Quality control of manufacturing and hot storage of crumb rubber modified binders

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HIGHLIGHTS

- The dual helical ribbon (DHR) guarantees sample stability while CRM binders are tested.
- Real-time monitoring provides control of CRM binders' rheological properties during manufacturing and hot storage.
- The procedure allows to provide recommendations for optimal hot storage.
- The procedure introduces parameters to establish targets for improved design and quality-control of CRM binders.

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The ultimate performance of crumb rubber modified (CRM) binders is linked to the accurate control of the properties during manufacturing and hot storage. However, due to their complexity, asphalt technologists find the characterisation of these materials still challenging. In this study, the adoption of a Dual Helical Ribbon (DHR), a novel mixing/measuring device for rotational viscometers, is proposed for the real-time monitoring of CRM binders during manufacturing and hot storage. According to the laboratory results, manufacturing periods of 45–60 min at 195°, as well as storage temperatures not exceeding 150 °C, are recommended for this type of modified binders.

1. Introduction

The European countries have established management systems to organise the collection of used tyres and promote their valorisation in several applications [1]. However, tyre rubber is engineered to be chemically very stable, durable and suffers biodegradation slowly. For the same reason, reclaiming natural rubber from waste tyres thorugh devulcanization is not that easy, hence the natural application of reusing tyre rubber for new tyre is not a straight-forward option. The necessity of re-using tyre rubber as secondary material in other application has therefore arisen and a very succesfull application has been incorporating the crumb rubber modifier (CRM) into asphalt mixtures. In fact, the use of CRM in asphalt applications provides a solution to improve pavements performance [2] including the reduction of the tyre/road noise when applied in wearing course [3]. The use of the ground rubber in asphalt paving materials can play a notable role in reducing the landfill of used tyres.

The enhanced engineering properties obtained by using these modified binders in different asphalt applications are well documented. The rutting resistance of rubber-binders is superior to that of the pure bitumen before being modified with crumb rubber [4]. At intermediate temperatures, the CRM binders display a reduced stiffness [5] and even at low temperatures, as is the cases of Russia, this family of materials have shown good performance [6].

Nevertheless, the properties of CRM binders depend on a number of variables, including not only the proportions and nature of the components (bitumen and rubber) but also the parameters of the manufacturing procedure (temperature, mixer type and mixing time), with significant influence on the viscosity and the rheology of the final product [7,8]. In this regards, the increasing of the CRM content usually turns into more modified binders and higher viscosities, as reported by Lee et al. [9]. The same authors report that the CRM produced by ambient grinding generally increases the viscosity more than the same content of cryogenically ground CRM, because of the shape of the rubber particles and their larger specific area. Thodesen et al. [10] conclude that cryogenic crumb rubber yield lower level of bitumen modification and proves it via scanning electron microscope (SEM) images of bitumen modified with both cryogenically and ambient ground CRM particles. On the other hand, the high temperatures and prolonged mixing times foster rubber-bitumen interaction but, excessive temperatures, above 200 °C, can degrade the quality of the CRM binder [11].

To understand better the influence of these variables on the CRM binder it is necessary to consider the rubber-bitumen interaction mechanisms, as they explain how the components and the parameters of the manufacturing procedure can determine the ultimate properties of the modified binder. The rubber is affected in two ways: swelling of the rubber particles and a progressive degradation by depolymerisation and devulcanization [12]. The swelling of the rubber particles can reach a ratio of 250% as a result of the interaction until saturation with the oily fractions of the bitumen [13]. Lopez-Moro et al. [14] investigated the interchange of aromatic oils between the rubber particles and the bitumen and determined a high concentration of this asphalt fraction around the rubber particles via fluorescent microscopy. In the same way, they concluded that some portions of the rubber particles, including black carbon in their composition, were delivered to the bitumen. At low manufacturing temperatures (below 160 °C) the primary mechanism is the absorption of oily fractions by the rubber particles [15] as shown by the results of the thermogravimetric analysis.

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Moreover, according to the results of the viscoelastic study, the increased interaction parameters results in the delivery of portions of rubber into the bitumen, modifying its rheology. The importance of the processing conditions has also been insightfully described by Lo Presti et al. [16] making clear that CRM binder overcomes an evolution of properties that, in terms of viscosity, can be summarised as follows. Phase I starts with a predominant role of the swelling of the rubber particles due to the absorption of the oily fractions of the bitumen, producing a progressive increase of the viscosity. During phase II, although the swelling phenomena slow down, the maximum value for the viscosity is achieved because the oily fractions continue diminishing until the saturation of the potential of the rubber particles to capture light components of the bitumen. And finally, during phase III, if the temperature is high enough, the viscosity starts decreasing along time because of the depolymerisation of the rubber. Obviously, in technical terms, phase III should be as minimised as possible to prevent the loss of quality of the modified binder.

The objective of limiting the degradation of the CRM binder should be easy to achieve by controlling the storage time and temperature. Nevertheless, at this point, a new concern arises, the poor storage stability of these modified binders. After the manufacturing of the CRM binders, it is necessary to store them in tanks at high temperatures, until the modified binder is used in the production of asphalt mixtures. As the rubber particles are prone to settlement as a solid in suspension in the liquid phase (bitumen), the tank has to be provided with a stirrer to keep the rubber particles in suspension. Several attempts have been carried out to provide the CRM binders with enhanced storage stability by diminishing the size of the CRM particles [17], by adding sulfur [18] or reactive polymers [19]. But the problem has not been satisfactorily solved, and the stirring of the stored rubber binders continues being necessary at industrial scale.

Nevertheless, the stirrer in the tank during prolonged storage periods, beyond preventing decantation, could promote the third phase of the rubber-binder interaction, this is, the depolymerisation of the rubber an so the loss of quality of the stored CRM binders. The increase of the viscosity by increasing the temperature and the mixing time, as proposed by Hosseinnezhad [20], is a stringent rule only in some ranges of temperature and processing time. In experiments prolonged during 8 h, the increase of viscosity reaches a sort of horizontal asymptote over time [21]. This lack of progression in the evolution of the viscosity could be referred to the depolymerisation of the rubber that counteracts the hardening due to the capture of oily fractions and the ageing of the binder because of the high mixing temperature. In line with this result, a study by Ragab and Abdelrahman [22] showed that the separation index during resting periods of 8 h decreases as the mixer speed goes from 10 to 50 Hz in the manufacturing procedure. The authors interpreted this observation as the evidence of the extensive depolymerisation and degradation of the rubber particles along time when the interaction parameters increase. The depolymerisation destroys the rubber particles and makes them less prone to suffer settlement.

Because of the above reasons, it should be necessary to estimate the conditions for the storage period to prevent or minimise the loss of viscosity of the CRM binders during prolonged storage times. Since the tank stirrer is conceived to avoid the settlement of the rubber particles, the foremost parameters to be defined are the storage time and temperature.

Pais et al. [23] carried out a study of the evolution of the viscosity and the rheological parameters of CRM binders during processing times of 0.5, 8, 24, 30 and 40 h. The study included several percentages of ambient and cryogenically ground crumb rubber. The authors concluded that the viscosity slowly decreases during prolonged period times for the binders modified with ambient crumb rubber. Nevertheless, the cryogenic rubber achieves lower viscosity values although no decreasing was observed during the 40 h. Probably the first limitation of this investigation could be referred to the fact that the samples were collected at proper times and prepared for testing, instead of continuously monitoring the properties of the CRM binder.

Some attempts have been performed to achieve a procedure for the monitoring of the viscosity of the binders in real-time in the laboratory. Celauro et al. [24] tried to do it with a standard Brookfield spindle #27. The device was conceptualised as a low shear mixer. the Brookfield equipment. Nevertheless, the authors observed that the procedure was affected by the settlement of the rubber particles. To overcome this inconvenience, Lo Presti et al. [25] developed and calibrated a dual helical impeller to substitute the conventional spindle. The new device was able to keep the rubber particle continuously in suspension measuring the viscosity of the CRM binder at the same time. The new tool and the procedure has been checked and improved with a Computational Fluid Dynamics model [26].

The present study aims to introduce an improved quality control procedure for CRM binders aiming at assessing their viscosity development during a laboratory simulation of their manufacturing and stirred hot storage. The procedure developed in this work consists in adapting a rotational viscometer to act as a low shear mixer/storage tank with a modified dual helical ribbon impeller (DHR) previously designed and calibrated [27]. With this setup, several processing variables are investigated (types, contents, sizes of CRM and different storage times and temperatures) to withdraw practical recommendations.

2. Materials and methods

The experimental programme showed in Fig. 1 was tailored to evaluate the effect of the processing variables affecting the CRM binders rheology during the product manufacturing and the hot storage. Eight different binders were included in the experimental programme: four modified binders were produced by adding 10% and 18% by bitumen weight of fine CRM size (125-250μ) in 50/70 grade bitumen and four by adding 10 and 18% of raw crumb rubber size (125-800μ). This programme was undertaken by using both ambient and cryogenic CRM. The proposed laboratory simulation for the manufacturing and agitated storage of CRM binders was performed by manufacturing the above-mentioned blends at 195 °C and simulating the storage at different temperature (195 °C, 180 °C, 165 °C, 150 °C). Each of the reported results was obtained as the average of at least two replicates. It has to be noted that, in this investigation, two Brookfield DV-II PRO Digital Viscometer were used: a low torque (LV model) and high torque (HA model). The two equipment differ mainly for the viscosity measurements range and as a consequence, they offer different accuracy of the measurement. In fact, torque measurement accuracy specified by the manufacturer is 1% of the full-scale range, with the LV model designed for low viscosity fluids (100% torque = 673.7 dyne cm), while the HA model is more appropriate for medium–high viscous fluids (100% torque = 14,374 dyne cm). An overview of the experimental programme is provided in Fig. 2.

2.1. Laboratory simulation of manufacturing and hot storage

According to Caltrans [28], rheologists have used the high-temperature viscosity of rubberised binders in the range 100–200 °C to ensuring the proper pumpability and compactability of the mixtures but also to guarantee adequate coating of the aggregates. Usually, this exercise is carried out in the field with hand-held viscometers or more typically in the laboratory by using a rotational viscometer with coaxial cylinders testing geometries. The dynamic/rotational viscosity is, therefore, a key engineering parameter that asphalt technologists need to control and target the CRM binder during product development. Hence, real-time monitoring of the viscosity of rubberised binders as a function of processing time and temperature provides asphalt technologists with a better understanding of the actual physical change throughout the processing of the material and overall allows higher confidence during product development and quality control. Furthermore, adapting a rotational viscometer as rheo-mixer offers the chance to study the change in material rheology with different base bitumens, or different crumb rubber content, by drastically reducing the material and time consumption.

On this regard, recent studies [24,25,27] have successfully demonstrated the concept of adapting a rotational viscometer as a low shear mixer for CRM binders. In all these studies, viscosity measurements were carried out according to international standards on viscosity measurements of rubberised binders (ASTM D4402 [29], ASTM D7741M [30]), but they all presented specific peculiarities: Celauro [24] proposed a procedure where the real-time monitoring was undertaken by using conventional spindles, Lo Presti [25] introduced the use of improved test-

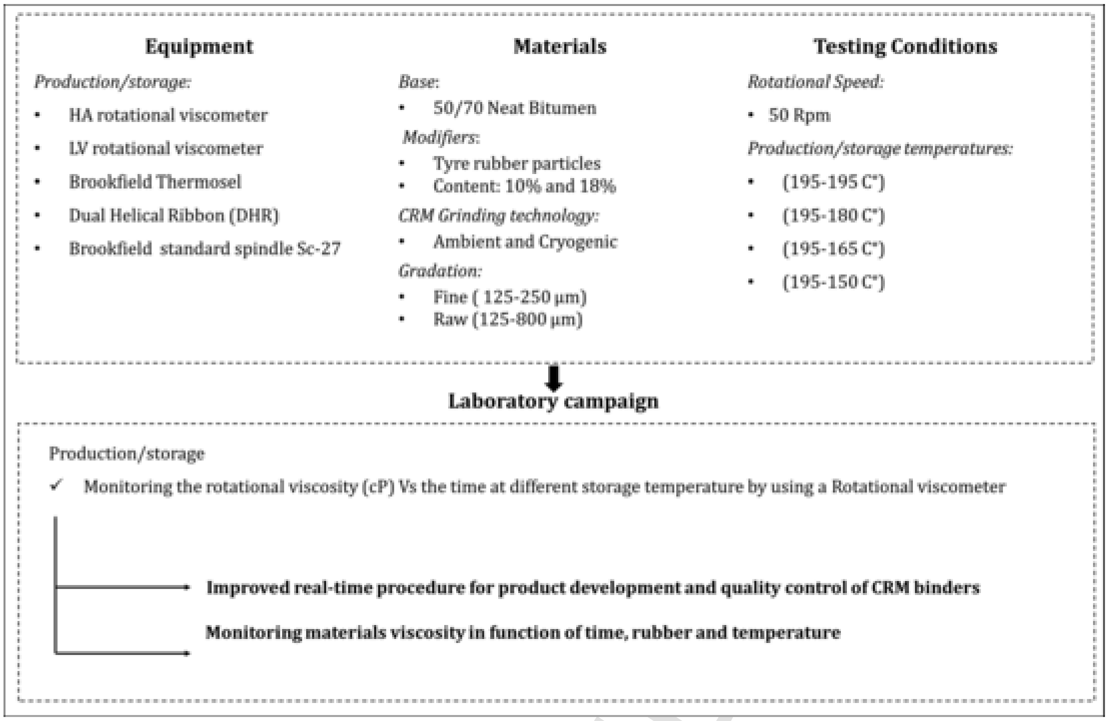


Fig. 1. Experimental programme.

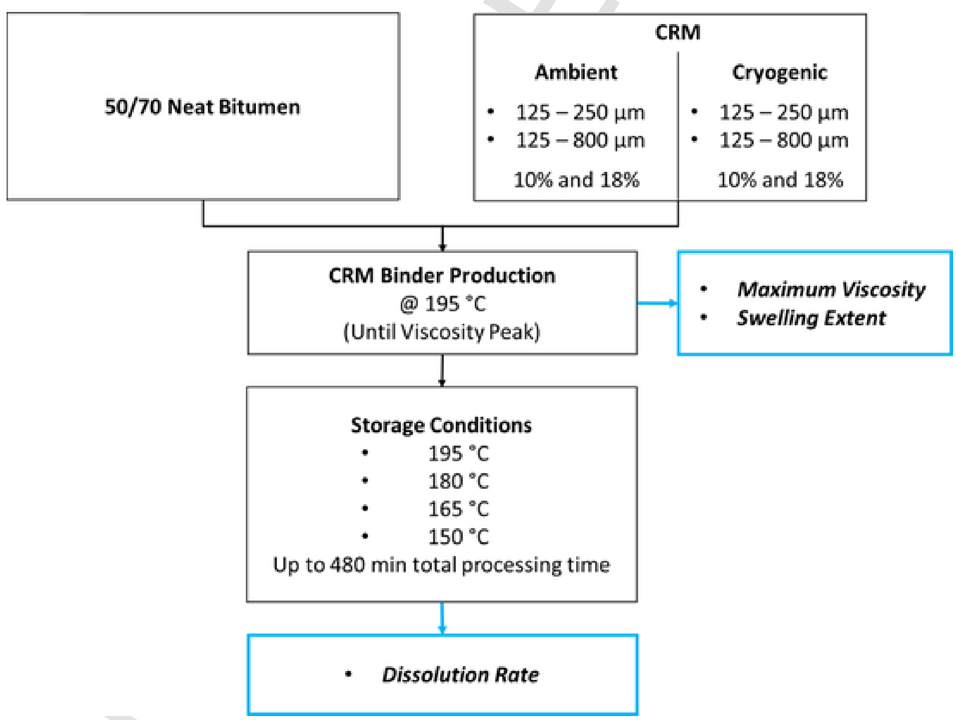


Fig. 2. Flow chart of experimental design procedures for evaluating the interaction effects on the production of CRM binders.

suggested the use of specific parameters such as the swelling extent and swelling rate to monitor the viscosity change over time.

2.1.1. The dual helical ribbon

The adaptation of a Brookfield viscometer with standard impellers is not trivial. In fact, the conventional testing geometry of rotational viscometer (coaxial cylinders with spindle geometry as impellers) seems not being well suited for complex materials such as CRM binders. Recent studies [26,27] highlighted that such setup is not designed for heterogonous bituminous binders. Indeed, it is common incurring in phenomena like phase separation, sedimentation or agglomeration, which could lead to having not representative samples. To overcome these issues, this work introduces a novel geometry, the DHR (Fig. 3), where, *c* (1.25 mm) is the clearance between blades and the container wall and *d* (35 mm), *s* (17 mm) and *w* (3 mm) are respectively the diameter, the pitch and width of the impeller. The DHR was specifically designed and calibrated to guarantee the sample stability minimising the above-

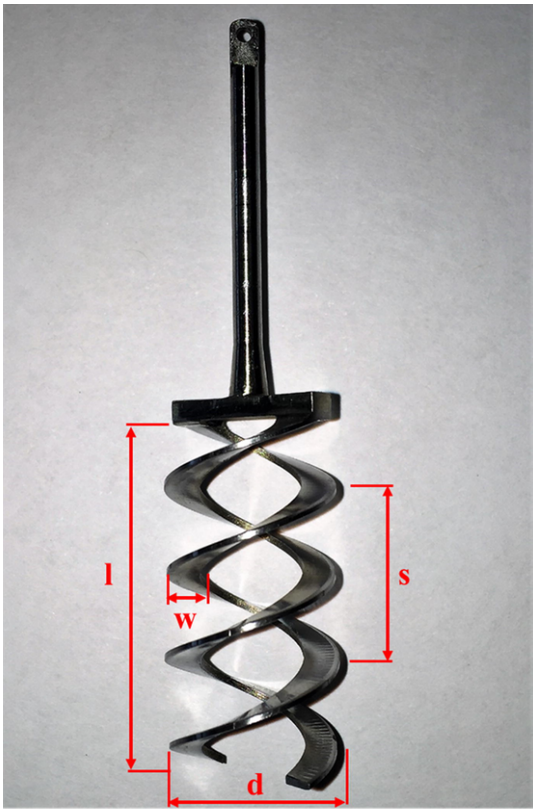


Fig. 3. The DHR impeller for use in a Brookfield Viscometer.

mentioned phenomena [27]. In fact, during viscosity measurements at high operational temperature, CRM binders seem to suffer significantly of sample instability, and this does not allow having a homogenous distribution of rubber particles within the binder, hence moving the spindle up and down during the mixing is necessary to help distribution of the rubber particles. Of course, this doesn't guarantee the homogeneity of the sample, it doesn't provide reliable measurements, and it could even lead to damaging the equipment [25,26]. The DHR, was designed to reduce the level of heterogeneity of complex binders and to guarantee the sample stability during the high temperature rotational test. This configuration allows the investigation of the variables associated with the manufacturing and storage of CRM binders (e.g. swelling extent and dissolution rate) while measuring viscosity in real time without incurring in common issues related to standard equipment such as phase separation and sedimentation. Overall, the Brookfield viscometer with DHR has several benefits including the precise control of mixing and testing temperatures, continuous monitoring of viscosity measurements, the ability to keep the rubber uniformly distributed within the blend, and all of this by using small quantities (10–15 g) of material. The DHR presented in this work is an upgrade of a dual helical impeller (DHI) designed and developed by Lo Presti [25]. The previous investigation showed the benefit of using the DHI to reduce the heterogeneity of complex systems such as the case of crumb rubber modified binders and, in turn, to provide more stable and realist viscosity measurements. Fig. 4 shows the homogeneity of the sample being proceeded with the DHR device. To take this pictures, a transparent container was used along with a transparent liquid, a standard visco-fluid of known viscosity (50 cp) + CRM. It can be seen the positive evolution of the homogeneity of the sample thanks to the mixing action of the DHR device. In the beginning, the settlement of the rubber particles is obvious (a). After the DHR starts rotating the rubber follows an ascendent flow along the axel of the container (b), and after some seconds the distribution of the rubber particles gets homogeneous (c).

2.1.2. Real-time monitoring of CRM binder's properties during manufacturing and hot storage

In this study, the real-time monitoring of the properties' development of CRM binders is extended to perform quality control also during the stirred storage phase. As widely described above, maintaining sample stability allows achieving the target viscosity for the CRM binders after both manufacturing and storage. Hence, a laboratory procedure enabling to predict the viscosity changes during storage would allow assessing the right conditions during hot storage. Here is the laboratory procedure used in this investigation:

- The low shear manufacturing process is simulated by using a rotational viscometer with a small cylindrical container testing geometry and a coaxial Dual Helical Ribbon (DHR) (Fig. 3) in place of more conventional spindles (I.e. SC-27).
- Manufacturing of materials is carried out at 50 rpm. This value is not simulative of field production, and it was chosen since it is high enough to guarantee sample stability. The value is kept constant to avoid possible complications due to the shear-thinning nature of CRM binders as well as to keep consistent the shear rate and obtain homogeneous data from the tests.
- After the viscosity peak is reached, the simulation of the hot storage is undertaken by simply changing the temperature of the thermal chamber, when needed. This is done under the assumption of storing the binder in an agitated tank with a rotational speed of 50 rpm
- The monitoring of the hot storage phase is undertaken for at least 8 h. The experiment could be prolonged to simulate longer storing time (i.e. 24 h, 48 h), but for practical reasons, 8 h can be considered long enough.

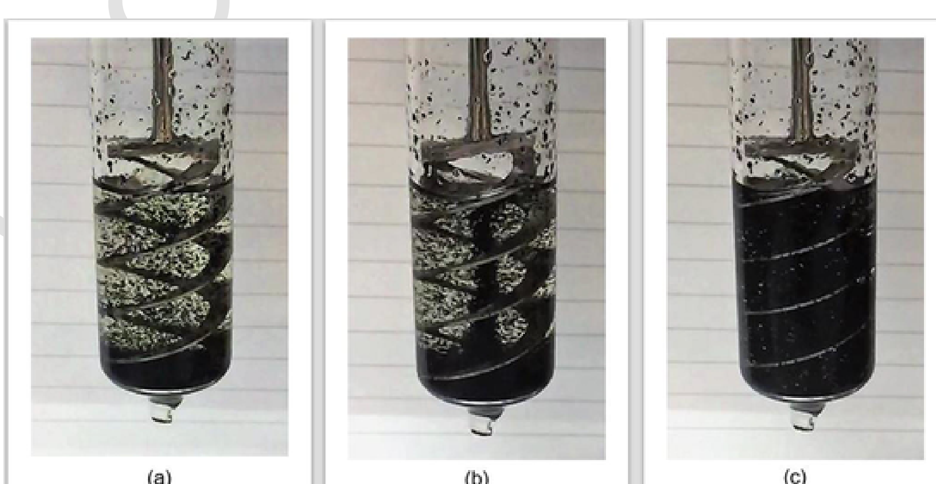


Fig. 4. Photographs of the DHR impeller showing the axial pumping force with a mixture of a visco-fluid (η = 50 cP) and rubber particles at the start (a), after few seconds (b) and at the end (c).

- The effect of different storage temperatures can be investigated by changing the temperature setting of the thermal chamber and keeping monitoring the change in viscosity.
- This procedure can be adapted to any rotational viscometer equipped with a DHR; however, asphalt technologists are recommended to compare a-priori the expected high torque for the lower range of temperatures and the viscometer's limits.
- The real-time monitoring produces five parameters that can serve asphalt technologists to optimise manufacturing and storage conditions (more details in the next section). These five parameters are:
  - o 1) the swelling extent, which is the time to reach the maximum viscosity at the production temperature;
  - o 2) the swelling rate, a ratio given by the maximum viscosity at production temperature over the time elapsed from the start of the production;
  - o 3) the maximum viscosity, which is the peak viscosity recorded at the end of the manufacturing stage and/or at the beginning of the storage period, after the storage temperature has been reached;
  - o 4) the dissolution rate, given by the ratio of the final storage viscosity over the initial storage viscosity, which provides information about the percentage of viscosity loss during the storage period;
  - o 5) final viscosity, which provides information about the viscosity value at the end of the storage period

A qualitative evaluation of the mixing capabilities of the DHR was preliminarily undertaken in this investigation and reported below. The experiment was carried out to provide an example of the results obtainable with real-time monitoring of CRM binder manufacturing and storage, as well as to highlight the importance of using a DHR for the suggested procedure.

2.1.3. Qualitative assessment of the proposed procedure

Fig. 5 shows the curve of a laboratory simulation of the manufacturing of CRM binder made of a 50/70 pen base bitumen mixed with 18% of crumbs (Raw size). For this example, the whole process was monitored for a total time of 360 mins. At first, manufacturing at 177.5 °C was performed by using both SC-27 and DHR and ended once the peak viscosity was reached. Immediately after, a simulation of hot storage at the same temperature was carried out, and viscosity values monitored up to the end of the 5 h.

The rubber is gradually added within 5 min after which, only for the SC-27, a drastic increase in viscosity is followed by a severe decrease up to more realistic viscosity measurements. This phenomenon is due to the necessity of the operator to move up and down the impeller to ensure proper distribution of rubber particles within the bituminous matrix. After this phase, the SC-27 records a build-up of viscosity values typical of a swelling process. This increment is due to the rubber particles swelling which results in a reduction in the inter-particle distance as well as stiffening of the binder by reducing the oily fractions of the bitumen (adsorbed by the rubber particles). As the swelling ends, the rubber particles start devulcanizing/depolymerising which leads to a decrease in viscosity. On the other hand, using the DHR guarantees appropriate mixing of the material, allows having a smooth curve of viscosity build-up and does not need the intervention of the operator. Also, the swelling process seems to act faster and to a higher peak value that is a symptom of a more intimate interaction between bitumen and CRM. On this regard, Table 1 provides a quantitative estimation of the parameters used to compare the simulation carried out by using both SC-27 and DHR and displayed in Fig. 5.

At last, to have a confirmation of the different interaction achieved by using the SC-27 and the DHR, at the end of the tests the samples were poured to allow a visual evaluation (Fig. 6). Results show a clear difference in terms of system heterogeneity: the material manufactured using the SC-27 presents particles agglomeration that inevitably affects the viscometer readings while the DHR provides a final blend where the rubber is clearly better integrated, showing an aspect similar to that of industrial scale CRM binders. Fig. 7 shows SEM images (x50) of the aspect of the surface for both samples. The sample processed with the DHR displays a surface smoother than that of the sample produced with SC-27, meaning that the DHR device promotes advanced the rubber-bitumen interaction.

3. Results

The idea behind the experimental programme tests was to reproduce the binder manufacturing as well as the storage periods under different temperatures, aiming at investigating the impact of different processing conditions on the viscosity evolution. As listed in Fig. 1 eight different materials were analysed as a result of blending CRM binders having different rubber content (10 and 18%) size (raw 125–800 µm and fine 125–250 µm) and grinding technology (ambient and cryogenic) for a total duration (production + storage) of 480 min. Real-time monitoring procedure was used for both manufacturing and storage stage as described in section 2.1. CRM binders were produced at 195 °C with the DHR device. Once peak viscosity was reached, the temperature of the thermal chamber was reduced and the simulation of the hot storage phase at different storage temperatures undertaken.

In order to facilitate a comparative analysis of the real-time monitoring exercise of the blend's viscosity evolution with storage time and temperature, the results are summarized in Table 2, then to better analyse the trends observed, results are plotted in Figs. 8–15 and discussed in the following paragraphs

As a first result, from the real-time monitoring of the manufacturing phase (Fig. 8), it can be highlighted that it is evident that higher CRM content requires more time to reach the maximum viscosity at production temperature

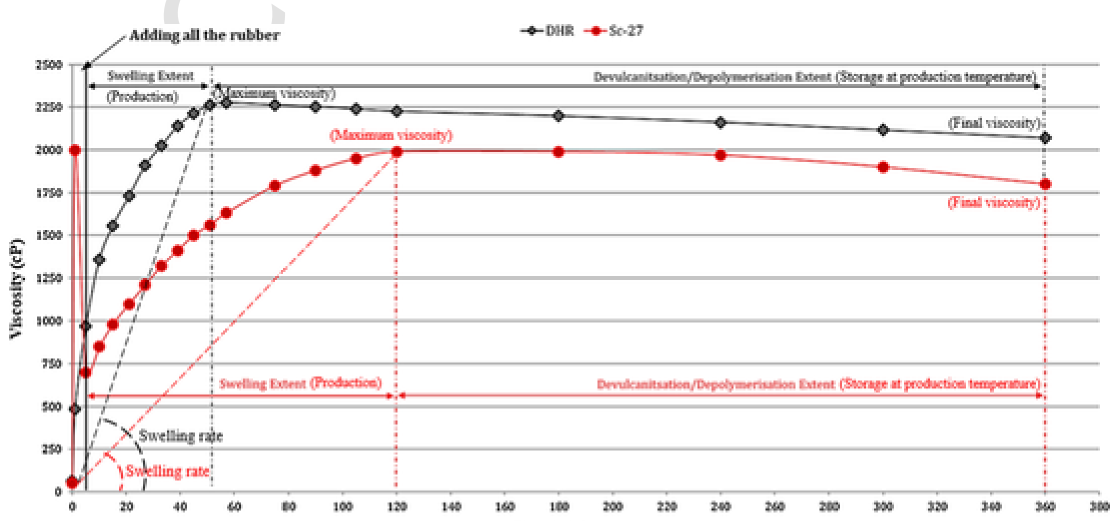


Fig. 5. Real-time monitoring of the manufacturing stage (177.5 °C, 50 rpm 18% ambient 125–800 µm CRM).

Table 1
Quantitative parameters associated with the interaction process.

	DHR	SC-27
Swelling Extent (min)	51	120
Swelling Rate (cP/min)	46.9	18.5
Maximum Viscosity (cP)	2263	1990
Dissolution rate (cP/min)	0.8	1.1
Final Viscosity (cP)	2070	1800

of 195 °C. These results agree with the general rule at the industrial scale that the minimal time to reach a proper rubber-bitumen interaction is 45 min and 60 min for low and high rubber content modified binders, respectively. The higher the content of rubber, the larger the time needed to reach the maximum viscosity.

3.1. Investigating the relation between storage temperatures and initial storage viscosity

With regards to the influence of the CRM type on the initial storage viscosity, Fig. 9 confirms that ambient rubber produces CRM binders with slightly higher viscosities. This result can be referred to the larger specific surface of the ambient rubber that allows the rubber particles absorbing more oil and swell more. Of course, CRM binders with 18% of CRM display higher viscosity values that when incorporating 10% of rubber, however all the blends show having a similar trend of initial storage viscosity by changing the storage temperature.

3.2. Investigating the relation between storage temperatures and CRM size

For this analysis, four storage temperatures have been considered (195 °C, 180 °C, 165 °C and 150 °C). To make the discussion of results easier, here only results of blends with 18% ambient CRM are reported, while a complete quantitative assessment of these simulations is reported in Table 2.

Figs. 10and 11 show the viscosity trend over the time at different storage temperatures; the first shows the details of a blend manufactured by using 18% ambient raw CRM (125–800 μm), while the latter refers to the blend with the ambient fine CRM (125–250 μm). In both graphs, the eventual bump in viscosity values at approx. Fifty minutes are due to the transition from the production temperature (195 °C) to the different storage temperatures. Results show a similar trend for both cases in terms of swelling extent and viscosity peak during the production period. Regarding the storage period, while it is clear that CRM particles are easier to be digested/dissolute at 195 °C, as was shown by the highest dissolution rates, only slight differences between raw and fine CRM binders were recorded. This partial conclusion is thoroughly analysed in section 3.4 by considering all the dissolution rates obtained in this investigation.

3.3. Investigating the relation between storage temperatures and CRM content and type

Fig. 12 provides a comparison of the viscosity evolution of the blends obtained with different rubber content, respectively 10% and 18%. As expected, using higher rubber content allows reaching a higher viscosity peak at the end of the production period (approx. 50 min). In addition to that, it can also be

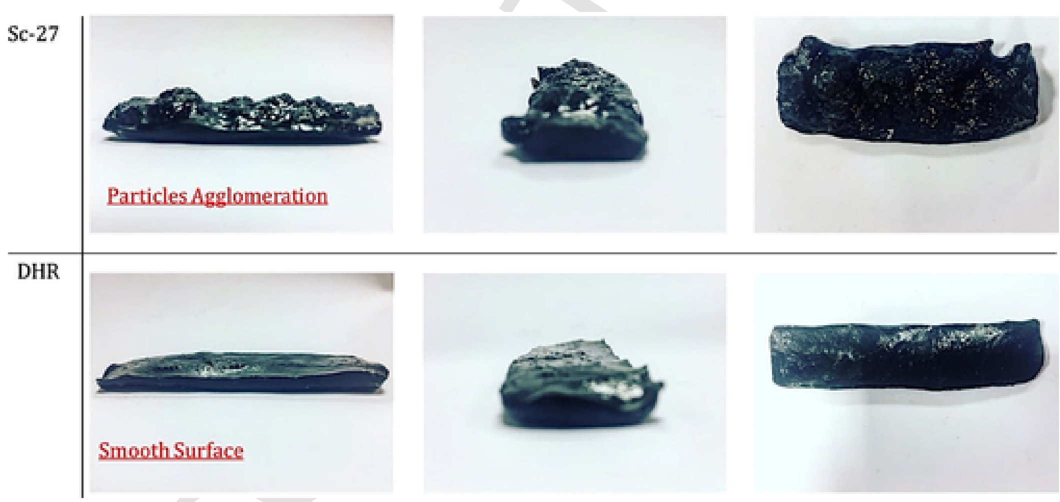


Fig. 6. Qualitative assessment of the improved bitumen-rubber interaction obtained using DHR instead of SC-27.

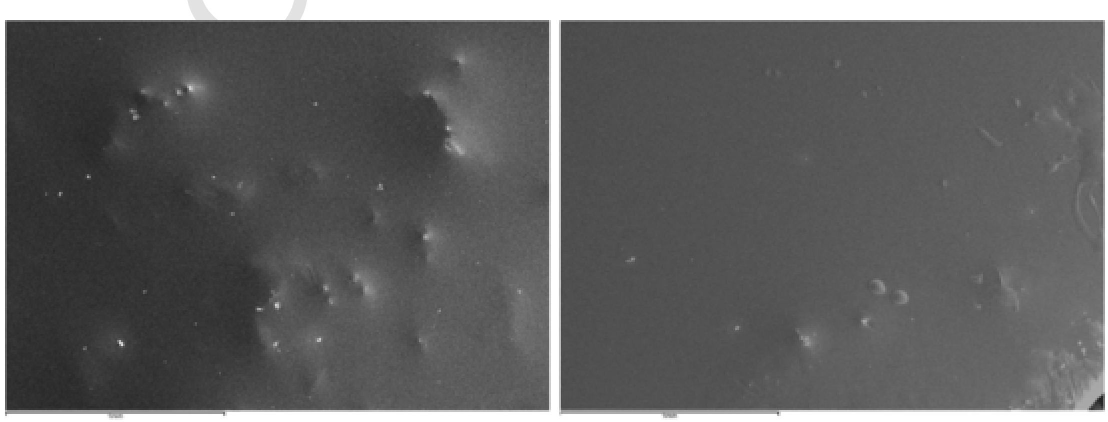


Fig. 7. SEM images (× 50) of the samples processed with SC-27 and DHR, respectively.

Table 2
Quantitative assessment of the real-time monitoring exercise.

Particle Content	(%) Grinding Tech	Particles size	Production Temp (°C)	Swelling Extent (minutes)	Storage Temp (°C)	Initial Storage Viscosity (cP)	Dissolution Rate (%)	Final Storage Viscosity (cP)
10%	Ambient	Raw	195	45	195	276	33	184
					180	418	28	276
					165	662	13	602
					150	1002	9	910
		Fine	195	39	195	211	33	137
					180	349	25	262
					165	616	10	552
					150	818	10	740
	Cryogenic	Raw	195	39	195	179	13	156
					180	326	7	312
					165	547	4	529
					150	938	2	915
		Fine	195	33	195	151	12	133
					180	234	6	220
					165	427	5	404
					150	763	3	745
18%	Ambient	Raw	195	63	195	1159	36	740
					180	1734	35	1140
					165	2258	26	1665
					150	3215	11	2944
		Fine	195	60	195	1209	49	611
					180	1656	23	1274
					165	2208	24	1633
					150	2994	10	2668
	Cryogenic	Raw	195	51	195	1030	21	851
					180	1527	10	1370
					165	2231	7	2070
					150	2792	5	2645
		Fine	195	45	195	1071	18	878
					180	1527	8	1412
					165	1932	7	1826
					150	2589	8	2484

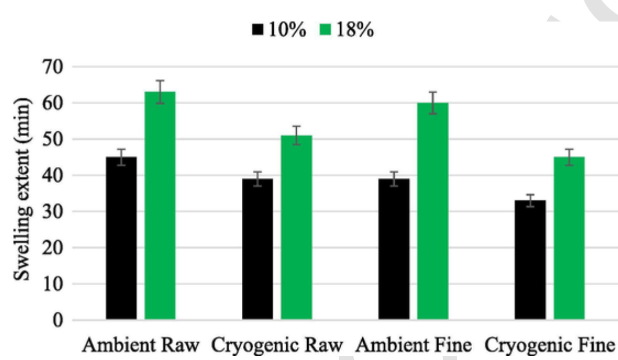


Fig. 8. Swelling extent for ambient and cryogenic CRM.

observed for both cases that the viscosity dissolution is affected by the storage temperature: while the dissolution rates at 150 °C are 9 and 11% for the modified binders with 10 and 18% CRM respectively, at 180 °C storage temperature these rates reach values of 28 and 35% respectively, around three times higher. These results mean that for both modified binders, with 10 and 18% CRM, the increase in the storage temperature brings about higher dissolution rates.

Fig. 13 shows that using cryogenic rubber the interaction mechanism between rubber and bitumen is reduced. For example, with a storage temperature of 180 °C and 18% content of raw rubber using the cryogenic technology a dissolution rate of 10% is obtained, lower than the 35% value obtained in Fig. 12 with CRM by the ambient technology. A similar trend is also recorded at 150 °C storage temperature (5% and 11% dissolution rates, for cryogenic and ambient CRM respectively), indicating once again that the cryogenic CRM reacts more slowly with the bitumen. In other words, cryogenic CRM produces modified binders with lower dissolution rates during the storage period.

For the sake of brevity, this section presents only a few detailed comparison examples to show the capabilities and the method to analyse the results from the DHR device. Beyond these examples, all the values for dissolution rates obtained in this investigation are plotted together in the following section to withdraw and support general conclusions and recommendations aimed to maintain the quality of CRM binders during the storage period.

3.4. Investigating the relation between storage temperatures and dissolution rates

Due to the high temperature during the storage period, the CRM binders loses part of their initial storage viscosity. This phenomenon has been quantified in this study as the dissolution rate, which represents the percentage of viscosity that has been lost from the beginning to the end of the storage period. By looking at data in Table 2 and trends in Figs. 14 and 15, as a general comment, it can be stated that ambient CRM always presents the highest dissolution rates. This fact could be related to the lower specific surface of the cryogenic rubber particles that slow the depolymerisation of the rubber. On the contrary, the ambient CRM, due to its higher specific surface, is more vulnerable to the effect of temperature during stirred storage, resulting in higher dissolution rates.

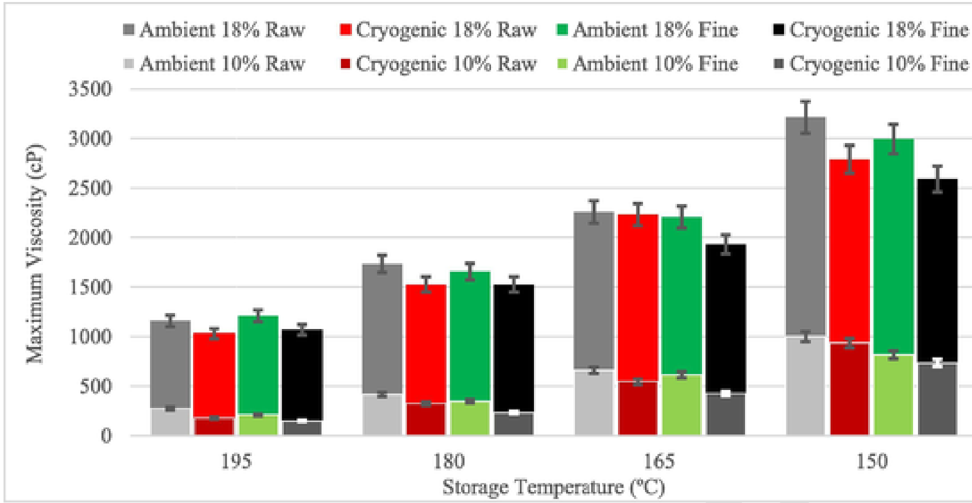
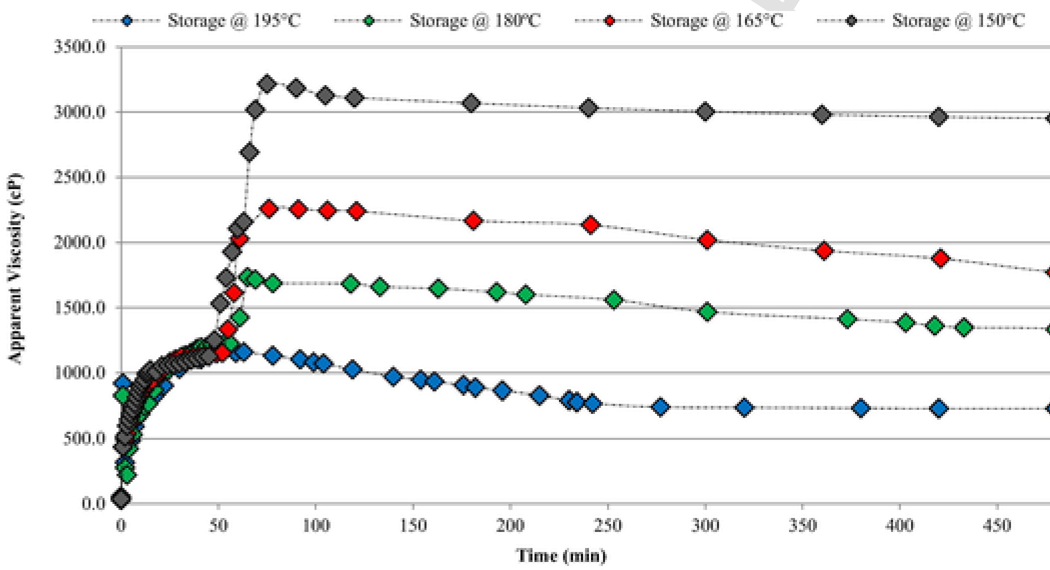


Fig. 9. Initial storage Viscosity vs storage temperature, 10% and 18% CRM.



Particle Content (%)	*Grinding Tech*	*Particles size*	*Production Temp (ºC)*	*Swelling Extent (min)*	*Storage Temp (ºC)*	*Initial Storage Viscosity (cP)*	*Dissolution Rate (%)*	*Final Storage Viscosity (cP)*
18	Ambient	Raw	195	63	195	1159	36	740
					180	1734	35	1140
					165	2258	26	1665
					150	3215	11	2944

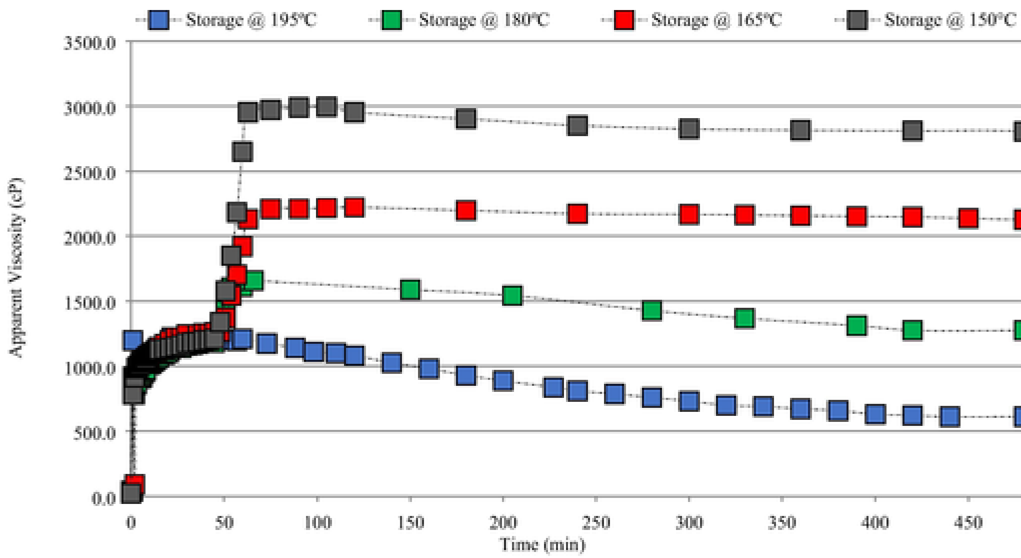
Fig. 10. Viscosity evolution at different storage temperatures. 18% ambient CRM 125–800 µm.

The size of the rubber appears not to have an essential influence on the dissolution rate. From these results, it seems that cryogenic rubber should be recommended to minimize the loss of viscosity during the storage of the CRM binder at the asphalt plant. Anyway, by reducing the storage temperature to 150 °C, the dissolution rate diminishes considerably for both ambient and cryogenic rubber.

4. Conclusions and recommendations

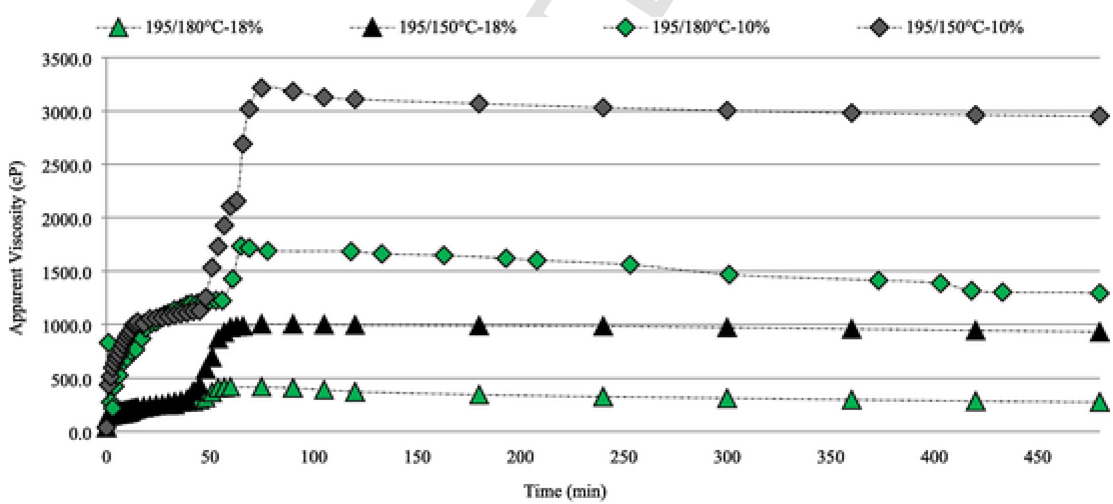
In this investigation, the conventional Brookfield spindle SC-27 has been substituted by a novel Dual Helical Ribbon (DHR) In fact, the DHR not only allows to continuously measure the viscosity of the sample but also provides improved mixing performance which guarantees the stability of the sample during measurements. Previous studies have shown the possibility of using rotational viscometers to emulate the manufacturing of CRM binders. This research extends the possibility of governing the modification process also to the hot storage phase.

Several CRM binders with different CRM type and content have been manufactured and tested at different storage temperatures to show the capabilities of the procedure. Blends were manufactured at 195 °C, and after the peak viscosity was reached, the chamber temperature was reduced at different levels to reproduce different storage temperatures. The impeller was always kept at 50 rpm to avoid settlement problems during manufacturing and keep constant the shear rate to achieve a fair comparison among all the data obtained in the testing program. Real-time monitoring of each blend can be translated into five parameters that allow performing a comparative analysis of the modification process during manufacturing and hot storage. The percentage of viscosity lost during the simulation of the storage period has been defined as the dissolution rate. High dissolution rates indicate a loss of properties of the binder during



Particle Content (%)	Grinding Tech	Particles size	Production Temp (°C)	Swelling Extent (min)	Storage Temp (°C)	Initial Storage Viscosity (cP)	Dissolution Rate (%)	Final Storage Viscosity (cP)
18	Ambient	Fine	195	60	195	1209	49	611
					180	1656	23	1274
					165	2208	24	1633
					150	2994	10	2668

Fig. 11. Viscosity evolution at different storage temperatures. 18% ambient CRM 125–250 µm.



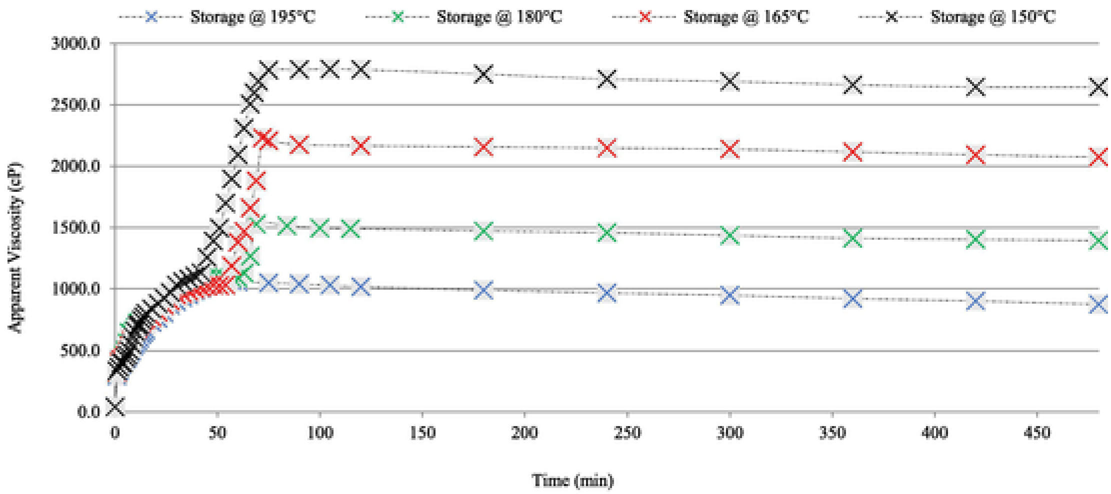
Grinding Tech	Particles size	Production Temp (°C)	Particle Content (%)	Swelling Extent (min)	Storage Temp (°C)	Initial Storage Viscosity (cP)	Dissolution Rate (%)	Final Storage Viscosity (cP)
Ambient	Raw	195	10	45	180	418	28	276
					150	1002	9	910
			18	63	180	1734	35	1140
					150	3215	11	2944

Fig. 12. Evolution of viscosity at different concentration of ambient CRM 125–800 µm.

the storage period. As a result of the work done, this study helped to produce evidence to support the following conclusions:

- It has been observed that the dissolution rate of the blends is strongly influenced by the storage temperature and the grinding technology of the rubber.
- In terms of industrial production, it seems that the use of cryogenic rubber will result in slightly lower viscosities during the production phase of the CRM binders. However, cryogenic CRM is less affected during hot storage, leading to lower dissolution rates.
- In any case, reducing the temperature of the storage tanks at the asphalt plant to 150 °C will minimise the dissolution rates, this is, will better keep the quality of the CRM binders in the tanks.

In future investigations, the relation between the shear rate of DHR at several speeds and the shear rate of full-scale mixers and stirrers should be



Particle Content (%)	*Grinding Tech*	*Particles size*	*Production Temp (°C)*	*Swelling Extent (minutes)*	*Storage Temp (°C)*	*Initial Storage Viscosity (cP)*	*Dissolution Rate (%)*	*Final Storage Viscosity (cP)*
18	Cryogenic	Raw	195	51	195	1030	21	851
					180	1527	10	1370
					165	2231	7	2070
					150	2792	5	2645

Fig. 13. Evolution of viscosity at different storage temperature using cryogenic CRM 125–800 µm.

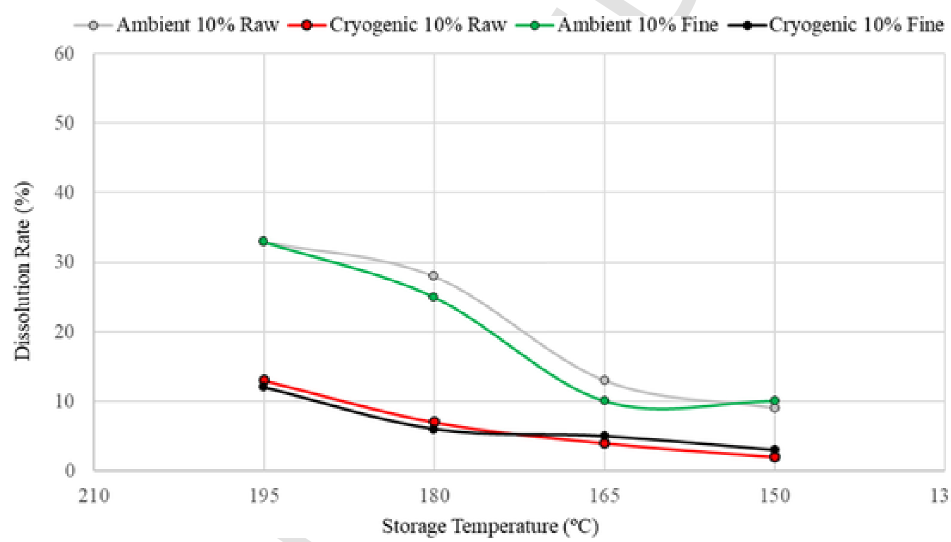


Fig. 14. Dissolution rates vs storage temperature, 10% CRM.

investigated. This knowledge could help to adapt the speed of the DHR to the specific industrial equipment to be used for the production of the CRM binder in a determined construction project and obtain more accurate predictions, adapted to each study case. Further studies should also look at evaluating the procedure with an extended experimental programme which includes longer storage time. In fact, the dissolution rate seems to be a promising parameter to engineer the optimal storage conditions of modified binders, even beyond CRM binders.

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.

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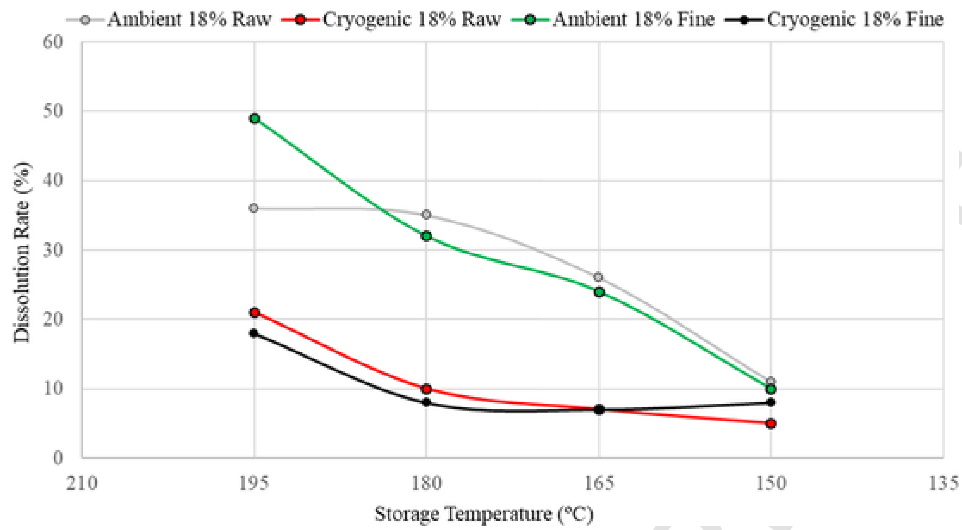

Fig. 15. Dissolution rates vs storage temperature, 18% CRM.

Authors' contribution

The authors confirm contribution to the paper as follows: Study conception and design: Juan Gallego Medina, Davide Lo Presti; Data collection: Gaspare Giancontieri, Juan Gallego Medina; Analysis and interpretation of results: Juan Gallego Medina, Gaspare Giancontieri, Davide Lo Presti; Draft manuscript preparation: Davide Lo Presti, Gaspare Giancontieri, Juan Gallego Medina,. All authors reviewed the results and approved the final version of the manuscript.

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