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Evaluation of swelling and degradation rates in crumb rubber modified bituminous binders

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ABSTRACT

The mutual interaction between crumb rubber (CR) and base bitumen is crucial for the stability and performance characteristics of crumb rubber-modified (CRM) binders. This study evaluates the swelling and degradation phenomena during the rubber-bitumen blending process, focusing on selecting the appropriate CR type and optimizing CRM binder production. Laboratory experiments assessed the evolution of viscosity over time for various CRM binders with differing CR types and contents. The Swelling and Degradation rates were introduced as key parameters to quantitatively measure interaction kinetics. The results indicated that the maximum swelling rate reached 160 cP/min at a 22 % CR dosage, while the degradation rate was observed at 55 cP/min. The swelling degree, quantified by the peak viscosity of 12,000 cP, was predominantly influenced by CR content. Additionally, CRM binders containing finer CR particles exhibited a faster swelling rate and lower degradation, reducing viscosity by up to 30 % post-peak. The findings of this research demonstrate that CR characteristics, including particle size and surface area, significantly impact the stability and performance of CRM binders, providing valuable insights for optimizing binder formulations.

1. Introduction

In recent years, there has been a new and increasing interest in using crumb rubber (CR) derived from end-of-life tires to modify bituminous binders due to the benefits provided in terms of reduced environmental impact and improved pavement performance [1–4].

The mutual interaction between CR and base bitumen is a key factor in the production of crumb rubber-modified (CRM) bituminous binders since it may significantly affect the homogeneity and stability of the final product [5,6].

Rubber employed in the car and truck tyre industry is usually subjected to vulcanization, a process entailing the addition of sulfur that leads to the formation of cross-links between polymer chains. As a consequence of this peculiar chemical structure, different interaction mechanisms take place between CR and base bitumen as compared to polymers commonly employed for binder modification [7,8].

In the case of Ethylene-Vinyl-Acetate (EVA) and Styrene-Butadiene-Styrene (SBS), polymer-bitumen interaction consists of a two-stage process that begins with swelling of polymer particles consequent to the absorption of the light fraction of bitumen and continues with their

progressive dispersion in the oily matrix, resulting in a final material in which the chemical structure of the original binder has been completely changed [9,10].

When blending CR with base bitumen, after an initial swelling phase similar to that of traditional polymers, the presence of cross-linked chains prevents the complete dissolution of rubber particles, thus leading to the formation of a final non-homogeneous medium [9]. In the case of long interaction periods or high interaction temperatures, the depolymerization of CR may occur, causing the release of rubber components in the bitumen [11].

CR-bitumen interaction phenomena are governed by bitumen composition, physical properties of CR (including size distribution and surface morphology), and CR dosage [12–14].

The chemical composition of a binder depends on its origin and source. Soft bitumen with low asphaltene content and high aromatic fraction shows greater interaction with CR particles than hard binders [15].

The physical properties of rubber particles are influenced by the technology adopted for CR production [1,16–18]. CRs are generally divided into ambient and cryogenic products, depending upon the

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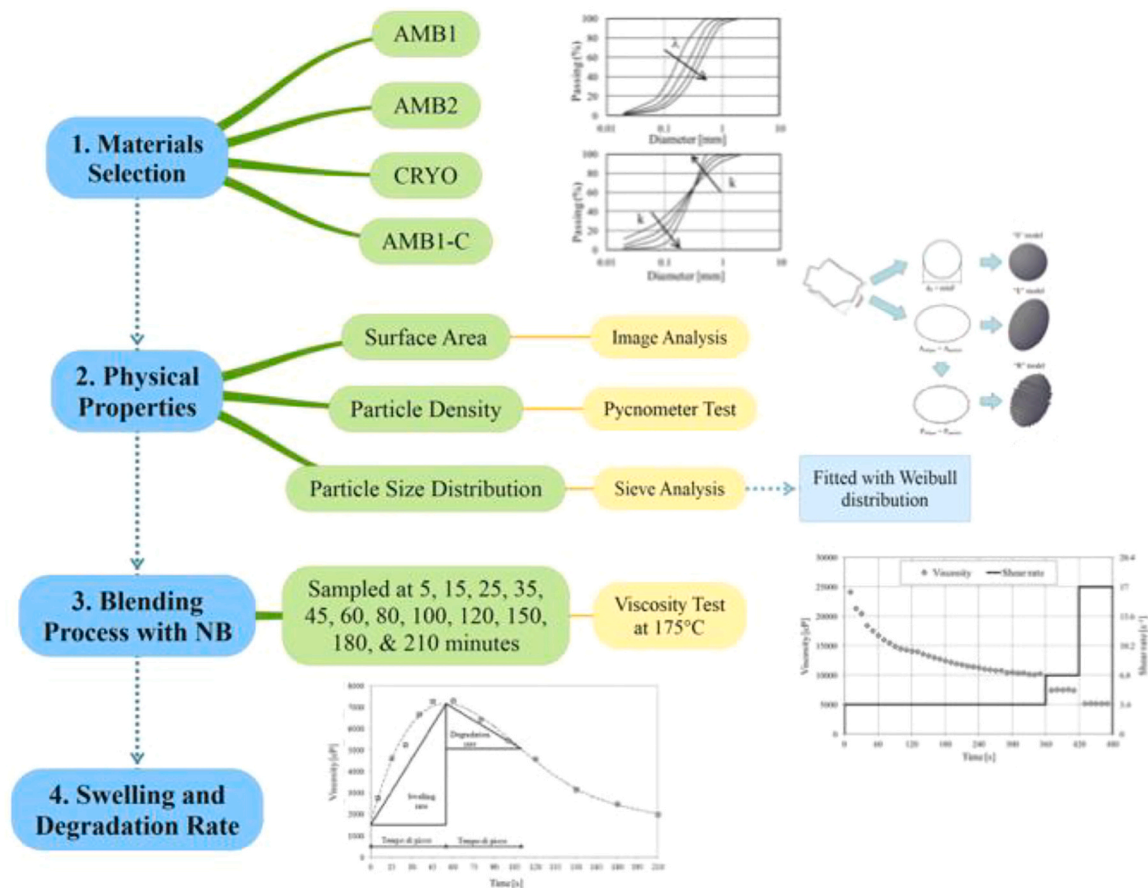


Fig. 1. Schematic of the experimental investigation.

temperature conditions adopted during milling operations on the end-of-life tires. Several studies have shown that the grinding process in cryogenic conditions results in rubber particles with regular shapes and smooth surfaces characterized by a lower specific surface area than particles with the same average size produced at ambient conditions [19,20]. CR particles with a high surface area are more prone to absorb the oily fraction of the bitumen, resulting in CRM binders with higher viscosity [13,21]

Investigations have proven that CR-bitumen interactions are also affected by temperature, duration, and mode of mixing [10,22–24]. In fact, the increase in mixing time, temperature, and shearing rate accelerates swelling and degradation processes [25]. Particle degradation improves the stability of CRM binder but produces a negative impact on its visco-elastic properties [26]. Attia and Abdelrahman [10] suggested using a high temperature (200–220 °C) and high shear rate (30–50 Hz) at the beginning of the mixing process to maximize the swelling rate of the CR particles and then reducing the temperature to 170 °C and shear rate to 10 Hz with the aim of minimize particles depolymerization. They demonstrated that the adoption of such an approach leads to a stable material with high performance-related properties due to the presence of very fine CR particles suspended in the binder.

Despite the recognized benefits of using crumb rubber (CR) in modifying bituminous binders, there are still unresolved challenges in optimizing CRM binders for consistent performance. As reported above, previous studies have often focused on general aspects of CR-bitumen interaction, but there is a lack of comprehensive understanding regarding the effects of specific CR characteristics, such as particle size, surface area, and processing methods, on the rheological properties of CRM binders. Additionally, the existing literature does not fully explore the kinetics of swelling and degradation phenomena in these materials, especially under varying blending conditions. These gaps in knowledge

hinder the development of standardized guidelines for CRM binder production, resulting in variability in product quality and performance.

The experimental study described in this paper aims to provide a further contribution to understanding CR-bitumen interaction by investigating a set of CRM binders produced with different types of CR at various dosages and blending temperatures. The experimental program included the physical characterization of CRs, and the evaluation of dynamic viscosity of CR-bitumen blends as a function of mixing time. Synthetic indicators derived from viscosity curves were introduced to assess the rate of swelling and degradation mechanisms quantitatively. The results were analyzed and discussed to highlight the synergistic roles of CR morphology and dosage on the interaction between rubber particles and base bitumen.

2. Materials and methods

The CRs employed in the experimental investigation were sampled from three end-of-life tire processing plants. Two of them adopt the ambient particle size reduction, while the third one operates in cryogenic conditions. The CRs produced through the ambient process (AMB1 and AMB2) were characterized by nominal particle dimensions comprised in the ranges 0–1 and 0–0.5 mm, respectively, while in the case of cryogenic product (CRYO), particles ranged from 0 to 0.6 mm. To highlight the influence on material response related to CR gradation of the same source, the coarser fraction of AMB1 was also used as an additional modifier (AMB1_C). This last one was obtained in the laboratory by subjecting AMB1 to sieve separation and considering only the particles retained at 0.354 mm sieve.

The selected CRs were subjected to a laboratory characterization to determine their particle size distribution, density, and surface area, as schematically represented in Fig. 1.

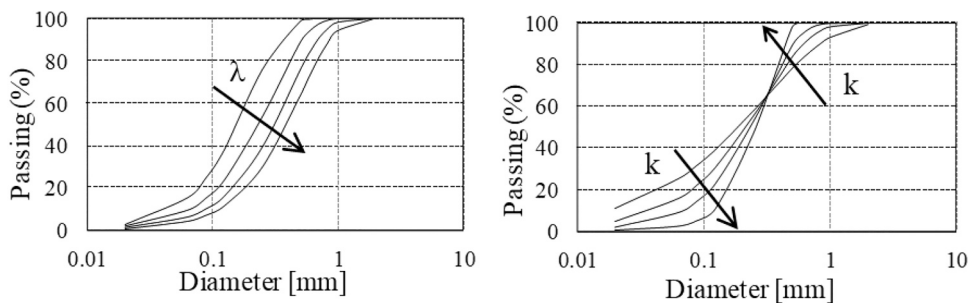


Fig. 2. Weibull distribution with λ and k parameters.

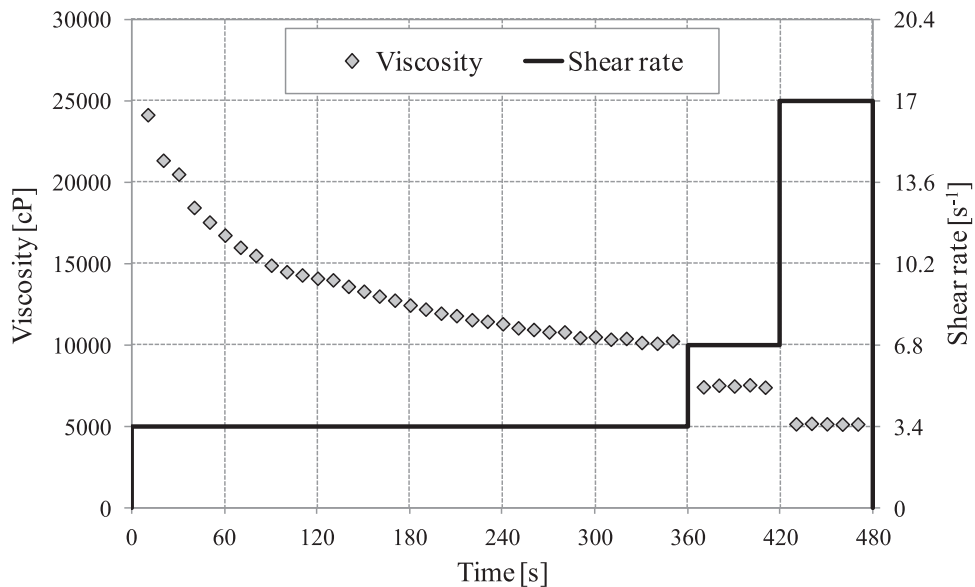


Fig. 3. Test sequence adopted for viscosity measurements.

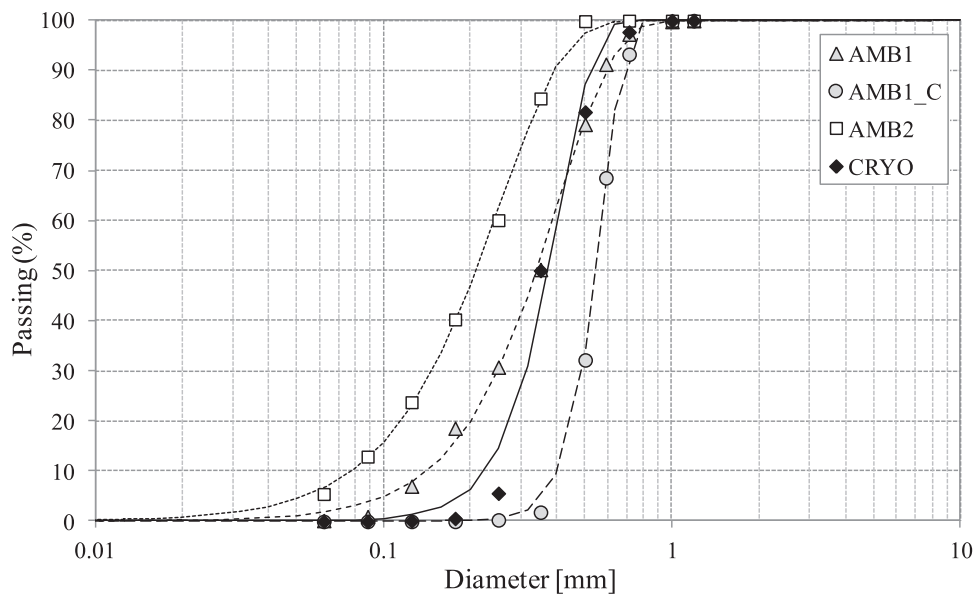


Fig. 4. Particles size distributions of the four CRs used in the investigation.

2.1. Particle size distribution

The particle size distribution of the crumb rubber (CR) samples was

determined using a standard sieve analysis method performed in dry conditions. The procedure involved the use of a series of ASTM standard sieves with the following mesh sizes: 1 mm, 0.841 mm, 0.71 mm,

Table 1
Characteristics of the four CRs used in the investigation.

CR Type	λ (mm)	k (-)	Average SA (cm ² /g)	Particle density (g/cm ³)
AMB1	0.402	2.167	221	1.180
AMB1_C	0.580	6.213	114	1.205
AMB2	0.254	1.908	386	1.196
CRYO	0.413	3.726	171	1.223

0.589 mm, 0.5 mm, 0.354 mm, 0.25 mm, 0.177 mm, 0.125 mm, 0.088 mm, and 0.063 mm. Approximately 100 g of each CR sample was placed on the top sieve and subjected to mechanical shaking for 10 minutes to ensure proper segregation of particles. The shaking was conducted using a sieve shaker with an amplitude setting that provided uniform and consistent shaking action.

After shaking, the material retained on each sieve was weighed to the nearest 0.01 g, and the mass percentage of the total sample retained on

each sieve was calculated. The cumulative percentage passing through each sieve was then plotted against the sieve size to produce a particle size distribution curve for each CR sample. The results were then fitted to the Weibull distribution obeying the following expression:

$$P_d = 1 - e^{-\left(\frac{d}{\lambda}\right)^k}$$

where d is the opening sieve (in mm), P_d is the percentage passing to the sieve with opening d, λ and k are Weibull distribution parameters. These parameters were used to describe the distribution curve, with the first one (λ) being related to the mean dimensions of particles and the second (k) being related to distribution uniformity. In particular, by referring to the qualitative example depicted in Fig. 2, it can be observed that increasing λ values correspond to increased mean particle dimension, and decreasing values of k correspond to even more uniform (well-graded) distributions.

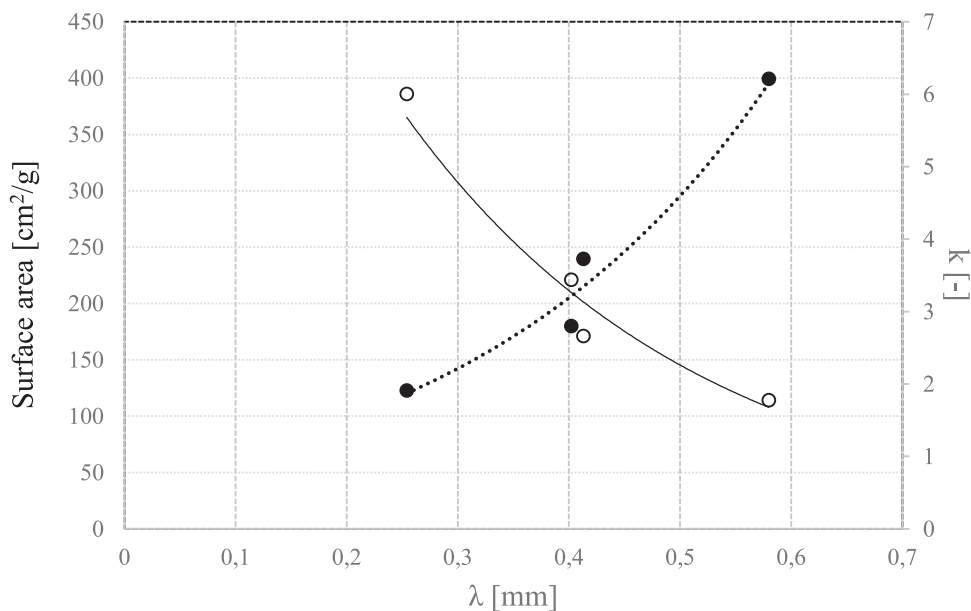


Fig. 5. Correlation between surface area and Weibull parameter λ .

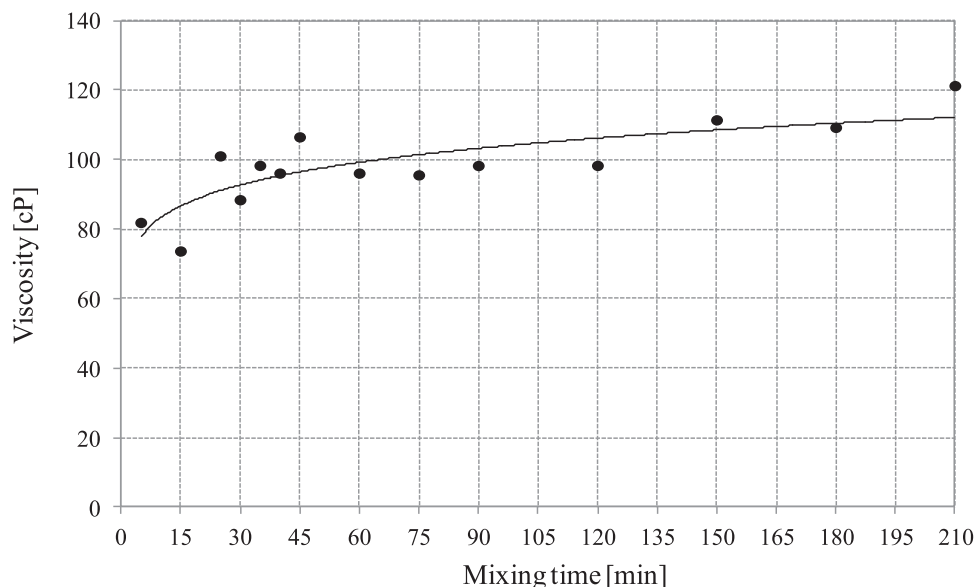


Fig. 6. Viscosity values as a function of storage time for base bitumen.

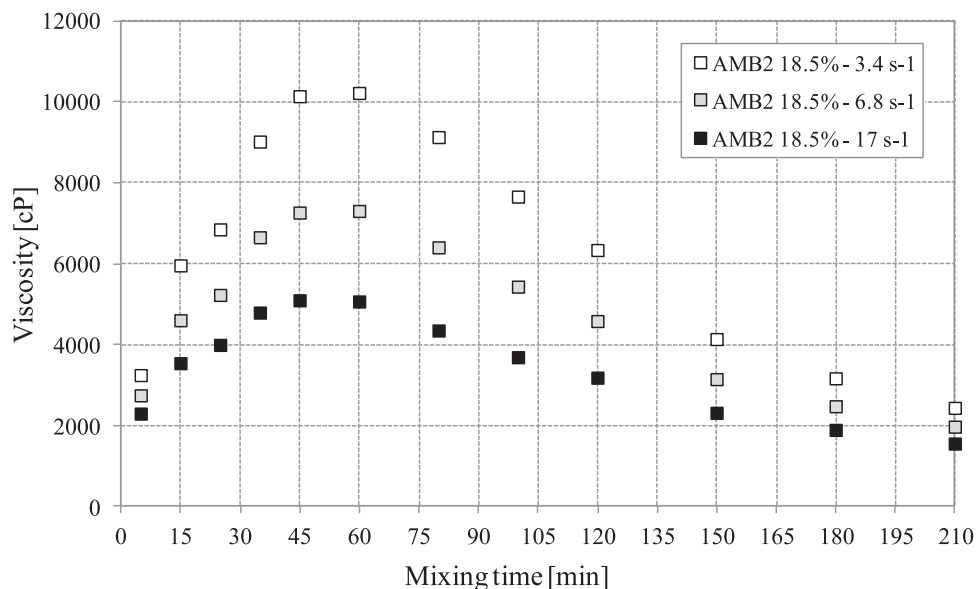


Fig. 7. Evolution of viscosity with mixing time in the case of blend containing 18.5 % AMB2 tested at different shear rates.

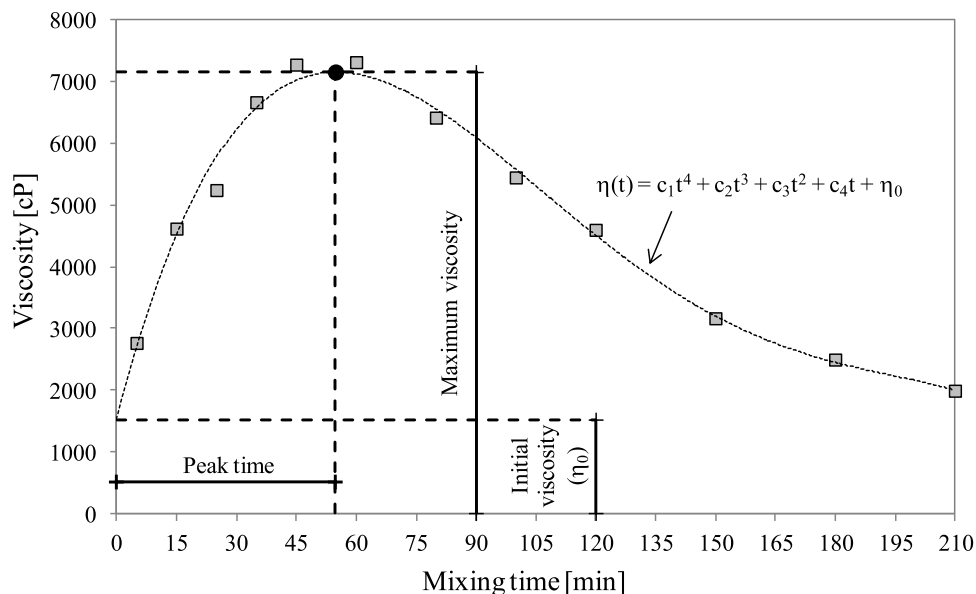


Fig. 8. Degree 4 polynomial function used to fit raw data and synthetic parameters (initial viscosity, maximum viscosity and peak time).

2.2. Crumb rubber particle density and surface area

The particle density of the crumb rubber (CR) samples was determined using the pycnometer method. This technique involves measuring the volume displaced by a known mass of CR particles when immersed in a fluid of known density. Ethyl alcohol was selected as the reference fluid due to its lower density compared to rubber, which prevents the CR particles from floating and ensures accurate volume displacement measurements.

Approximately 10 g of each CR sample was weighed and placed in a pycnometer. The pycnometer was then filled with ethyl alcohol, ensuring that all CR particles were fully submerged and no air bubbles were trapped. The mass of the pycnometer with the sample and alcohol was recorded. The pycnometer was then filled with ethyl alcohol to a calibrated mark, and the excess liquid was removed to ensure consistent volume measurement [21].

Surface area per unit mass (SA), expressed in m²/g, was assessed

based on digital images of rubber granules retained in various sieves. Images were generated using a stereomicroscope equipped with a digital camera and subsequently processed through the freeware software ImageJ. The following formula, proposed by Santagata et al. [20], was used:

$$SA = \phi \cdot \frac{6}{\rho} \cdot \sum_i \frac{f_i}{d_{m,i}} \tag{2}$$

where ϕ is a correction factor, given by the product of the shape (ϕ_f) and roughness (ϕ_r) factors, ρ is the density (in g/m³), f_i is the frequency (in decimal units) of the i -th single-size fraction, $d_{m,i}$ is the mean particle diameter (in mm) of the i -th fraction.

2.3. Crumb rubber-modified bitumen preparation and sampling

The four CRs were combined with a standard 50/70 penetration grade bitumen adopting three different dosages for each CR (15, 18.5,

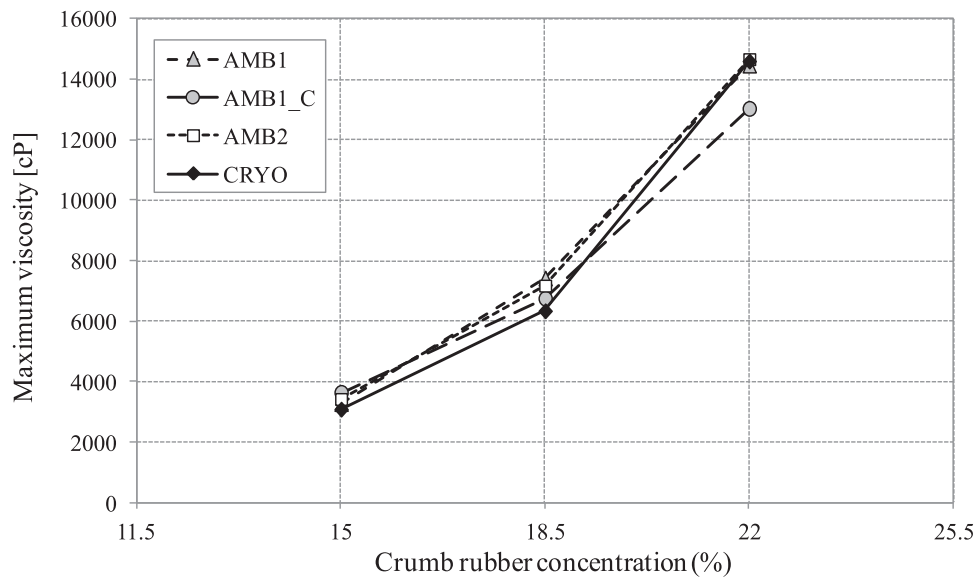


Fig. 9. Maximum viscosity values as a function of CR concentration.

and 22 % by weight of the base bitumen), for a total of twelve CRM binders. These were prepared by adding rubber particles kept at room temperature to the bitumen, pre-heated at the mixing temperature of 190 °C within batches immersed in a thermostatic oil bath. Blends were then stirred with an anchor-shaped stirrer for a total of 210 min at a rotational speed of 600 rpm.

During the mixing process, samples of materials were taken from the batch at predefined intervals (5, 15, 25, 35, 45, 60, 80, 100, 120, 150, 180, and 210 min) and immediately subjected to viscosity tests, to assess the evolution of viscosity as a function of mixing time. The viscosity has been demonstrated to be a reliable and effective indicator for evaluating CR swelling and degradation processes [27]. For comparison purposes, the described procedure was also adopted for the base bitumen stored at the same temperatures and total times of the CR-modified binders.

2.4. Viscosity test

Measurements were performed with a Brookfield viscometer (DVIII-Ultra) equipped with a SC4-27 spindle and operated at a single test temperature of 175 °C. Test sequence consisted in three subsequent stages characterized by different rotational speeds and durations: i) a first stage during which rotational speed was set at 10 rpm (corresponding to a shear rate of 3.4 s^{-1}) for 6 min, ii) a second stage in which rotational speed was increased to 20 rpm (6.8 s^{-1}) for 1 min, and iii) a final stage with a rotational speed of 50 rpm (17 s^{-1}) for an additional 1 min (resulting to a total time of 8 min for each test run). Adopting a longer duration for the first stage was dictated by the fact that materials need a certain amount of time to reach steady-state conditions.

From the example reported in Fig. 3, it can be observed that during the first stage, viscosity decreased with time, and approximately 5 min were necessary to reach a plateau value: as a consequence, only data recorded in the last minute of the testing stage were effectively considered in the analysis. Conversely, in the second and third stages, the achievement of steady-state flow was quite immediate due to higher shear rates imposed during rotation. All tests were performed in duplicate runs, and average results were considered thereafter.

3. Experimental results

3.1. CRs characteristics

Fig. 4 reports the results obtained from sieve analysis conducted on

CRs. Particle gradation curves corresponding to the different materials are clearly distinguished from each other, with AMB2 and AMB1_C showing the finest and the coarsest distribution, respectively. AMB1 and CRYO are characterized by intermediate properties, with the cryogenic product having a slightly higher amount of coarse fraction and a slightly lower amount of fine particles than the ambient one.

The Weibull distribution parameters λ and k derived from the fitting operation are summarized in Table 1, which also lists particle density and SA. As expected, AMB1_C exhibits the highest λ and k values, indicating the highest mean particle size and the least uniform distribution, followed by AMB1, CRYO, and AMB2.

Higher SA values are observed in CRs produced through ambient processes compared to those produced in cryogenic conditions. The only exception is AMB1_C, which showed the lowest SA since it is constituted by particles all coarser than 0.354 mm.

A strong relationship is also observed between SA and the Weibull parameters λ and k , as shown in the diagrams reported in Fig. 5. SA decreases with the increase of λ and λ increases with the increase of k , indicating that lower mean particle size and more uniform particle gradation result in higher surface area.

3.2. Viscosity

Fig. 6 shows the viscosity variation as a function of storage time obtained for the base bitumen tested at 175 °C. The combined effects of volatilization of the lighter weight molecules and progressive oxidation due to the exposure to high temperature during mixing (190 °C) produced the increase of viscosity all over the considered time interval. The scattering observed for data points can be attributed to the variability of test samples taken from the batch.

Adding CRs to the bitumen leads to a completely different response of the resultant CRM binders. The example reported in Fig. 7 shows the viscosity curves obtained at different shear rates for CRM binder containing AMB2 at 18.5 % dosage. Each curve is characterized by a first phase in which viscosity increases with the increase of mixing time until reaching a peak value, followed by a second phase in which viscosity decreases with the further increase of time. A similar response was detected for all other investigated CRM binders at any shear rate.

The observed viscosity evolution with mixing time reflects the CR-bitumen interaction within the material. When rubber particles are added to neat bitumen, their volume gradually rises (swelling) due to the progressive absorption of the oily fraction of binder with the

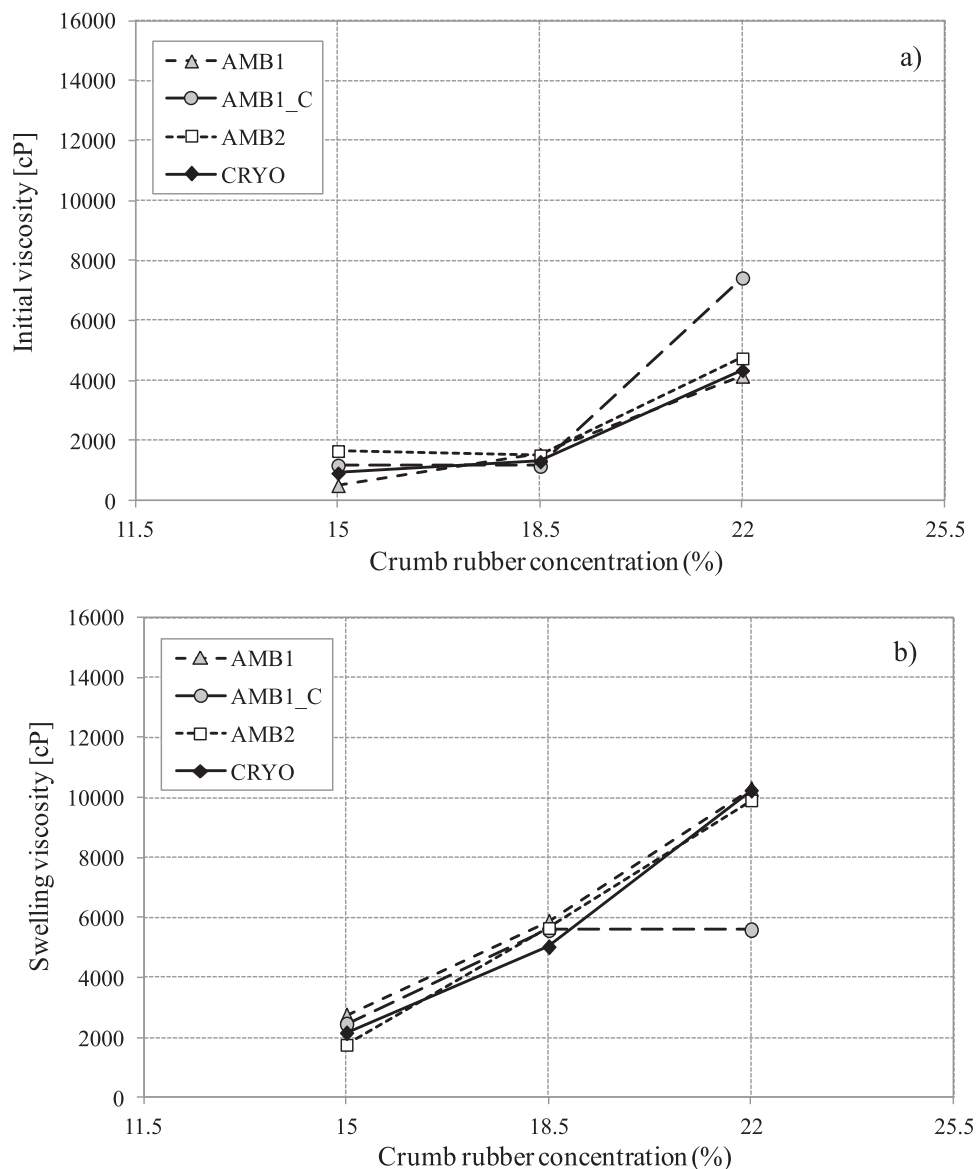


Fig. 10. Initial viscosity (a) and swelling viscosity (b) as a function of CR concentration.

consequent reduction of inter-particle distances. The stronger interaction among the particles increases the material viscosity. It has been shown that aromatics are more likely to penetrate CR particles than other bitumen components [28]. This is linked to the binding energy between CR and binder fractions, which was found to be the largest for aromatics and followed by saturated oils, resins, and asphaltenes [29, 30].

The degree of swelling depends on the capability of rubber particles to absorb bitumen, the maximum amount of which corresponds to the peak point of the curve. Afterward, the thermal and mechanical energy provided to the blend cause the partial depolymerization and devulcanization of the particles, which begin to break down within the bituminous matrix. Smaller particles result in higher inter-particle distances with the consequent drop of material viscosity.

Fig. 7 also indicates that a clear distinction can be made between the curves corresponding to different rotational speeds, providing evidence of a strong dependence on the applied shear rate of the material's response. At any given mixing time, viscosity values decrease as the shear rate increases, indicating a typical shear thinning behavior. The same outcome was found for all other materials considered in this study.

A fourth-degree polynomial function was used to fit and describe the

evolution of viscosity during blending, i.e. time, according to the following equation:

$$\eta(t) = c_1 t^4 + c_2 t^3 + c_3 t^2 + c_4 t + \eta_0$$

where $\eta(t)$ is the dynamic viscosity [cP] at mixing time t [min] while c_1 , c_2 , c_3 , c_4 and η_0 are model parameters determined from regression analysis. The values of maximum viscosity, initial viscosity, and peak time (Fig. 8) were then analytically derived and used further analyse the influence of CR dosage on CR-bitumen interactions.

The magnitude of maximum viscosity can be related to the stiffening effect introduced by rubber particles. Fig. 9 reports the values of maximum viscosity versus CR dosage obtained for the four CRM binders tested at a shear rate equal to 6.8 s^{-1} . It is observed that maximum viscosity increased with the increase of rubber concentration. Moreover, curves corresponding to different CR types are quite close to each other, revealing that the characteristics of CRs marginally influence stiffening effects. Similar results were obtained for other shear rates.

Rubber particles contribute to the overall increase of binder's viscosity in two different manners: 1) by acting as a filler and 2) by changing the structure of the composite system due to the absorption of

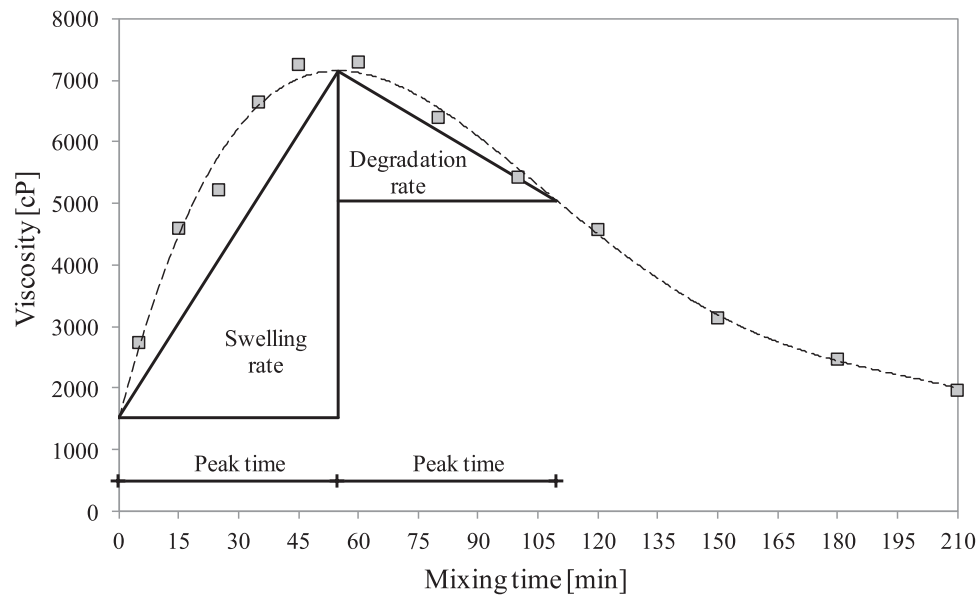


Fig. 11. Swelling and Degradation rates determined from viscosity curves.

the oily fraction of base bitumen.

Two contributions can be distinguished and quantitatively assessed by splitting the maximum viscosity value into two components: the initial viscosity and the so-called “swelling” viscosity.

The first component is calculated from the abovementioned fourth-degree polynomial functions at a hypothetical mixing time equal to zero, i.e., immediately after adding CR to the base bitumen. At this point, the swelling process has not yet been triggered, and the rubber particles still have their original shapes and dimensions; it follows that initial viscosity is evidently related only to the bulking effect of CR. The second component is given by the difference between the maximum and the initial viscosity. It indicates the maximum increase of viscosity reached during the first phase of blending and can be assumed to measure the overall degree of swelling in the CR-bitumen blend.

Fig. 10 reports the initial viscosity and swelling viscosity values as a function of CR concentration for the materials investigated. It is noticed that initial viscosity shows a slight growth when passing from 15 % to 18.5 % concentration. This indicates that particles are still diluted into the bitumen and cause minor effects on CR-bitumen blend properties. Conversely, particles become more concentrated and packed at the highest dosage (22 %), resulting in a more marked reinforcement of the blend. Swelling viscosity is more sensitive to CR dosage variations, as denoted by an almost linear increase observed for all binders. The only exception is represented by blends containing AMB1_C, for which no substantial differences are observed between 18.5 % and 22 % concentrations. The presence of a plateau suggests that for this type of CR, the quantity of particles corresponding to 18.5 % by mass can adsorb all the available aromatic fraction of the base bitumen and that any supplementary amount of CR contributes in terms of bulking effect only. It is finally observed that the curves of the remaining CRM binders are very close to each other, indicating that swelling viscosity was mostly governed by CR dosage rather than CR characteristics for these materials.

3.3. Swelling and degradation rates

The analysis of viscosity curves revealed that CR-modified binders might reach their maximum viscosity at very different mixing times and that a drop in viscosity after the peak point may occur at very different rates. Therefore, peak time must be considered for a comprehensive analysis of CR-bitumen interaction and its kinetics.

To this purpose, two parameters, namely “Swelling rate” (SR) and “Degradation rate” (DR), were introduced. As illustrated in Fig. 11, SR is

given by the ratio of swelling viscosity to peak time and DR by the ratio of viscosity loss after peak time to peak time.

Fig. 12 illustrates the relationship between these parameters and CR content, showing that both rates increase as CR content grows in all cases. This indicates that in the presence of a higher amount of particles, the available oily fraction of the bitumen is absorbed in a shorter time, and the subsequent dissolution of swollen particles occurs more rapidly. Data points also reveal a significant difference in the occurring speeds between swelling and degradation phenomena: the SR is almost three times as big as the DR for each CRM binder.

Further observations can be drawn from the comparison of the various materials. At the lowest CR content (15 %), blends containing different CRs exhibited very similar values for both parameters. At increased dosages (18.5 % and 22 %), the difference between CRM binders becomes progressively larger, especially in the case of SRs. In particular, binders containing AMB1 showed the fastest swelling and degradation, followed by those containing AMB2, AMB1_C, and CRYO. For the latter, however, SRs and DRs of the maximum CR-dosage exceeded those of the AMB1_C blend, with values that are very close to those of the blend containing AMB2. Such outcomes confirm that the type of CR used to produce rubberized bitumen plays a significant role in the kinetics of swelling and degradation processes.

To highlight the role of CR physical properties on the interaction mechanisms, SRs and DRs were correlated with particle-specific surface area SA and the Weibull distribution parameters λ and k . Fig. 13 shows that for lower CR dosages, SRs and DRs are not dependent (CR 15 %) or slightly dependent (CR 18.5 %) from the surface area of the rubber particles; conversely, variations of rates with variation of SA appear much more noticeable at the highest dosage (CR 22 %). In this last case, the bell-shaped curves indicate an initial increase in SRs and DRs with the increase of SA, up to a threshold beyond which a further increment of SA results in the progressive decrease of both parameters. Due to the strong correlation between SA and Weibull distribution factors, similar trends were found by plotting SRs and DRs as a function of λ and k .

It can be argued that greater surface area combined with more uniform particle size distribution promotes, to a certain extent, the absorption of the oily fractions of bitumen. Rubber polymer chains absorb more bitumen molecules, resulting in a higher swelling degree and faster swelling [31]. Swollen particles, in turn, degrade more rapidly due to an intensification of bond-breaking and particle breakage. However, at very high SA values (and corresponding very low λ values), the mean dimension of rubber particles becomes too small, thus hampering the

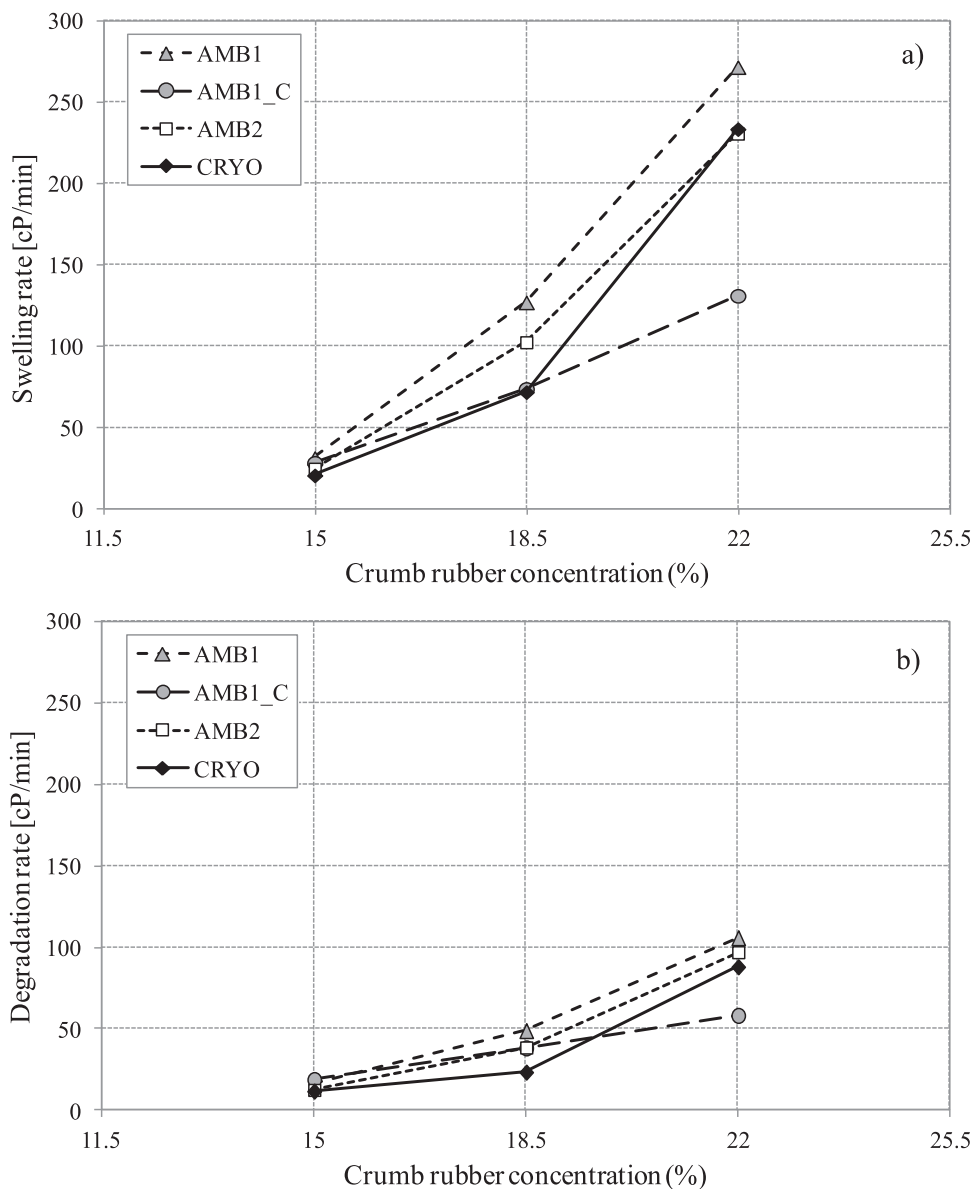


Fig. 12. Swelling rate (a) and Degradation rate (b) versus CR concentration.

mechanisms described above. This is reflected in a deceleration of the bitumen absorption process and subsequent degradation of CR particles.

It follows that the control of CR characteristics may represent a key subject in producing CRM binders with high CR content. In the specific case of materials investigated in this study, even though similar swelling degrees were recorded for blends at 22 % CR dosage, it was demonstrated that their swelling rates were significantly different due to different mean particle sizes and gradation of CRs employed for modification.

4. Summary and conclusions

The laboratory study investigated a set of CRM binders to evaluate the influence of CR characteristics and concentrations on rubber-bitumen interaction mechanisms. Viscosity tests conducted at increasing blending times revealed that the degree of swelling in rubber-bitumen blends was primarily governed by CR content, with a maximum swelling rate observed at 160 cP/min at a 22 % CR dosage. The properties of CR, including surface area, mean particle size, and gradation, significantly affected the kinetics of swelling and degradation

phenomena, particularly at higher CR concentrations. For instance, CRM binders containing ambient-processed CR with higher surface areas exhibited faster swelling rates and lower degradation, as indicated by a viscosity reduction of up to 30 % post-peak, where peak viscosity reached up to 12,000 cP.

Specific conclusions from the study include:

- CRs produced using ambient processes exhibited higher surface areas (e.g., AMB2 with 386 cm²/g) compared to those produced in cryogenic conditions (e.g., CRYO with 171 cm²/g). Ambient CRs also displayed a higher mean particle size and less uniform gradation, characterized by Weibull parameters $\lambda = 0.254$ mm and $k = 1.908$ for AMB2. These characteristics significantly influenced the swelling and degradation kinetics in CRM binders.
- A bell-shaped relationship was observed between surface area and the rates of swelling and degradation at higher CR dosages (22 %), with an initial increase followed by a decrease as particle size reduced. This indicates a complex interplay between particle size and surface area in affecting binder properties.

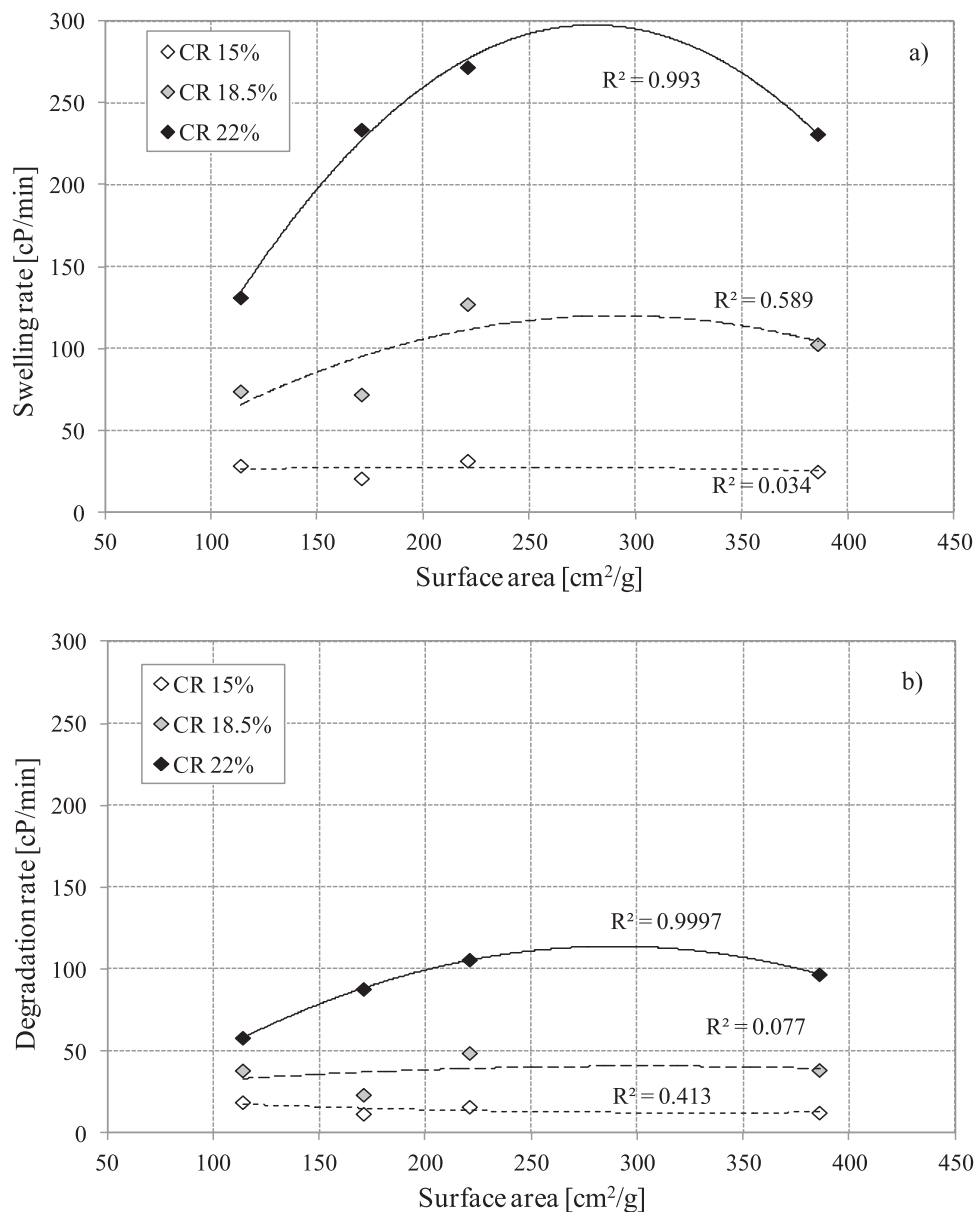


Fig. 13. Swelling rate (a) and Degradation rate (b) as a function of particles surface area.

- The viscosity evolution demonstrated a strong dependence on the applied shear rate, with increased shear rates (e.g., from 3.4 s^{-1} to 17 s^{-1}) leading to higher viscosities at any given mixing time. This shear-thinning behavior was consistent across different CR types and concentrations, underscoring the importance of controlling processing parameters to achieve desired binder performance.
- The degree of swelling, associated with the peak viscosity value, was predominantly influenced by CR content. The swelling viscosity component increased from 8000 cP at 15 % CR to 12,000 cP at 22 % CR, while the CR type had a relatively marginal influence. Swelling rates were observed to reach up to 160 cP/min, while degradation rates were up to 55 cP/min, particularly at the highest CR content (22 %).
- The physical characteristics of CRs, particularly surface area and particle size distribution, significantly influenced the kinetics of interaction phenomena. Smaller, more uniformly graded particles (higher k values and lower λ values) enhanced the absorption of the oily fractions of bitumen, leading to higher swelling degrees and faster degradation.

Further investigations should explore a broader range of CR materials to validate these findings and refine CRM binder formulations. Additionally, laboratory results should be corroborated with CRM binders produced in actual manufacturing plants to ensure practical applicability. Finally, future studies should focus on creating a link between the evolution of swelling and degradation phenomena with the chemo-structural properties of the CRM binders.

CRediT authorship contribution statement

Ezio Santagata: Writing – review & editing, Supervision, Methodology, Conceptualization. **Michele Lanotte:** Writing – original draft, Methodology, Investigation, Conceptualization. **Orazio Baglieri:** Writing – review & editing, Investigation, Data curation.

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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