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Assessing RAP Multi-Recycling Capacity by the Characterization of Recovered Bitumen Using DSR

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Abstract: This paper addresses the changes in bitumen properties during multi-recycling cycles, both before and after ageing. The rheology of recovered bitumen was characterized using the dynamic shear rheometer. The softening point and penetration value were also determined. The analysis showed that the bitumen's properties could be recovered even after more than one recycling cycle. The bitumen recovered from the second recycling cycle presented an average reduction of 45% in terms of complex modulus when compared with the first recycling cycle. The bitumen from the RAP mixtures presented a similar susceptibility to ageing. The analysis clearly showed that RAP has the potential to be multi-recycled.

Keywords: RAP; multi-recycling; DSR; rejuvenator; ageing; sustainability

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1. Introduction

Every year, thousands of kilometres of road are maintained and rehabilitated across the globe [1]. The pavement layers are subjected to several solicitations (traffic, climate changes and stressors that cause ageing), which create stresses and consequential damages [1,2]. Hence, maintenance and rehabilitation (M&R) interventions with the occasional replacement of layers are expected and needed to guarantee the comfort and safety of road users. These operations produce a considerable amount of Reclaimed Asphalt Pavement (RAP), which is a valuable waste. RAP recycling is a step forward in the direction of sustainable approaches promoted by recent policy goals [1,3].

RAP incorporation in new bituminous mixtures (RAP mixtures) can be associated with different techniques, rejuvenators and additives [1,4–8]. RAP mixture production and applications are increasing every year, which represent the first cycle of the RAP's life [1,9]. The circular economy approach presupposes those materials are submitted to several cycles during their life cycle without downgrading their functionality. Therefore, the multi-recycling capacity of the RAP needs to be evaluated, and only a few investigations have been carried out on this subject [7,9–11].

Aged bitumen from RAP is one of the main limitations for high RAP incorporations in bituminous mixtures due to both the unknown degree of blending and diffusion that occurs between the virgin bitumen and the RAP bitumen and the unawareness of the properties that may cause pavement cracking failures [1,6,12–18]. Several studies performed during recent years showed that the degree of blending between RAP and neat bitumen varies from no blending to full bending [6,12–16,19]. Another critical issue for the RAP bitumen is oxidation, which causes embrittlement in RAP mixtures. The oxidation occurs during the ageing process of bitumen by the loss of volatilized components (e.g., aromatic components) of bitumen. As the bitumen viscosity behavior is dependent on its chemical composition, the reduction in the aromatics percentage leads to an increase in asphaltenes, leading to a stiffness increase and a reduction in adhesion and coating ability.

Presently, there is scarce information on the characterization of the multi-recycling capacity of bituminous mixtures. The bitumen, one of the bituminous mixture constituents, is the element that is most affected by ageing effects [17,20–25]. Nonetheless, it is fundamental to understand how this valuable constituent of the bituminous mixture behaves during its entire lifespan under several recycling cycles. Usually, the ageing of bitumen is simulated by the rolling thin film oven test (RTFOT) or the thin film oven test (TFOT) and pressure ageing vessel (PAV), representing the short- and long-term ageing, respectively [11,17,19,21,22,25]. However, both methodologies did not simulate mixing between the bitumen from RAP and the new bitumen, the rejuvenator and the other components of the bituminous mixture, which can lead to changings in the bitumen binding matrix [19,26,27]. The multi-recycling cycles can be simulated through ageing methodologies on the bituminous mixture, which lead to a closer simulation of what happens in the entire lifespan from the mixing process until the end-of-life [9,10].

Cracking is one of the main distresses affecting bituminous pavements—notably, when RAP is used—and it can be divided into two types: fatigue cracking and thermal cracking [1,28–31]. Fatigue cracking occurs when the number of load repetitions exceeds the fatigue life of the pavement by considering the average temperature of the pavement layer in the field. Thermal cracking is a thermally induced stress on top of the layer, under low temperatures, which exceeds the tensile strength of the mixture. The susceptibility of the mixture to cracking depends on features such as stiffness, relaxation capability and ageing; the bitumen plays an important role in this regard [23,31–33].

There are different ways to describe and characterize bitumen. Moreover, an advanced characterization based on the Dynamic Mechanical Analysis (DMA) is required to obtain further insight into the behavior of the materials on the basis of their rheology [34,35]. Different procedures are able to run Dynamic Shear Rheometer (DSR) tests and carry out a multiple-analysis of the results, depending on the bitumen behavior or the test conditions.

The main objective of this paper is to analyze the multi-recycling capacity of RAP in new bituminous mixtures and its behavior during the entire lifespan by the characterization of the recovered bitumen. To achieve this objective, three compositions were formulated: a bituminous mixture containing virgin materials and a 35/50 neat bitumen; another containing 75% RAP, obtained from a road M&R operation, and 25% virgin material; and, lastly, another mixture containing 75% RAP, produced in the laboratory to simulate two cycles of recycling, and 25% virgin material. For all bituminous compositions, two sets of specimens were produced by applying a laboratory ageing methodology and considering another set of non-aged specimens. A vegetable rejuvenator was used for both RAP mixtures. The bitumen from all bituminous compositions was recovered and characterized.

2. Background

The rheological properties of the bitumen are normally presented in terms of the following: complex modulus (G^*), which provides the total resistance to deformation when the bitumen is subjected to shear loading; and phase angle (δ), which is the difference between the stress and strain in harmonic oscillation and is an indication of the viscoelastic balance of the material behaviour. G^* can be divided into two components: storage modulus (G') and loss modulus (G'') [34,36,37]. The rheological properties of bitumen are temperature- and frequency-dependent. These properties can be shown in the master curves of G^* and δ by the determination of the shift factors associated with temperature shifting [36,38]. The temperature dependency of the viscoelastic behavior is indicated using shift factors (α_T) (equation), i.e., the quotient between the tested frequency f and the reduced frequency at a reference temperature $\left(T_{ref}\right)$.

$$\alpha_T = \frac{f_r}{f} \tag{1}$$

The master curves are constructed by using a reference temperature to which all rheological data are shifted. A reference temperature should be defined, in which the value of α_T is equal to one. Several functions were defined to model the frequency-temperature superposition, notably: the quadratic shift function described by Pellinen et al. [39]; the Williams, Landel and Ferry (WLF) equation [40–43]; the Modified Kaelble equation [38,44,45]; the Arrhenius equation [27,46]; the Log-linear equation [36]; the Viscosity Temperature Susceptibility (VTS) equation [47]; the Laboratoire Central des Ponts et Chaussées (LCPC) approach [41]; and the Gordon and Shaw method [48]. The latter combines the use of two equations, WLF and Kaelble, for an initial estimation of the shift on the basis of parameters and standard constants. Afterwards, the fit is refined by a pairwise shifting technique and by straight lines representing each dataset [38,48]. It is preferable to use storage modulus and loss modulus isotherms, which give equal weight to the elastic and viscous components of the modulus. Each of them is shifted independently and then averaged to further refine the fit using pairwise shifting, with a polynomial representing the data being shifted. This gives a shift factor for each successive pair, which is then added to the lower temperature to obtain all shift factors from the lowest temperature. Then, the relationship between the shift factor and the temperature is used to determine the shift to the reference temperature, and the other shift factors are re-expressed in relation to this shift factor [38].

The WLF equation has been widely used to describe the relationship between α_T and temperature dependency. The modified Kaelble Equation (2) was proposed based on the WLF function, but a magnitude term was introduced in the difference of temperatures, which led to changing the overall shape from a hyperbolic curve to a sigmoidal one. Additionally, Kaelble introduced a characteristic temperature parameter—the glass transition temperature (T_d)—which divides the behavior of bitumen into two regimes, i.e., below and above this temperature. This parameter is similar to the defining temperature presented by Christensen and Anderson [34,49].

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

where C_1 and C_2 are constants to minimize the error between the measured and predicted G^* data, and T is the test temperature.

The master curve of a bitumen leads to an understanding of how the bitumen type and its chemical composition affect the viscoelastic behavior of the bitumen. This tool makes it possible to derive interpolated values of the properties of any combination of temperature or frequency within the range covered by the laboratory measurements, such as the ones obtained from DSR, which are generally conducted within the linear viscoelastic (LVE) region [39]. Furthermore, master curves can also be used to describe the rheological properties of bituminous mixtures. Different studies were performed to develop models to predict the rheological master curves. The Sigmoidal Model used in the Mechanistic-Empirical Pavement Design Guide (MEPDG) is widely used and accepted [50]. This mechanical model is represented in Equation (3).

$$\log|G^*(F,T)| = \delta + \frac{\alpha}{1 + e^{\beta + \gamma(\log f_r)}} \tag{3}$$

where $|G^*(F,T)|$ is the complex modulus as a function of frequency and temperature; $\log f_r$ is the log reduced frequency; δ is the lower asymptote; α is the difference between the values of the upper and lower asymptotes; and β and γ are defined as shape coefficients. Rowe et al. [44] proposed the use of a Generalized Logistic Sigmoidal Model (or Richards

Model) to obtain a better fit of the non-symmetric curve of the master curve. This model is represented by Equation (4).

$$\log|G^*(F,T)| = \delta + \frac{\alpha}{\left(1+\lambda e^{(\beta+\gamma(\log f_r))}\right)^{\frac{1}{\lambda}}} \tag{4}$$

where the coefficients are the same as those previously defined. The λ coefficient allows the curve to take a non-symmetrical shape. When λ decreases to one, the above equation shrinks to the standard sigmoidal function represented by the sigmoidal model.

The LVE model developed by Christensen and Anderson (CAM model) during the Strategic Highway Research Program (SHRP) A-001A [34,49] involves four parameters: the glassy modulus (G_g), the crossover frequency (ω_{cT_d}), the rheological index (R) and the steady-state viscosity (η_0). G_g is the value that the complex modulus approaches at low temperatures and high frequencies, which is normally assumed to be 1×10^9 Pa in shear loading for most bitumen. The ω_{cT_d} is the frequency at a reference temperature, where δ corresponds to 45° and is a parameter that indicates the general consistency of a given bitumen at the selected temperature, being also bitumen-specific. The R-value is the difference between the log of the glassy shear modulus and the log of the modulus in shear loading at the crossover frequency. This value is directly proportional to the scale parameter, or the shape of the relaxation spectrum, and is also related with the degree of skewness in the spectrum. The rheological index may be thought of as a shape parameter. The last two parameters are bitumen-dependent parameters and change with the hardening of the bitumen, hence allowing for the evaluation of the ageing and rejuvenation of the bitumen. In this model, both the complex modulus and the phase angle are calculated according to Equations (5) and (6), respectively. By combining the previous equations, the R-value can be calculated by Equation (7).

$$|G^*| = G_g\left(1+\left(\frac{\omega_r^d}{\omega_{cT_d}}\right)^{\frac{\log 2}{R}}\right)^{\frac{R}{\log 2}} \tag{5}$$

$$\delta = \frac{90}{\left(1+\left(\frac{\omega_r^d}{\omega_{cT_d}}\right)^{\frac{\log 2}{R}}\right)} \tag{6}$$

$$R = \frac{\log(2)\times\log\left(\frac{G^*}{G_g}\right)}{\log\left(1-\frac{\delta}{90}\right)} \tag{7}$$

In this model, the WLF equation is used for the temperature-frequency shifting.

More recently, the use of the Glover-Rowe (G-R) parameter was proposed, which is based on the functional model that describes the stress state in a low-temperature ductility test and is expressed using the LVE complex modulus and the phase angle measured in DSR [51,52]. G-R can be expressed as presented in Equation (8).

$$\text{G-R} = \frac{G^*\times(\cos\delta)^2}{\sin\delta} \tag{8}$$

This parameter is calculated for 0.005 rad/s at a temperature of 15 °C. Three limit values were defined: 180 kPa and 450 kPa, whenever damage onset and significant cracking were considered likely; and 600 kPa, whenever cracking was expected. This parameter is referred to as a durability and fatigue evaluation parameter. It was verified that the G-R parameter effectively describes the various damage levels in RAP mixtures [51,52]. This parameter allows for the assessment of the ability to develop stress via the use of a complex shear modulus and then relax stress by viscous dissipation captured via the phase angle.

As to the fatigue cracking parameter, the Strategic Highway Research Program (SHRP) proposed a limit value for the loss modulus $(G'' = G^* \sin\delta)$ that should be less than or equal to 5000 kPa at intermediate temperatures. This maximum value is considered for bituminous binders subjected to long-term laboratory ageing [33].

3. Materials and Methods

3.1. Materials

In this study, six bitumens were extracted from the three different non-aged and aged bituminous compositions as follows: one virgin mixture (0RAP and 0RAPA, respectively) and two RAP mixtures with a 75% incorporation (75RAP0, 75RAP0A, 75RAP1 and 75RAP1A) (Figure 1).

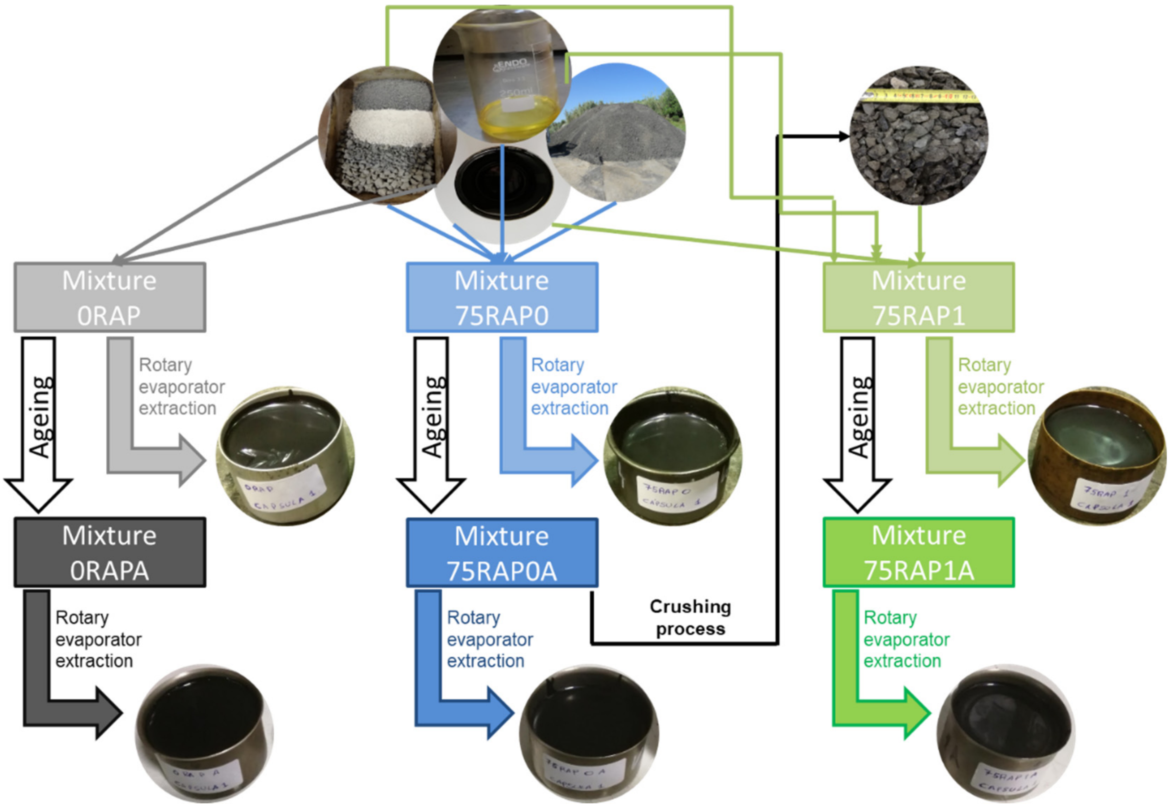

Figure 1. Schematic representation of the recovered bitumen origin.

The first recycling cycle RAP mixture (75RAP0 and 75RAP0A) was produced with RAP from a roadway M&R operation, i.e., the replacement of the surface layer, and the second recycling cycle RAP mixture (75RAP1 and 75RAP1A) was produced with RAP made in a laboratory by crushing aged slabs from the first RAP mixture (75RAP0A). Table 1 presents the softening temperature and penetration value of the RAPs used to produce the mixtures of the first (75RAP0 and 75RAP0A) and second (75RAP1 and 75RAP1A) recycling cycles. Both RAP mixtures were produced using a Crude Tall Oil (CTO) rejuvenator to recover the RAP bitumen properties. The choice of CTO was based on the fact that it is a renewable bio-based material, additionally being a by-product of the paper industry. All the compositions were produced using a neat 35/50 nominal penetration virgin bitumen. The three compositions were subjected to laboratory ageing comprising both short-term oven ageing (STOA) and long-term oven ageing (LTOA), according to the standard AASHTO R30-02 [53]. The STOA was performed on a loose mixture after the mixing process and before compaction for 2 h at 155 °C. The LTOA was applied on roller-compacted samples for 5 days at 85 °C.

Table 1. RAP bitumen properties used in the 75RAP0 and 75RAP1 mixtures.

Recycling Cycle	Softening Point (°C)	Penetration 25 °C (10^{-1} mm)
1st recycling cycle	67.5	19.5
2nd recycling cycle	68	22

The bitumen was extracted from the mixtures with the help of a centrifugal extractor and rotary evaporator equipment according to EN 12697-3 [54]. This procedure was performed at temperatures below the mixing temperature of the bitumen and for a limited period, hence adding an insignificant contribution to the ageing. Moreover, this procedure was performed equally for all the samples, and the results were comparable.

Table 2 shows the characteristics of the bitumen and of the bituminous mixtures from which it originates.

Table 2. Bitumen constituents, ageing conditions and mixture origin.

Samples	Bitumen			Mixture	
	Neat Bitumen (by Mass of Total Bitumen)	RAP Bitumen (by Mass of Total Bitumen)	Rejuvenator (by Mass of RAP Bitumen)	Ageing	Composition
0RAP	100%	0%	0%	No	Virgin aggregates
0RAPA	100%	0%	0%	Yes	
75RAP0	30%	70%	4.5%	No	25% virgin aggregates 75% RAP
75RAP0A	30%	70%	4.5%	Yes	
75RAP1	28%	72%	5.0%	No	25% virgin aggregates 75% RAP
75RAP1A	28%	72%	5.0%	Yes	

3.2. Experimental Procedures

Two tests—softening point and penetration tests—were performed according to EN 1426 and EN 1427, respectively. The penetration test evaluates the consistency of the bitumen by assessing the depth to which a needle can penetrate a bitumen under specified conditions of time (5 s) and temperature (25 °C). The softening point is viewed as an indicator of the high-temperature performance of the bitumen. The softening temperature was defined by Pfeiffer and Van Doormal [55] as the temperature at which most bitumen reach a penetration of 800×10^{-1} mm. Both measures can be correlated by the Penetration Index (I_p), (Equation (9)), which is the measure of the temperature sensitivity of the bitumen [56].

$$I_p = \frac{20 \times T_{R\&B} + 500 \times \log P - 1950}{T_{R\&B} - 50 \times \log P + 120} \tag{9}$$

where $T_{R\&B}$ is the softening point in °C and P is the penetration value in 10^{-1} mm.

The DSR tests allow for the characterization of the elastic, viscoelastic and viscous properties of the bitumen under a wide range of temperatures and frequencies (times of loading) (Figure 2). These tests can be performed in either controlled stress or controlled strain modes using different geometries. In this study, a controlled stress mode, combining two testing parallel-plate geometries, was adopted due to the wide range of temperatures tested. These were selected as shear stress and shear rate and were constant over the entire area of the plate, which simplified calculations and provided accurate fundamental rheological properties. Table 3 presents the test conditions.

(a)

(b)

Figure 2. (**a**) Dynamic shear rheometer; (**b**) Testing sample on 8 mm geometry.

Table 3. DSR test conditions.

Parameter	Configuration 1	Configuration 2
Temperature	10–40 °C (5 °C intervals)	25–80 °C (5 °C intervals)
Frequency	0.1–20 Hz	0.1–20 Hz
Geometry	Parallel plates ø 8 mm	Parallel plates ø 25 mm
Gap	2 mm	1 mm

The data from the frequency sweep tests on DSR were evaluated using RHEA™ software. The latter converts dynamic mechanical data from the frequency domain to the time domain and vice versa, hence giving the possibility of determining the master curves of stiffness from the dynamic data. The time-temperature superposition was performed by shifting the isothermal curves using a modified non-linear Marquadt–Levenburg least-squares optimization [39]. The software numerically optimizes the number of relaxation/retardation modes used in the analysis [57]. The shifting procedures use the methodology defined by Gordon and Shaw [48]. Both the CAM model and the Generalized Sigmoidal model (Richards model) were used to calculate the master curves of all the bitumen.

The evolution in the properties throughout the successive recycling cycles, before and after ageing, was analyzed by the representation of several parameters that assess the brittleness of both aged and non-aged bitumen blends (aged bitumen from RAP + neat bitumen + the rejuvenator percentage).

4. Results and Discussion

4.1. Softening Point and Penetration Value

Table 4 shows the results of the softening point, penetration value and penetration index for the six bitumen samples. In general terms, it can be observed that the bitumen recovered from the virgin mixture before (0RAP) and after ageing (0RAPA) presents a lower softening point than the bitumen recovered from both RAP mixtures. 75RAP1 and 75RAP1A showed a 4.4% and 4.1% lower softening temperature when compared with 75RAP0 and 75RAP0A, respectively. The bitumen recovered from the second recycling cycle RAP mixtures (75RAP1 and 75RAP1A) presented 60% and 50.7% higher penetration values than the bitumen from the first recycling cycle mixtures (75RAP0 and 75RAP0A), even after ageing, hence showing less viscosity. This result, together with the softening point, indicates that the multi-recycling process cannot significantly affect the consistency of the bitumen when a rejuvenator is used. The differences obtained for both aged RAP mixtures, when compared with the correspondent mixture without ageing, showed that the successive recycling cycles did not affect the ageing sensitivity. The penetration index showed an increase with ageing and throughout the recycling cycles, which seems to indicate that the temperature sensitivity of the bitumen diminished with ageing and recycling cycles.

Table 4. Softening point, penetration and penetration index.

Bitumen	Softening Point (°C)	*P* (10^{-1} mm)	Penetration Index
0RAP	55.2	31	−1.00
0RAPA	57.9	27	−0.75
75RAP0	62.7	20	−0.40
75RAP0A	67.5	15	−0.12
75RAP1	60.0	32	0.00
75RAP1A	64.7	23	0.16

4.2. Rheological Analysis

4.2.1. Complex Modulus and Phase Angle

Figure 3 presents the isochronal curves at 4 Hz of the complex modulus and the phase angle as a function of temperature. The increase in G^* and the decrease in δ show the hardening of the bitumen. Surprisingly, the 75RAP0A presented the highest value of G^* and the lowest value of δ for all the testing temperatures, followed by the 75RAP1A. For the non-aged RAP mixtures, it was possible to observe the same in the 75RAP0, which showed the highest value of G^* and the lowest value of δ. The bitumen was expected to present a tendency to hardening throughout the successive recycling cycles. However, the bitumen from the first recycling cycle RAP mixtures (75RAP0 and 75RAP0A) presented higher values of G^* and lower values of δ when compared with the bitumen from the second recycling cycle (75RAP1 and 75RAP1A, non-aged and aged, respectively). This reduction in the second cycle could be related to the higher percentage of rejuvenator used (0.5% more) when compared with the first cycle. Additionally, the rejuvenator added in the first cycle could have a contribution in this second recycling cycle. This indicates that the addition of a rejuvenator combined with a fraction of new bitumen can recover the physical properties of the aged bitumen by diminishing its viscosity. This behavior suggests a promising potential for the RAP to be multi-recycled without affecting the bitumen properties if a rejuvenator is added.

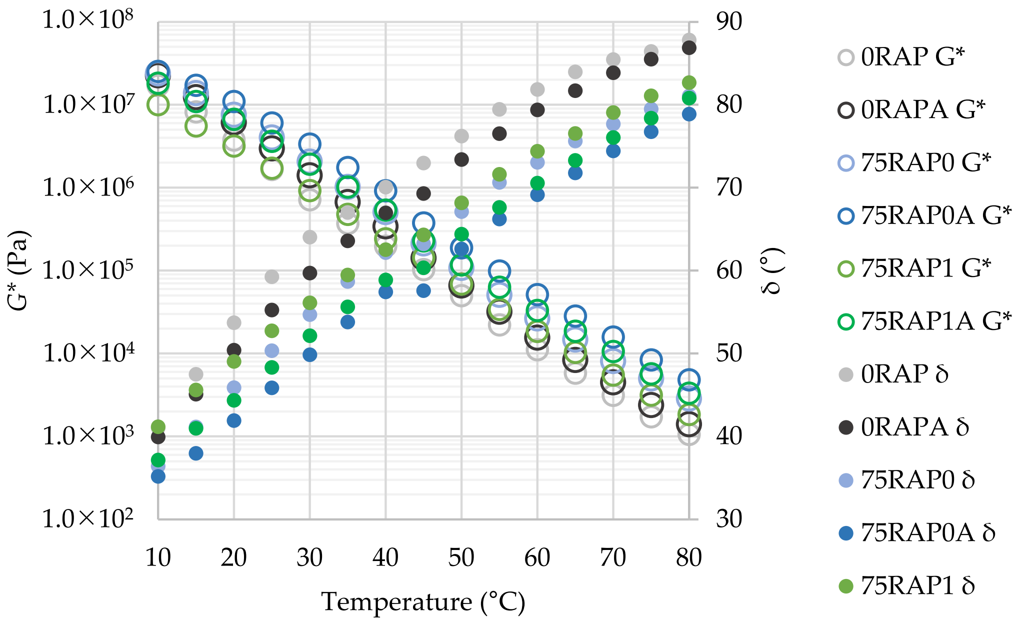

Figure 3. Isochrones at 4 Hz.

From the frequency sweep DSR tests, it was possible to assess the range of complex moduli and phase angles for each recovered bitumen (Table 5). In general, all the bitumen presented values within the same order of magnitude, in terms of the maximum value of G^*, and similar ranges, in terms of δ. By a deeper analysis, it was possible to verify that the RAP mixture from the first cycle of recycling (75RAP0 and 75RAP0A) presented the highest values for G^* when compared with the bitumen from the second recycling cycle RAP mixture (75RAP1 and 75RAP1A). In terms of phase angle, 75RAP0 and 75RAP0A presented lower ranges when compared with 75RAP1 and 75RAP1A, respectively.

Table 5. Range of complex moduli (G^*) and phase angles (δ).

Bitumen	0RAP	0RAPA	75RAP0	75RAP0A	75RAP1	75RAP1A
G^*_{min} (Pa)	3.4×10^1	5.2×10^1	1.3×10^2	2.4×10^2	7.7×10^1	1.4×10^2
G^*_{max} (Pa)	3.6×10^7	4.9×10^7	4.9×10^7	5.4×10^7	2.2×10^7	3.9×10^7
δ_{min} (°)	34.7	34.3	31.7	29.7	36.1	32.7
δ_{max} (°)	88.3	89.3	86.6	86.1	88.0	87.6

4.2.2. Master Curve Development from Dynamic Shear Rheometer Data

Black space diagrams were used to capture both relaxation and stiffness together to assess the relative cracking behavior of the bitumen. Figure 4 shows the master curves for all the studied bitumen using the shifting methodology proposed by Gordon and Shaw [48] and the Generalized Logistic Model. The high temperatures are indicated on the left (lower complex modulus and higher phase angle) and the low temperatures are indicated on the right (higher complex modulus and lower phase angle). In general, the lower complex modulus (stiffness) and the higher phase angle (relaxation capability) are expected to improve the cracking resistance of mixtures. The phase angle normally decreases with the increase in RAP content, as happened in this case. However, it was possible to verify that the phase angle did not show a tendency to decrease in the second recycling cycle, having presented a similar or higher value. Ageing also contributed to the decreasing phase angle as the bitumen hardened. For 75RAP1, the tendency to a significant decrease in the relaxation capacity was not observed when compared with 75RAP0. As observed in the softening point and penetration tests, the bitumen from the second recycling cycle RAP mixtures (75RAP1 and 75RAP1A, non-aged and aged, respectively) did not present a tendency to continuous hardening throughout the recycling cycles. With the use of a rejuvenator combined with the fraction of virgin neat bitumen, it was possible to obtain a bitumen with a lower complex modulus and a higher phase angle, which presented a decreased tendency to develop fatigue and thermal cracking. The second recycling cycle did not show negative effects on bitumen behavior.

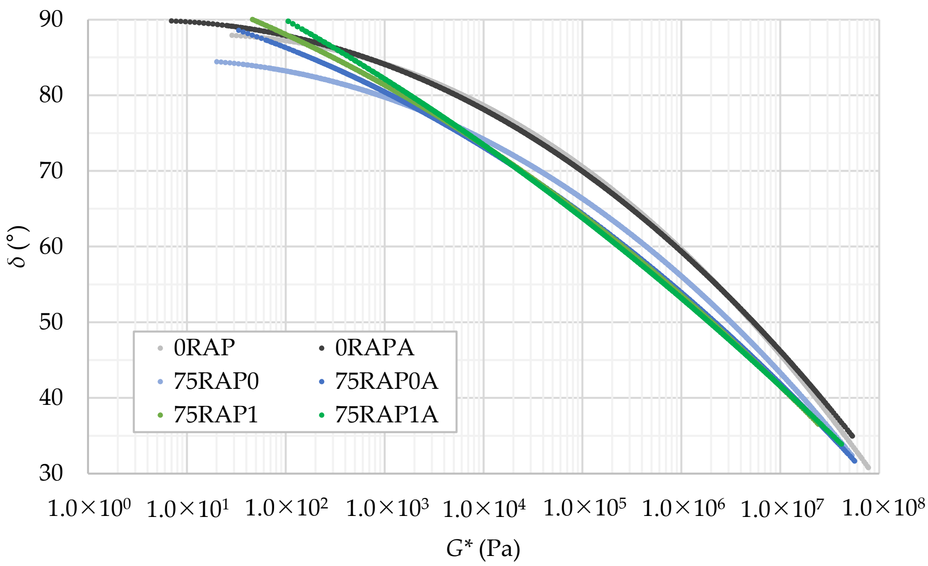

Figure 4. Black diagram.

4.2.3. Ageing Indices

The ageing indices were calculated to better understand the ageing effect on the rheological response of the neat bitumen and the bitumen blends from the RAP mixtures (Figure 5). This index was calculated according to the Equation (10). Moreover, the ageing indices led to understanding the evolution in the ageing susceptibility throughout the successive recycling cycles. The complex modulus $|G^*|$ at 4 Hz and over a range of temperatures (10 °C to 80 °C) was determined for the bitumen recovered from both the non-aged and aged mixtures. The results of the ageing indices suggest that both the virgin mixture and the RAP mixtures presented insignificant differences in terms of ageing susceptibility; the ageing index varied between 1.06 and 2.21. It was observed that the testing temperature significantly affected the determination of the ageing indices, which presented significant variations for lower temperatures (below 40 °C), and the subsequent stability. At this temperature, all bitumen presented a softer consistency due to being close to the softening point, and, consequently, an elastic behavior was observed. These conclusions were in line with the results of the penetration index, which indicated a reduced susceptibility to ageing.

$$\text{Ageing Index} = \frac{G^*{}_{Aged}}{G^*{}_{Unaged}} \tag{10}$$

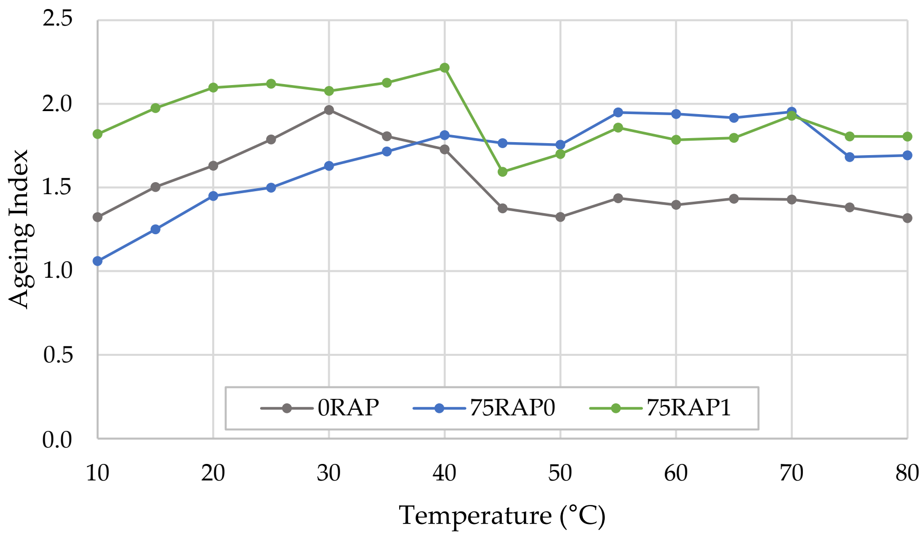

Figure 5. Ageing indices for the neat bitumen and the bitumen blend from the RAP mixtures.

The results suggest that there are insignificant differences in terms of the ageing susceptibility of the multi-recycled bitumen, which points to the viability of RAP being recycled several times.

4.2.4. Rheological Index

Figure 6 shows the R-values and crossover frequencies obtained using a reference temperature of 20 °C. In general, the aged bitumen from the RAP had a discernible effect on the increase in the R-value and on the decrease in the crossover frequency. It is possible to observe two regions: one with a variation in the R-value between 1.48 and 1.56, where the non-aged and aged virgin mixtures (0RAP and 0RAPA, respectively) present a slight increase in R-value due to ageing but also a surprising increase in crossover frequency; and another, where the bitumen from the first and the second recycling cycle RAP mixtures present higher R-values, varying between 1.85 and 2.05. In terms of crossover frequency, the bitumen from the RAP mixtures presented higher (75RAP1) or similar (75RAP0) values when compared to the non-aged mixtures, which seems to indicate that a rejuvenation took place. In the case of the aged mixtures, the 75RAP0A and 75RAP1A presented lower values of crossover frequency. This indicates that the bitumen from the RAP mixtures remains stiffer for a given degree of viscoelastic response, but it can produce this viscoelastic response at higher frequencies. It should be noted that the values obtained for the rheological index were quite similar; the total range varied between 1.48 and 2.05, a difference of 0.57 between the 0RAP and the 75RAP0A. It is known that, for very high values of *R* (*R* = 3), the bitumen is prone to develop stresses more gradually, and, consequently, when subject to repeated subcritical thermal stresses, it presents a tendency to thermal fatigue.

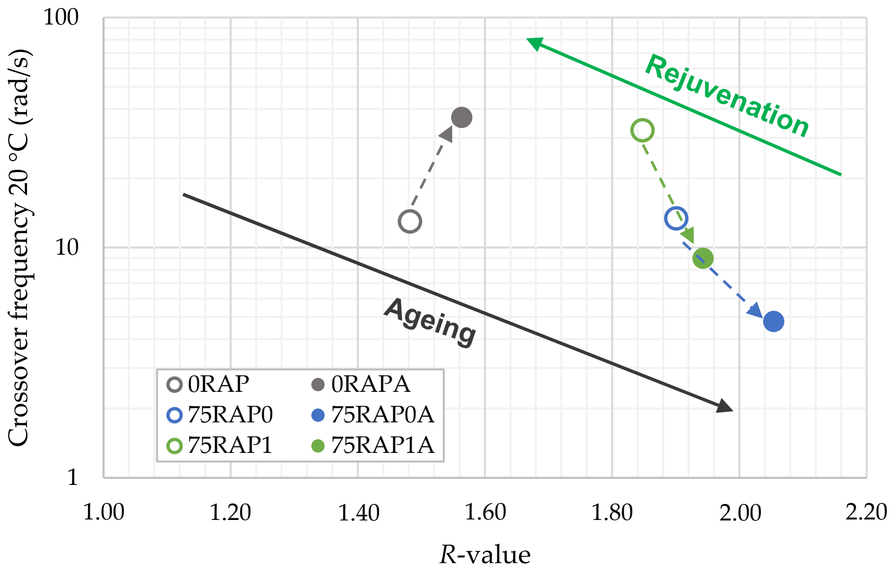

Figure 6. *R*-value and crossover frequencies.

Contrary to expectations, it is possible to observe that the second recycling cycle did not have significant effects on the bitumen blend properties. The bitumen from the second recycling cycle RAP mixtures (75RAP1 and 75RAP1A) presented lower *R*-values and higher crossover frequencies when compared with 75RAP0 and 75RAP0A, respectively. The bitumen from the second recycling cycle RAP mixtures presented lower *R*-values (2.8% and 5.4%) and higher ω_{cT_d} (141.2% and 88.0%) when compared with the bitumen from the first recycling cycle RAP mixtures, for non-aged and aged conditions, respectively.

This is an indication of the multi-recycling capacity of RAP mixtures, which depends on their rejuvenation and on the rejuvenator percentage and type defined in the mixture formulation.

4.2.5. Fatigue Criteria

Figure 7 presents the complex modulus and the phase angle plotted in Black Space for an angular frequency of 0.005 rad/s tested at a temperature of 15 °C. These results are in line with the previous ones, which suggest that the multiple recycling cycles did not significantly affect the performance of the mixture when a rejuvenator was used in each cycle. The minimum limit defined for the G-R parameter (G-R = 180 kPa) and the SHRP fatigue criteria ($G'' \leq 5000$ kPa) was broadly respected, which shows that there is no risk of brittleness and consequent cracking of the mixture, even after ageing. Moreover, the results indicated that the used dosage of the rejuvenator played an important role in the final G-R value obtained. Indeed, even for a second recycling cycle (75RAP1 and 75RAP1A), the bitumen presented lower G-R values (77.8% and 55.3%) when compared with the bitumen from the first recycling cycle RAP mixture (75RAP0 and 75RAP0A), for non-aged and aged conditions, respectively. This confirms the results obtained in empirical tests in which the bitumen recovered from the mixture of the second recycling cycle (75RAP1 and 75RAP1A, non-aged and aged, respectively) presented a lower softening temperature and a higher penetration when compared with the mixtures of the first recycling cycle (75RAP0 and 75RAP0A).

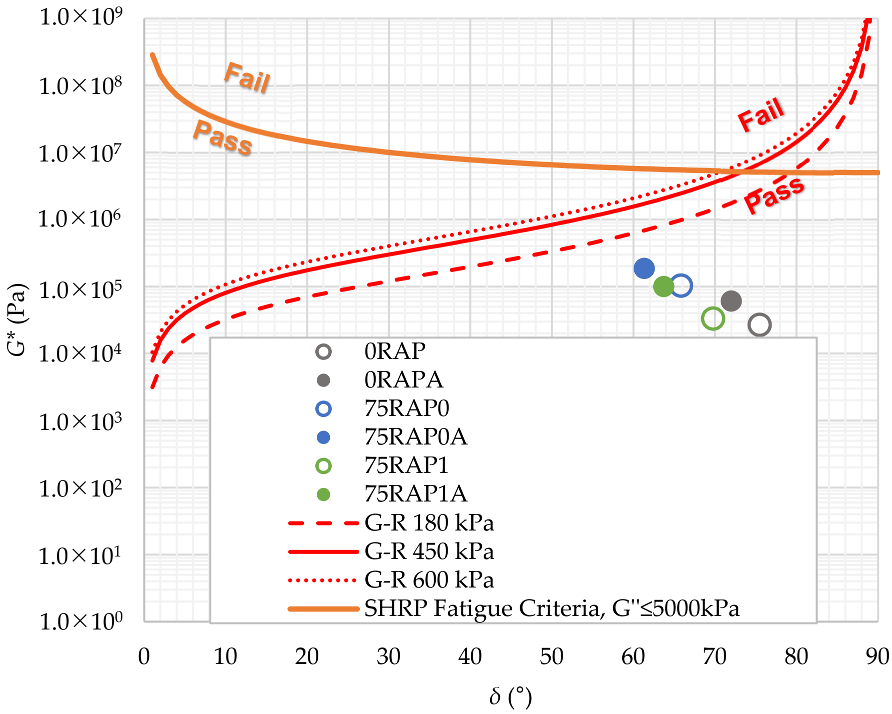

Figure 7. Complex modulus and phase angle for an angular frequency of 0.005 rad/s tested at a temperature of 15 °C.

From the empirical tests, it was possible to predict the multi-recycling capacity of the bitumen in bituminous mixtures and to understand how successive cycles of recycling could affect the bitumen viscosity. Figure 8 presents the relations between the penetration value and the softening point with the Glover-Rowe parameter (Figure 8a) and the rheological index (Figure 8b). The Figure 8a shows that, with the increase in the G-R parameter, the $T_{R\&B}$ increases and the penetration value decreases. Similar tendencies are observed for the increase in R-value.

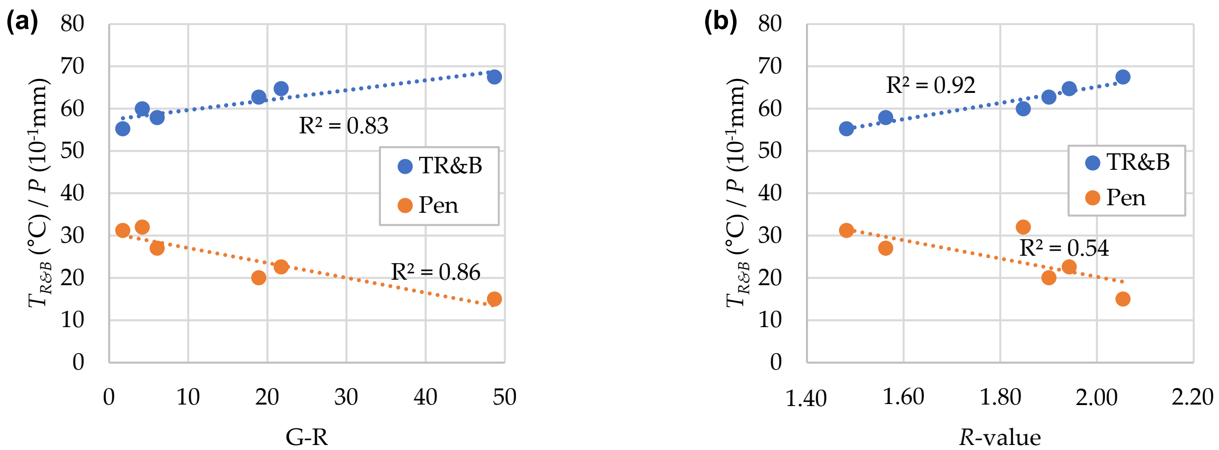

Figure 8. $T_{R\&B}$ (°C)/Pen (10^{-1} mm) as a function of: (**a**) Glover–Rowe parameter; (**b**) R-value.

5. Conclusions

The main goal of this paper was to assess the multi-recycling potential of bituminous mixtures based on the recovered bitumen behavior. This subject, although an innovative topic, is now a near-future reality due to the increasing RAP recycling in bituminous mixtures. For this purpose, the empirical properties of bitumen and the rheological properties of bitumen samples were evaluated. Six bitumen samples were recovered from the three different bituminous compositions, both non-aged and aged, as follows: one virgin mixture produced with a neat 35/50 nominal penetration bitumen and virgin aggregates (0RAP and 0RAPA, respectively); and two RAP mixtures with a 75% incorporation, one representing the first recycling cycle (75RAP0 and 75RAP0A, respectively) and the other representing the second recycling cycle (75RAP1 and 75RAP1A, respectively). Both RAP mixtures were produced using a CTO rejuvenator to recover bitumen properties and consisted of a 35/50 neat bitumen and 25% virgin aggregates. The mixtures were aged in a laboratory by short- and long-term oven ageing, according to AASHTO R30-02. The bitumen was recovered from the mixtures with the help of a centrifugal extractor and rotary evaporator equipment using toluene.

The most important conclusions that can be drawn from this study are the following:

- The bitumen recovered from the second recycling cycle RAP mixtures revealed a lower softening point (4.4% and 4.1%) and a higher penetration value (60.0% and 50.7%) than the bitumen recovered from the first recycling cycle RAP mixtures (non-aged and aged, respectively).
- The penetration index increased from −1.00, in the bitumen from the non-aged virgin mixture, to 0.16, in the second recycling cycle aged RAP mixture, which seems to indicate a decreasing ageing susceptibility throughout the recycling cycles and with ageing.
- The bitumen rheology plotted on the Black diagram showed that the successive recycling cycles did not affect the rheological performance of the bitumen. The use of a rejuvenator containing a percentage of virgin bitumen led to the obtention of a behavior that does not compromise the performance of the bituminous mixture.
- The bitumen recovered from the RAP mixtures and virgin mixtures presented similar ageing indices, which shows that the bitumen blends (aged bitumen from RAP + rejuvenator + virgin bitumen) were not prone to ageing when compared with the virgin bitumen.
- The *R*-value-ω_{cT_d} space diagrams showed that the successive recycling cycles were not translated into an increased tendency toward crack development. Furthermore, the bitumen from the second recycling cycle RAP mixtures presented lower *R*-values (2.8% and 5.4%) and higher ω_{cT_d} (141.2% and 88.0%) compared with the bitumen from the first recycling cycle RAP mixtures, for non-aged and aged conditions, respectively. This demonstrates the multi-recycling potential of the RAP.
- The results plotted on the Black diagram of the complex modulus and phase angle, determined for a frequency of 0.005 rad/s at 15 °C (G-R parameter), confirmed that the bitumen recovered from the RAP mixtures did not present a tendency to develop cracking, as the values obtained were less than G-R = 180 kPa. Moreover, it was verified that the bitumen from the second recycling cycle RAP mixtures presented lower G-R values (77.8% and 55.3%) than the bitumen from the first recycling cycle RAP mixtures, for non-aged and aged conditions, respectively. This indicates that the rheological properties of the bitumen can be recovered using a rejuvenator. The bitumen from the second recycling cycle RAP mixture and from the virgin mixture were similar.
- All bitumen presented values far from the SHRP fatigue criteria limit ($G'' \leq 5000$ kPa), hence confirming the results obtained for the G-R parameter.
- It was found that both the softening point and penetration tests presented a good correlation with the G-R parameter and that the softening point presented a strong correlation with the *R*-value. Despite the tests failing to give further information on the rheological performance of the bitumen and, consequently, on the cracking and ageing susceptibility, these tests could be used as a first indicator in practices of multi-recycling feasibility; however, rheological characterization is fundamental to an accurate evaluation.

Taking into consideration all the results obtained in this study and the evaluations performed, it was possible to conclude that the bituminous mixtures have the capacity to be multi-recycled when properly formulated. The aged bitumen, despite being the critical part of a RAP mixture, can be properly rejuvenated, and its recovered properties can present similar functions to the new ones. As RAP incorporation into new bituminous mixtures is currently carried out worldwide, the multi-recycling capacity evaluation is clearly a step forward in the circular economy for the paving industry. This evaluation must continue by the execution of tests on multi-recycled bituminous mixtures to validate these conclusions.

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