	34.3	56.0
5	3.0	31.9
4	0.1	19.5
2	-	14.1
1	-	11.2
0.5	-	8.8
0.25	-	7.0
0.125	-	5.6
0,063	-	4.4

The set of investigated RE agents included three different commercially available products, named A, B and C. These recycling agents are proprietary products labelled by generic descriptors that define their origin: A and B were vegetable oils, while C consisted of a mix of paraffinic hydrocarbon oils. Their basic characteristics, gathered from the manufacturer's datasheets, are shown in Table 3.

Table 3. Basic characteristics of RE products.

RE	Density ρ (Mg/m³)	Dynamic viscosity η (mPa·s)
A	0.86 ÷ 0.90 (@20 °C)	34 ÷ 45 (@20 °C)
B	0.98 (@20 °C)	500 (@20 °C)
C	0.93 (@20 °C)	100 (@25 °C)

A neat asphalt binder was also employed in the investigation to prepare reference RAMS. The selected material belonged to the 50/70 pen grade, which is the same grade used in the production of the original asphalt mixtures reclaimed from the existing pavement.

2.2 Model systems

Base materials were combined to obtain different types of RAMS, as described in the following:

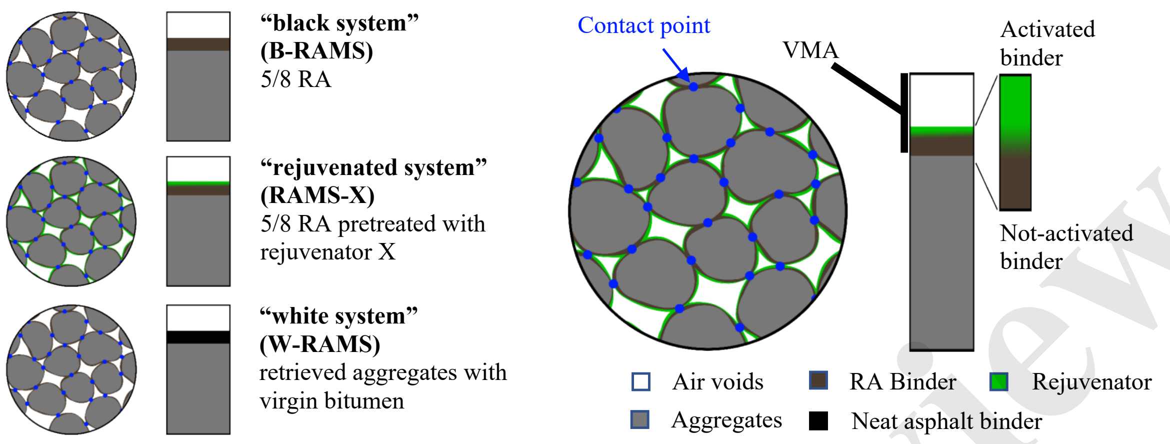
- black model system (B-RAMS), made of 100% of 5/8 RA without any RE,
- white model system (W-RAMS), made of aggregates recovered from ignition and 50/70 pen grade asphalt binder. The binder content was set equal to that of the 5/8 RA,
- rejuvenated model systems (RAMS-X), made of 100% of 5/8 RA with the addition of RE of type X (A, B or C). Three dosages (4, 8 or 12% by weight of binder) were used for each RE type.

B-RAMS was assumed as reference system for the assessment of self-activation aptitude of the RA binder by thermal effects, i.e. the bonding capability of aged binder triggered by the sole action of heat during mixing and compaction. W-RAMS was assumed as reference system to quantitively assess the effectiveness of RE in achieving the properties of virgin binder.

All model systems were prepared following a procedure that included few simple steps. Specifically, in the case of black and rejuvenated systems the procedure consisted in i) heating the 5/8 RA fraction until reaching the target temperature of 150 °C, ii) mixing the material for 5 minutes, iii) blending the hot RA with RE (this step was obviously omitted for B-RAMS). The RE was added at the selected dosage by spreading it on RA particles in two subsequent stages (after 1 and 2 minutes of mixing), to achieve a uniform distribution of the agent [42]. In the case of W-RAMS, aggregates and neat asphalt binder were mixed at the same temperature and blending time adopted for the other systems to avoid any possible effect associated to these variables.

The blends were compacted to the same volume of voids in mineral aggregates (VMA), to obtain an internal structure common to all models (Figure 1a). It is worth noting that the binder film in rejuvenated RAMS is effectively activated (or regenerated) to an extent that depends on the interaction mechanisms between RE and aged binder, driven by gradual diffusion of the rejuvenator agent within the binder thickness [43-44]. The relative proportion between activated and not activated binder (Figure 1b) has a direct influence on the mechanical response of RAMS.

Compaction of systems was carried out by means of gyratory shear technique, according to EN 12697-31 [45]. Gyratory specimens were fabricated using cylindrical moulds of 100 mm diameter with imposed heights of 65 mm and 135 mm. Specimens with one or the other height were used depending on protocol adopted for the specific test. Number of gyrations were adjusted to obtain the target VMA value, set equal to 30% to create distributed particle-to-particle points contacts within the RAMS skeleton.



(a) (b)

Figure 1. (a) Internal structure of model systems and (b) phase distributions in rejuvenated RAMS.

It is worth noting that RAMS used in the investigation did not intend to replicate, by any means, asphalt mixtures constituted by 100% RA, which are properly designed to obtain targeted volumetric properties and mechanical performances [31].

2.3 Testing

Measurements of the dynamic modulus of model systems were carried out following the procedure indicated by AASHTO T378 [46]. Cylindrical specimens with 135 mm height were subjected to controlled haversine compressive stress at multiple loading frequencies (25, 20, 10, 5, 2, 1, 0.5, 0.2, and 0.1 Hz) and at three different temperatures (4, 20 and 40 °C).
The dynamic modulus |E*| (i.e. the norm of the complex modulus E*) was calculated from sinusoidal axial stresses and resulting axial strains as the ratio of the peak-to-peak stress to the peak-to-peak strain. By applying the time-temperature superposition principle, |E*| values obtained at various temperatures were shifted to the reference temperature of 20 °C to construct the corresponding master curves. As suggested by Rowe et al [47], a free-shifting technique was preferred as it allows the properties of material to be obtained independently, without imposing any constraints on the outcome of the analysis. In accordance with the recommendations set by AASHTO R84 [48], the test data were fitted to a sigmoidal function, which describes the general form of the dynamic modulus master curve as follows:

$$\log|E^*| = \delta + \frac{Max - \delta}{1 + e^{\beta + \gamma \log f_r}} \quad (1)$$

where Max, β, δ, γ are the fitting parameters and f_r is the reduced frequency (Hz). This last one was evaluated by employing the shift factors $a(T)$ according to Equation 2:

$$f_r = a(T) \cdot f \quad (2)$$

where $a(T)$ is the shift factor at the generic test temperature T (°C), and f is the physical frequency (Hz). Due to the peculiar composition and volumetric characteristics of the model systems, it was considered appropriate not to apply the equations given by AASHTO R84 for estimating the limiting maximum modulus and the reduced frequency, leaving these parameters as fitting parameters in equations 1 and 2.

According to Zaumanis et al. [29] and Shen et al. [49], who investigated the rheological behaviour of rejuvenated binders extracted from RA, the optimum rejuvenating content can be defined as the content that allows the rheological properties of the original binder to be restored. In this study, the original-like state of binder was assumed to be reproduced by W-RMAS, containing the 50/70 neat asphalt. Consistently with such a hypothesis, considerations on optimum dosage of the selected RE products were based upon the comparison of master curves of corresponding RAMS systems with respect to reference W-RAMS.
Failure properties were evaluated via cyclic direct tension fatigue tests carried out on 135 mm height specimens by following the procedure indicated in AASHTO T400 [50]. Measurements were run in the strain-controlled mode, with peak-to-peak on specimen strain amplitude varying in the range 150-200 μm/m, at temperature of 15 °C and frequency of 10 Hz. The test outputs were expressed in terms of number of loading cycles to failure (N_f), determined by considering the number of cycles corresponding to actual rupture of sample.
Indirect tensile strength tests were performed according to EN 12697-23 [51]. Gyratory specimens with 65 mm height were used for testing, with an imposed vertical displacement rate of 50 mm/min. The Indirect Tensile Strength (ITS) of model systems was measured at 4 °C, in order to assess their strength properties at relatively low temperature conditions.
In the perspective of multiple recycling of RA, as required by circular economy principles, the influence of ageing on RA binder treated with RE was also evaluated. The adopted approach consisted in running the dynamic modulus tests on RAMS specimens, after being subjected to short- and long-term ageing in accordance with AASHTO R30 [52]. The short-term ageing (STA) procedure entailed conditioning the loose mixture in oven at 135 °C for 4 hours before compaction; in the long-term ageing (LTA) procedure, compacted specimens were stored at a temperature of 85 °C for 120 hours.

Susceptibility to ageing of virgin and rejuvenated binders was evaluated by comparing the master curves of materials after STA and LTA conditioning to that of B-RAMS (which obviously was not subjected to any conditioning). An Aging Index (AI) was also introduced to quantitatively assess the degree of aging after STA or LTA treatment, as follows (Equation 3):

$$AI = \frac{|E^*|_{AGED}}{|E^*|_{B-RAMS}} \qquad (3)$$

with a clear meaning of symbols. AI was calculated from dynamic modulus values at various temperatures (4, 20 and 40 °C) and at single reference frequency of 10 Hz.

All measurements were run in at least three replicates and averaged data were used in the analysis.

3 Results and discussion

3.1 Dynamic modulus

The master curves of dynamic modulus of B-RAMS, W-RAMS and the rejuvenated RAMS-X systems obtained from testing data are reported in Figure 2.

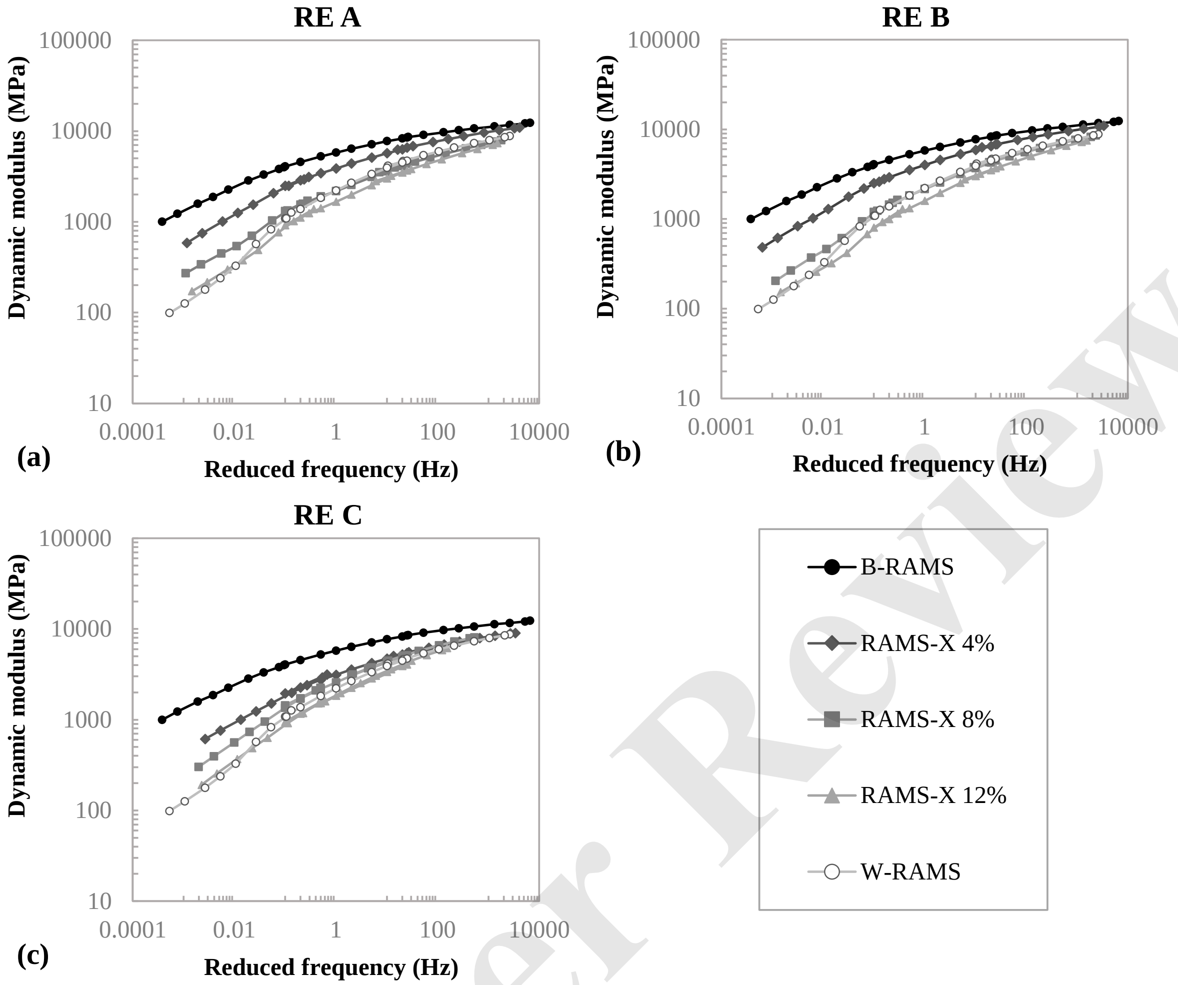


Figure 2. Master curves of dynamic modulus at 20 °C of B-RAMS, W-RAMS and the rejuvenated RAMS-X systems treated with a) RE of type A, b) RE of type B, and c) RE of type C.

B-RAMS and W-RMAS exhibited the highest and lowest |E*| values, respectively, over the whole considered range of reduced frequencies. This is clearly coherent with expectations due to the presence of aged binder in B-RMAS and virgin binder in W-RMAS. For any RE product, the increment of dosage added to RAMS from 4 to 12% reflected on the progressively decrease of stiffnesses, with corresponding master curves moving toward the master curve of W-RMAS. The curve of each RAMS-X at 8% RE dosage appears to be very close to that of W-RMAS, indicating such dosage as the optimum dosage for all the considered agents. This result is coherent with those reported by other authors in literature [29].

Table 4 reports the fitting parameters of sigmoidal function used to mathematically describe the master curves. The shift factors at the temperatures of 4, 20 and 40 °C are reported in the subsequent Table 5. The quality of the fitting was checked by the two statistics R^2 and SER: the former exceeded 0.998 and the latter was always between 0.011 and 0.049.

Table 4. Sigmoidal function fitting parameters.

	B-RAMS	RAMS-A 4%	RAMS-A 8%	RAMS-A 12%	RAMS-B 4%	RAMS-B 8%	RAMS-B 12%	RAMS-C 4%	RAMS-C 8%	RAMS-C 12%	W-RAMS
Max	4.20	4.18	4.13	4.11	4.18	4.12	4.14	4.11	4.14	4.16	4.05
δ	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50
β	-2.03	-1.66	-1.30	-1.15	-1.70	-1.30	-1.07	-1.60	-1.41	-1.14	-1.41
γ	-0.38	-0.40	-0.40	-0.44	-0.41	-0.45	-0.44	-0.41	-0.46	-0.45	-0.54
R^2	1.000	0.999	0.998	0.998	0.999	0.999	0.998	1.000	0.999	0.999	0.998

Table 5. Shift factors at various temperatures.

a(T) (°C)	B-RAMS	RAMS-A 4%	RAMS-A 8%	RAMS-A 12%	RAMS-B 4%	RAMS-B 8%	RAMS-B 12%	RAMS-C 4%	RAMS-C 8%	RAMS-C 12%	W-RAMS
4	2.42	2.21	1.85	1.78	2.13	1.97	1.79	2.12	1.33	0.78	2.02
20	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
40	-2.42	-1.93	-1.96	-1.84	-2.20	-1.93	-1.84	-1.57	-1.70	-1.65	-2.28

Parameters Max and δ define the range of variation of dynamic modulus of each RAMS. Preliminary results showed that all RAMSs had a δ value varying between 0.45 and 0.57. The parameter represents the response of the mixture at high temperatures or low frequencies, where the contribution of the binder phase is negligible, and the behaviour is mainly governed by lithic skeleton. It was then decided that a single value of δ should be considered, as all RAMS have the same lithic structure. A reference value of 0.5 was therefore chosen, corresponding to a dynamic modulus of 3.16 MPa.
Values of Max parameter of all RAMS-X systems fell between the upper asymptote Max of B-RMAS and the upper asymptote Max of W-RAMS, which yielded the values of 4.20 and 4.05, respectively. This confirms that the two systems may actually be considered as reference systems for evaluating, in relative terms, the response of model systems produced with the different rejuvenating agents.
The shape parameter γ, representing the steepness of the sigmoidal function, assumed the highest value for B-RAMS (-0.38) and the minimum value for W-RMAS (-0.54). This implies that presence of aged and stiff binder in the model system led to a reduced relaxation capability of the materials and, as a consequence of that, a flatter dynamic modulus master curve. Conversely, the virgin binder relaxed more, and this reflected in a steeper curve. γ values of rejuvenated RAMS-X systems are in between, showing a general increase with the increase of RE dosage. None of the additives allowed the γ value of the reference system W-RMAS to be reached.
The values of β, which is a parameter that determines the horizontal position of the inflection point, were -2.03 for the B-RAMS and -1.41 for the W-RAMS. Unlike the case of the γ parameter, a monotonic trend of β was observed for each RAMS-X. In particular, β increased with the increase of RE dosage, indicating a horizontal shift of the inflection point of the curves. This denotes that the transition in the rheological behaviour of the materials was shifted to the higher frequency region. In other words, the addition of RE produced the softening of the corresponding systems to an extent that was dependent on the RE dosage. It is observed that for all the rejuvenated products, the model system prepared with 8% RE dosage reached a β value comparable to that of reference W-RMAS.
Comparison between B-RAMS and W-RAMS reveals that ageing of binder reflects on changes in the shift factors, as shown by Table 5. This finding is in accordance with the results reported by Saleh et al. [53]. Data reported in Table 5 also reveal that regardless of the specific RE product, the shift factors decreased as the RE dosage increased, making the material more susceptible to temperature changes than the reference W-RAMS.

3.2 Cycles to failure

The results of the cyclic direct tension tests performed on the model systems at optimum RE dosage are shown in Figure 3, which report the values of cycles to failure N_f corresponding to lower and upper axial tensile strain amplitude (ε_0) adopted in measurements.

The results obtained from B-RMAS were not included in the graph because the very low values, less than 1,000 cycles. This outcome can be explained considering the extreme fragility of the RA aged binder. The heating of the material during preparation of test specimens allows the aged binder to soften and to impart cohesion to the stone structure, which is able to develop a high degree of stiffness. This translates, however, into a very low resistance to the action of cyclic loading, which leads to a sudden formation of micro-cracks with consequent rapid propagation of macro-cracks.

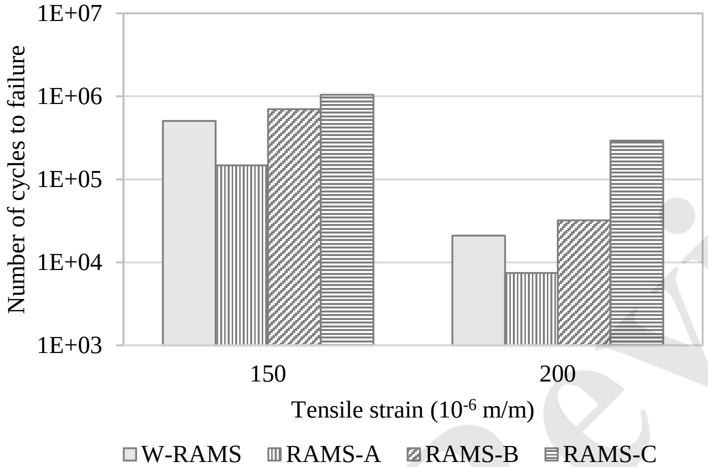


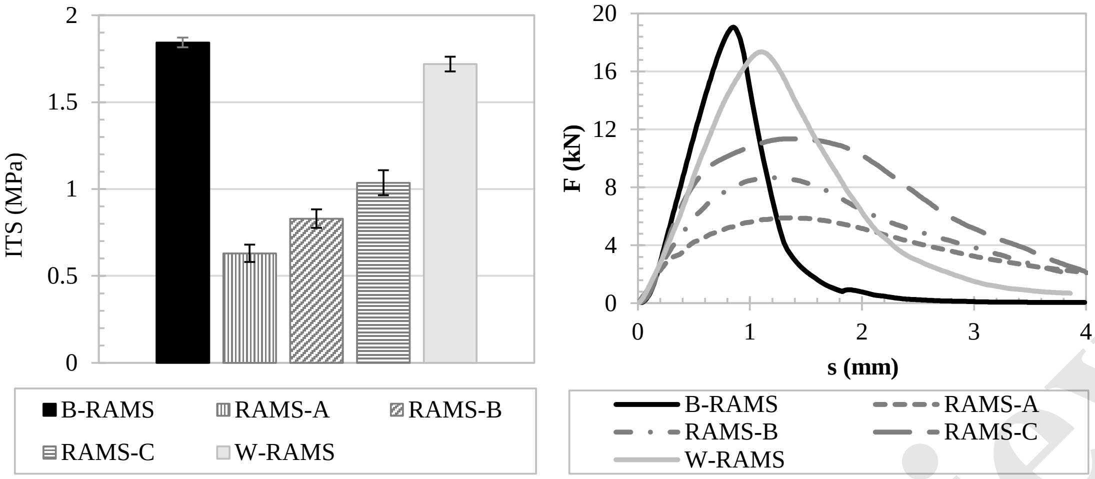
Figure 3. Results obtained from under cyclic sinusoidal direct tension test carried out on model system with 8% RE dosage.

The addition of RE improved the resistance of the material to cyclic loading, to an extent that depended on the type of product employed in the preparation of the model system blend. Contrary to the case of dynamic modulus data, for which no sharp differences were observed between one rejuvenated system and another, based on N_f parameter the distinction is much clearer.

RAMS-C exhibited the highest resistance at both strain amplitudes, followed by RAMS-B and RAMS-A. The relative difference in N_f values with respect to W-RAMS was of about -70% for RAMS-A, +40% for RAMS-B, and +100% for RMAS-C. It is worth noting that the cycles to failure recorded for RAMS-B and RAMS-C exceeded those of W-RMAS containing the neat binder, while N_f values of RAMS-a remained well below of the reference ones.

3.3 Indirect tensile strength

The values of ITS parameter are graphically represented in Figure 4, which also show an example of applied force (F) versus displacement (s) plot obtained from test data. As expected, the high stiffness of the RA binder is reflected in the higher strength of the reference system B-RAMS. The ITS value exhibited by the reference system W-RAMS was also high and very close to the one of B-RAMS (1.72 MPa versus 1.84 MPa): such result was quite surprising since the presence of virgin binder in W-RAMS was expected to significantly reduce the tensile strength of the system. ITS values of all rejuvenated systems were lower than those of reference ones (Figure 4a); moreover, a clear distinction can be made between different products, with RAMS-C showing higher strength (1.04 MPa) than RAMS-B (0.83 MPa) and RAMS-A (0.63 MPa). The qualitative analysis of force-displacement evolution during the whole test (Figure 4b), provides further insights into the mechanical response of considered materials. The reference systems B-RAMS and W-RAMS exhibited a quite similar type of behaviour, characterized by a steep linear increasing trend of the curve followed by a sudden decrease after the peak point. Conversely, the curves of rejuvenated systems showed a flattened shape, characterized by a more gradual increase and a more gradual decreasing of force with displacement before and after the peak region. Such an outcome is similar to that obtained in other studies based on the use of other testing methodologies, like the Illinois Flexibility Index Test [26].



(a) (b)

Figure 4. (a) Indirect Tensile Strength (ITS) at 4 °C and (b) force–displacement curves from ITS test.

The reduction of peak loads and overall flattening of the curves is a clear indication that RE have induced a softening effect on the material. The softening effect is reflected in the decrease of stiffness combined with the increase of ductility, resulting in the overall increase of materials toughness. The data demonstrate that the incorporation of all RE products into the RA aged binder led to a notable reduction in stiffness and an increase in ductility when compared to the 50/70 virgin binder. The differences between the various products may be indicative of different interaction mechanisms that took place at chemical level.

3.4 Ageing

Figure 5 displays the master curves of complex modulus obtained for W-RAMS and for each RAMS-X, after being subjected to STA and LTA conditioning. Figure 3 also contains the master curve of the reference model B-RMAS. The corresponding function fitting parameters are reported in the following Table 6 and Table 7.

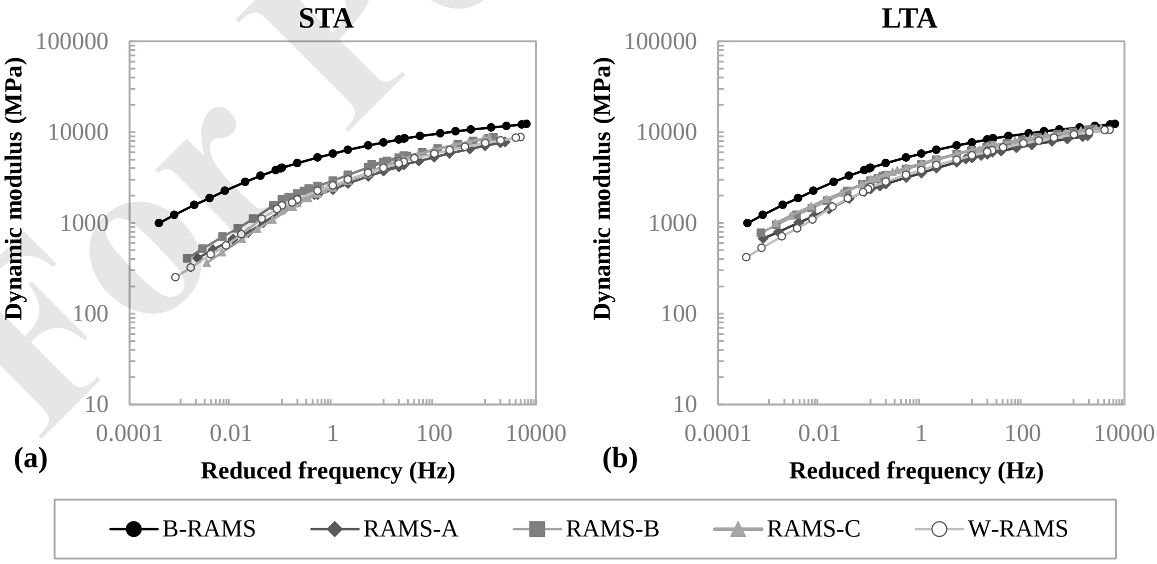


Figure 5. Dynamic modulus master curves of RAMS systems: (a) after short-term aging, (b) after long-term aging

Table 6. Sigmoidal function fitting parameters of aged systems.

	RAMS-A STA	RAMS-A LTA	RAMS-B STA	RAMS-B LTA	RAMS-C STA	RAMS-C LTA	W-RAMS STA	W-RAMS LTA
Max	4.10	4.20	4.13	4.20	4.17	4.20	4.07	4.17
δ	0.50	0.50	0.50	0.50	0.50	0.50	0.50	0.50
β	-1.39	-1.57	-1.53	-1.75	-1.32	-1.78	-1.51	-1.67
γ	-0.39	-0.34	-0.42	-0.37	-0.43	-0.37	-0.45	-0.39
R^2	0.998	0.999	0.999	1.000	0.999	0.999	0.998	0.999

Table 7. Shift factors of aged systems.

a(T) (°C)	RAMS-A STA	RAMS-A LTA	RAMS-B STA	RAMS-B LTA	RAMS-C STA	RAMS-C LTA	W-RAMS STA	W-RAMS LTA
4	2.00	1.88	1.77	1.99	1.68	2.15	2.30	2.31
20	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
40	-1.66	-2.12	-1.87	-2.16	-1.48	-1.87	-2.10	-2.45

The results in Table 6 indicated a general increasing of |E*| values after aging. The increase of stiffness was more pronounced after LTA treatment with respect to STA one. The master curve of W-RAMS subjected to long-term ageing never reached the master curve of B-RAMS. This reveals that combination of STA and LTA procedures didn't fully reproduce actual ageing occurred in RA material. This was expected and quite obvious, since ageing phenomena taking place in real pavements depend on a variety of factors that can be only partially simulated in the laboratory.
The curves of the rejuvenated systems are positioned below the curve of B-RAMS, being very close to W-RAMS after STA conditioning while a higher deviation is observed in the case of LTA treatment. Consistently with the trends described above, γ parameter shifted toward less negative values after ageing while the inflection point frequency determined from β shifted toward the lower frequencies/higher temperatures domain.
Looking at the values of the shift factors displayed in Table 7, an increase in the shift between the isotherms is generally observed for each RAMS after being subjected to STA and LTA conditioning. The shift factors of W-RAMS and rejuvenated RAMS systems subjected to short- and long-term ageing never reached those of B-RAMS. This supports the previous consideration that the laboratory ageing procedure didn't fully reproduce the actual ageing occurred in the RA material.
All this is reflected in the AI parameter, whose values determined at the reference temperature of 20 °C are summarized in Figure 6. For comparison purposes, the AI values of unaged materials were also calculated and reported.

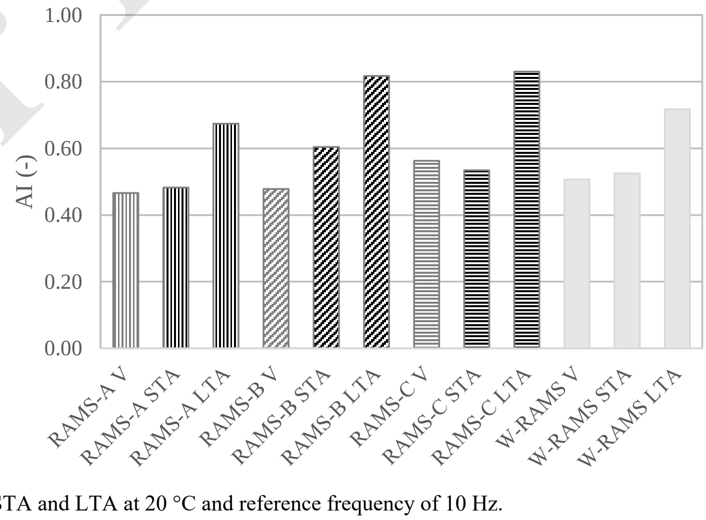


Figure 6. AI values after STA and LTA at 20 °C and reference frequency of 10 Hz.

Rejuvenated binders are usually reported to be more susceptible to aging than virgin binders [54-56]. This behaviour has been mainly observed for the bio-rejuvenators and might be due to the higher amount of oxygen in bio-oils than that of petroleum asphalt. However, other studies have reported improved aging resistance for rejuvenated binders compared to virgin binders [57].

For the materials considered in this study, the ageing index increased as the degree of ageing increased. The relative increase appears less pronounced in the case of rejuvenator A, whose AI value passed from 0.47 of the unaged state to 0.67 of the LTA state. This relative increase is comparable with that of W-RMAS containing the neat reference binder. For the other two RE products, the relative increase is higher and spans from 0.48 to 0.82 in the case of product B and from 0.53 to 0.83 in the case of product C.

This evidence is coupled with the fact that the AI values of RAMS-A after LTA conditioning are similar to or lower than those of the reference W-RAMS, whereas AI values were higher for the model systems containing the other two RE products (RAMS-B and RAMS-C). It is inferred that the restorative effects produced by rejuvenator A on the aged binder of RA made the latter less sensitive to further aging if recycled in the production of new mixtures. The opposite is true for the other two RE products, which exhibited very similar responses. as indicated by the corresponding AI values.

3.5 Interpretation of results

It is well recognized that rejuvenating agents soften the aged binder but can also modify its internal structure, by increasing the relative content of maltene fractions (resins and aromatics), and by decreasing the relative content of asphaltenes [58-60]. In fact, the recovery of the mechanical properties of binder-rejuvenator blends is commonly attributed to the restoration of the asphaltene-maltene ratio altered by oxidative ageing [20].

Depending on their chemical nature and the way they diffuse within the aged binder, some RE agents act mainly on a physical level (as softener), others interact at a chemical level (as replenisher).

In light of the results obtained from laboratory testing, it can be deduced that RAMS systems were able to capture the peculiarities of different rejuvenating products and driving mechanisms depending on the type and conditions of the test to which they were subjected.

Dynamic modulus tests proved to be suitable for evaluating the increasing softening of aged binder produced by rejuvenators as their dosage increased. Softening was evidently reflected in the reduction of stiffness and load-frequency sensitivity of the model systems. Based on the results of the dynamic modulus tests, however, no clear differences were observed between the investigated agents. This may be due to the fact that the way in which the model systems were stimulated under small oscillating loading allowed to highlight the effects produced by the rejuvenators at the physical level. The various products have similar physical characteristics in terms of density, and this may explain the similarity of their response. Specifically, a dosage of 8% yielded rheological properties very similar to those of the reference 50/70 binder for all products.

In contrast, cyclic direct tensile tests and indirect tensile strength tests made it possible to clearly discriminate between the different agents and to establish a ranking order among them. It can be assumed that the response obtained in cyclic tests was governed by the microstructural characteristics of the binder phase and the bonds that develop at the molecular scale. In these terms, the different behaviour of the blends is indicative of the different changes generated by each rejuvenator at the chemical level. The poorest performance exhibited by rejuvenator A, inferior to that of the reference neat binder, suggests that the modifications were very limited. It can be deduced that this agent acted more as a softener than as a replenisher. The superior performances exhibited by product C can be associated to an effective restoration of the components lost during oxidation and a rebalancing of the ratio between the various binder fractions. These performances also exceed those of the reference binder, leading to a general improvement of the properties of the aged RA bitumen to a level even higher than the original one. Rejuvenator B is in an intermediate condition where, however, effects at the chemical level seem to prevail over effects of pure physical nature.

Similar considerations can be drawn from the indirect tensile tests. In this case, the lower test temperature enhances the ductility of the rejuvenated systems (which is higher than those of both the aged and the reference neat binders) where the differences between the different products are attributable to the different interactions occurred at both physical and chemical levels.

4 Summary and conclusions

This paper presents the results of an experimental investigation on the use of a novel methodology for the evaluation of the effectiveness of rejuvenating agents employed in asphalt mixtures containing reclaimed asphalt. The methodology is based on mechanical testing of Reclaimed Asphalt Model Systems, constituted by a single-size reclaimed asphalt compacted to a high value of voids in mineral aggregates. The rationale behind this approach is that the response of model systems under external loadings is mainly dependent upon the bonding of the binder phase, and this allows the direct assessment of the effect of employed rejuvenator on the actual aged binder of reclaimed asphalt. Its use presents the advantage of avoiding the extraction of aged binder by solvents, thus avoiding alterations of its structure and composition. The investigation was conducted on three different rejuvenators that included two vegetable oils and a paraffinic hydrocarbon oil. The testing program involved determining the dynamic modulus, the fatigue failure and indirect tensile strength of the systems. The sensitivity to ageing effects of rejuvenating products was also investigated.
Although the results obtained from the study do not justly allow definitive conclusions to be drawn on the use of Reclaimed Asphalt Model Systems, they confirm that such systems have good potentialities as laboratory tools for assessing and ranking rejuvenating products.
Specific outcomes of the paper can be summarized as follows:

- B-RAMS and W-RMAS exhibited the highest and lowest stiffness, respectively, over the considered range of temperatures and frequencies. For each RE product, the increment of dosage added to corresponding RAMS-X reflected on the progressively decrease of its dynamic modulus. A rejuvenator dosage of 8% allowed to obtain rheological properties comparable to those of reference W-RAMS for all the considered products.
- The addition of RE improved the resistance of the aged material to cyclic loading, to an extent that depended on the type of RE product. RAMS-C exhibited the highest resistance at both strain amplitudes adopted for testing, with a relative difference in N_f values with respect to W-RAMS of about 100%. It was followed by RAMS-B (+30%) and RAMS-A (-70%).
- ITS values exhibited by the reference systems B-RAMS and W-RAMS were very close to each other (1.72 MPa versus 1.84 MPa). ITS values of rejuvenated systems were lower than those of reference ones. A clear distinction was observed between different products, with RAMS-C showing higher strength (1.04 MPa) than RAMS-B (0.83 MPa) and RAMS-A (0.63 MPa).
- The ageing index increased as the degree of ageing increased. In the case of RAMS-A, AI value passed from 0.47 of the unaged state to 0.67 of the LTA state. This relative increase is comparable with that of W-RMAS containing the neat reference binder. For the other two RE products, the relative increase was higher, spanning from 0.48 to 0.82 for RAMS-B and from 0.53 to 0.83 for RAMS-C.

Further investigations are certainly needed to better understand the mechanical behaviour of model systems, especially at intermediate and low temperatures. Moreover, the experimentation shall be extended to other RA sources to validate the concept of RAMS and to corroborate its effectiveness in comparing and ranking rejuvenating products.

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