Microstructure analysis and mechanical performance of crumb rubber modified asphalt concrete using the dry process

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Abstract

Globally, the vast majority of waste tires are landfilled, with catastrophic ecological consequences and in particular, serious threats to human health (e.g. fire, pests and soil contamination). Because of the increasing environmental awareness, the use of crumb rubber modified asphalt has become an important recycling strategy for waste tires. The so-called crumb rubber (CR), which is the recycled rubber from tires, has become a common additive in hot mix asphalt mixture due to its improvement of the mechanical performances of asphalt mixtures. The purpose of this study is to investigate the effect of adding crumb rubber to asphalt mixtures using the dry process by relating mechanical performances with microstructural characterizations. It was possible to observe the influence of conditioning process (time during which the asphalt mixture is kept at a high temperature after mixing) on the mechanical performance of the mixtures that depend primarily on the properties of the crumb rubber used. Specifically, it is shown that the conditioning time has an influence on the Marshall test results as well as the viscosity measurements. By using the Environmental Scanning Electron Microscope (ESEM), the distribution of the crumb rubber within the mixture has been investigated. In particular, the distribution of the crumb rubber by changing the conditioning time is of interest. The results showed that crumb rubber is well dispersed in the asphalt mixture when the conditioning time is increased. The effect of crumb rubber on the microstructure using Atomic Force Microscopy-Infrared Spectroscopy (AFM-IR) indicated that the main chemical change takes place in the para domain and catana or the so-called bee structures diminish on the CR modified bitumen as a consequence of more conditioning time.

Keywords

Asphalt; Recycled Tire Rubber; Crumb Rubber; Conditioning time; ESEM; AFM-IR

1. Introduction.

About 1.4 billion tires are sold worldwide each year and subsequently turn into the category of end of life tires [1]. Improper management of used tires causes many environmental problems. Depending on the geographical location, part of this waste material is used as fuel (energy recovery), material recovery, or it is landfilled. More than 50% of the 1,000 million tires that end their service life every year are discarded without any treatment [2]. The tire is a complex product that is made of three main components: elastomeric compounds, fabric, and steel.

Crumb Rubber (CR) is the common name used to identify the tire rubber parts used to modify asphalt binder. Because of the increasing environmental awareness, the use of CR modified asphalt has become a vital recycling strategy for waste tires. CR is usually incorporated into asphalt mixtures using two different methods, which are referred to as the wet process and the dry process. In the wet process, CR acts as an asphalt binder modifier, while in the dry process, CR is used as a portion of the fine aggregates [3].

It has been shown that increasing the amount of crumb rubber content increases the viscosity of the binder [4–6]. The benefit of increased viscosity is that additional binder can be used in the asphalt mix to reduce reflective cracking, stripping, and rutting while improving the binder's response to temperature change and long-term durability [7]. The addition of CR also resulted in a modification of the rheological response of the binder. Specifically, CR addition improved the resistance to rutting and its thermal cracking resistance [8–13]. Loderer *et al.* [14] showed that the larger the specific surface of the CR particles, the better the final properties of the CR-modified binder.

Most of the rubberized asphalt research conducted worldwide use the wet process to produce a CR-modified binder. However, the use of the dry process has several advantages. The dry process allows more flexibility for the asphalt manufacturers as it allows the CR to be added in the same manner as the aggregates, and therefore, no additional equipment is needed in the plants or laboratories. Also, the use of the dry process allows incorporating more CR content in the asphalt mixture than the wet process does. However, a proper design is necessary to ensure a good performance of the asphalt mixtures, and there are still many open questions that affect the mix design when using the dry process.

When using the dry process, the resulting asphalt mixture must be at relatively high temperatures for a designated minimum period (conditioning time) to permit interaction between the crumb rubber and asphalt. This conditioning time has a significant influence on the binder properties [15] and on the short-term performance of the modified asphalt mixtures [16,17]. The CR particles significantly change during this process by swelling up to five times its size [9,18]. Once CR comes in contact with the binder, the low molecular weight portion of the binder is absorbed by the CR and causes it to swell, and consequently, if the high-temperature exposure is continued, degradation occurs [19]. For this reason, some authors suggest increasing the amount of binder to compensate for this swelling effect [9,16].

The properties of the CR are a critical factor that influences the interaction between CR and binder. The rubber particle shape seems to affect a blending degree with the binder during the conditioning time. The rubber particle size is also an essential factor for the rheological and engineering properties of mixtures [20,21]. This effect was carefully analysed in another study [22], where two different CR-modified mixtures were studied by using Environmental Scanning Electron Microscopy (ESEM). It was shown how particles from different sources exhibit different roughness. In this study, it is attributed changes in binder viscosity to different particle morphology and surface area due to the unequal grinding and tearing properties of various tire components.

Generally, the effect of type of rubber and the influence of the conditioning time on the microstructure and physical properties of asphalt mixtures modified using the dry process has not been thoroughly investigated. Thus, it is vital to conduct studies to obtain a better understanding of the interaction of all components. The objective of this study is to evaluate the influence of conditioning time on the performance of asphalt containing two different types of CR in various asphalt mixtures fabricated using the dry process in the laboratory. This was accomplished through the evaluation of the effect of conditioning time (usually also known as digestion) on the viscosity evolution of binders in contact with different concentrations of CR as well as on the mechanical performance of different asphalt mixtures modified by the dry process. In addition, imaging techniques such ESEM and Atomic Force Microscopy Infrared-spectroscopy (AFM-IR) were used to evaluate changes at the microstructure level of asphalt mixtures and binder, respectively.

2. Experimental design.

2.1. Materials.

Three semi-dense asphalt mixtures with a maximum aggregate size of 4 mm and 12% air void content were prepared (SDA 4-12). Two of them were experimental CR-modified mixtures using the dry method, and the other was a control mixture. All of them had the same aggregates composition. The grading curve is plotted in Figure 1. The binder used in the CR-modified mixtures was a conventional 50/70 penetration grade binder. For the control mixture, a polymer-modified binder (PMB 45/80-65) was used as required by the Swiss standards (SNR 640 436: 2015). In this way, it is possible to investigate if CR modification can have a similar performance in comparison to polymer modified mixtures.

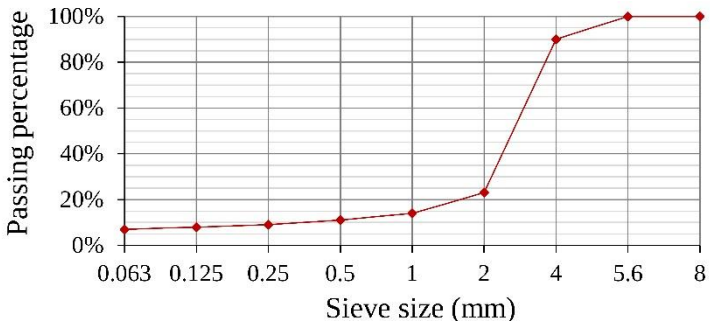

Figure 1. Particle size distribution

The only difference between experimental mixtures was the type of CR used. Both CR types were produced using the ambient grinding method. However, one was modified by a polymer coating treatment (CR-A), whereas the other was produced without any additional modification (CR-B). Both CR types had a maximum particle size of 800 µm. It can be seen from ESEM images in Figure 2 that the shape of both CR particles are rough, and no obvious differences in particle textures and shape can be found.

a) b)

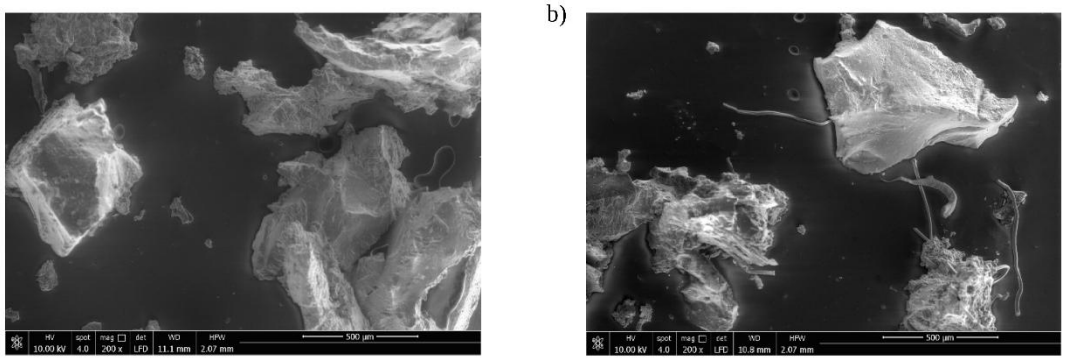

Figure 2. ESEM micrographs of (a) CR-A and (b) CR-B at 200 × magnification (scale bar = 500 μm)

2.2. Asphalt binder characterization.

In parallel to the evaluation of the asphalt mixes, the effect of the conditioning time on the binder-CR interaction was also evaluated by analysing the viscosity, chemical composition, and morphology of the asphalt binder samples with CR. The aim of this analysis was to compare the two rubber types and evaluate the changes they could cause during the mixing and conditioning processed in the properties of the binder. This study was carried out to understand the behaviour of the modified mixtures better, but in no case was this binder used to prepare asphalt mixes. Asphalt mixes have always been prepared using the procedure described in the previous section (dry method).

The binder samples were prepared by blending the reference binder (50/70 penetration grade) with the CR particles at 165 ºC (mixed by hand for 30 seconds). These samples were kept in the oven at 165 ºC for different conditioning times (30, 60, 120, 180, 240, and 300 minutes). This conditioning time plays an important role in allowing the interaction between CR particles and bitumen. To simulate the conditioning time applied to the mixtures, no mixing energy has been applied to the bitumen samples. Only every 30 minutes and at the end of each conditioning time, the samples were mixed (30 seconds by hand) in order to minimize the settlement of the CR to the bottom of the container. Dosages of 10 and 15% of CR by mass of binder were used to prepare each binder sample incorporating CR. These samples were stored at room temperature for their characterization.

2.2.1. Determination of dynamic viscosity of bituminous binder.

The rotational viscosity test (EN 13302:2010) was used to characterize the binder samples. This test was performed following the internal Standard Operation Procedure (SOP) established. Once the sample was poured into a pre-heated sample container, the spindle was lowered into the binder and the system was allowed to equilibrate at the test temperature (30 minutes). During this time, the spindle was kept rotating at a very low shear rate (5 rpm). During the test, the viscosity was measured using a rotational speed of 20 rpm. Once the viscosity was stabilized, this value was recorded as a unique value. This viscosity value allows to compare between the two types of particles used and to evaluate the influence of the conditioning time on the viscosity of the binder.

2.2.2. Chemical and micro-morphology surface characterization of asphalt binder samples (AFM-IR).

AFM-IR is a device that combines atomic force microscopy (AFM) with infra-red (IR) spectroscopy. Recent studies on asphalt binder in nanometre resolution using AFM-IR [23,24] revealed complex surface chemistry of bitumen surface. They showed that the chemistry of surface microstructures depends highly on the aging level and thermal history of the bitumen. In this study, we used this novel technique to provide information on the effect of conditioning time on the complex surface chemical makeup of bitumen samples with CR.

Bitumen samples with 15% weight of CR-B after 30 and 180 minutes of conditioning time were evaluated using an AFM-IR microscope. To reach a required smooth surface for AFM-IR specimens, the heat-casting method due to its less effect on the fundamental properties of the bitumen and to avoid using any solvent has been selected. From a few millimetres of the bitumen surface (1.5 ± 0.5 mg) of bitumen, samples were extracted using a laboratory spatula (at room temperature) to avoid surface layer oxidation. The spreading of bitumen on the thin glass coverslips created approximately 500 μm thickness bitumen film and covered about an area of ca. 4 mm^2 after annealing samples on a hot plate (at 110 ˚C, 5 minutes) and quenching to

close the room temperature in a refrigerator (3 ± 2 ˚C, 5 minutes). The samples in a Petri dish and were always covered during the sample preparation procedure (Figure 3) to avoid dust accumulation.

The AFM-IR scans were performed using a nanoIR2 device from Anasys Inc./Brucker equipped with a pulsed tuned IR laser gun from the Daylight Solutions model: MIRcat-2400. AFM-IR collects chemical information of the surface in nanometre resolution, principally by simultaneously collecting FT-IR data with conventional AFM scans. The scans were conducted in an enclosed environmental chamber at the low humidity condition (RH<3%) and temperature constant of 23 ºC.

AFM-IR scans were performed in tapping mode, using the first mechanical resonance of the AFM cantilever to collect AFM topography and AFM phase, and the second mechanical resonance for the chemical mapping. The first and second mechanical resonance frequencies of the gold-coated probes were 53.8±0.1 kHz and 346.80±0.5 kHz, respectively, with the spring constant of 40-45% of 1-7 N/m and the set point 5.1-5.4 V. The scan rate was set to 0.7 Hz and for the scan sizes of a 15 µm by 15 µm square area with a resolution of 500 lines in X and 250 lines in the Y direction of all scans.

The emitted IR laser source covers a range from wave number 892 cm^{-1} to 2000 cm^{-1} (wavelength: 5-11.5 µm and maximum power 50 mW). This available IR range is sufficient to characterize the main characteristic absorption peaks of asphalt binder such as 1456 cm^{-1} and 1376 cm^{-1}, covering critical functional groups of carboxyl and sulfoxide groups.

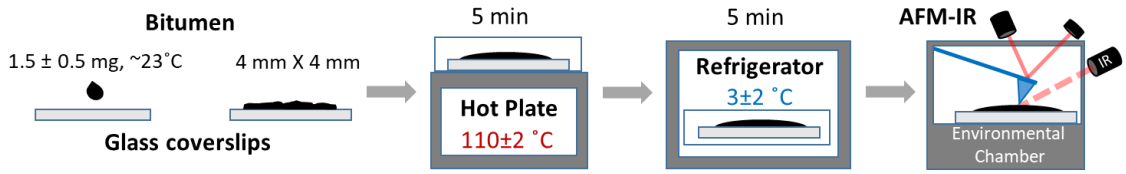

Figure 3. AFM-IR specimen preparation steps

2.3. Asphalt mixtures evaluation.

The dry method was used to prepare batches of 25 kg of the SDA 4-12 mixture. The CR content was fixed at 1% by mass of aggregates (equivalent to ca. 15% by mass of asphalt binder). The binder content was fixed at 6.2% by mass of mixture for all the mixtures. As for the manufacture protocol, first, the preheated aggregates (185 ºC) were mixed with the CR for 1.5 minutes. Then, the preheated binder (160 ºC) was added and mixed with the aggregates and CR for another 2 minutes.

Once mixed, the mixture was conditioned in the oven at 165 ºC (conditioning temperature) for 60, 90, and 120 minutes (conditioning time). These times were selected taking into account the minimum time required for a proper interaction between the crumb rubber and the binder, as well as the maximum time linked to practical and economic issues during the manufacture and transport of the asphalt mixture. After each conditioning time, the performance of the asphalt mixes has been evaluated by means of the following tests and analyses.

2.3.1. Volumetric properties.

Maximum density, bulk density, and air void content have been calculated. For determining the maximum density, the volumetric procedure of EN 12697-5 (procedure A) was used. For the bulk density determination, due to the percentage of voids that are expected to be higher than 10%, the specimen bulk density was determined following the procedure D of EN 12697-5. During this procedure, the calculation of the specimen bulk density is conducted geometrically. The air void content was calculated according to the specification in the standard EN 12697-8.

2.3.2. Marshall test.

This test was performed using cylindrical samples (102 mm diameter) compacted at 155 ºC using the Marshall hammer with 50 blows per side. Marshall tests were conducted following the standard EN 12697-35. This European Standard specifies a test method for determining the stability, flow, and the Marshall Quotient values of specimens of bituminous mixtures prepared using the impact compactor method of test EN 12697-30. In this case, the specimens were conditioned in the water at 60 °C for 45 minutes before the test.

2.3.3. Water sensitivity test.

Water sensitivity test. The influence of moisture on the asphalt mixture performance was evaluated through the water sensitivity test (EN 12697-12). Cylindrical samples (102 mm diameter) of each type of mixture with 60 and 120 minutes of conditioning time were prepared. These samples were compacted at 155 ºC using the Marshall hammer, with 35 blows per side were used and conditioned following the specification in the standard. In this case, the samples were tested under indirect tensile stress at 22 ºC.

2.3.4. Imaging analysis of asphalt mixtures (ESEM).

Mixtures prepared with CR-B and conditioning times of 60 and 120 minutes were evaluated using the ESEM. For this purpose, specimens with dimensions 28×47×10 mm^3 were cut from the centre of Marshall specimens (50 blows per side). Figure 4 shows the specimen preparation steps. The samples were impregnated with resin and polished following the method described in a previous study [25]. The ESEM experiments were performed with an FEI Quanta 650 by Thermo Fisher. This study was conducted in a low vacuum mode to avoid any specimen perturbations [26]. Specifically, lower inert gas purge (IGP) equal to 1.9×10^{-7} mbar and upper IGP equal to 1.2×10^{-9} mbar and current emission of 309 mA.

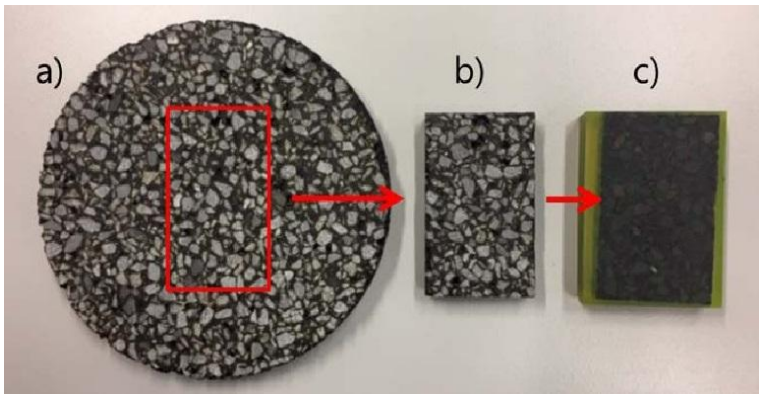

Figure 4. ESEM specimen preparation steps. (a) Marshall sample cylinders, 102 mm diameter and 64 mm high; (b) Centre cut 28x47x10 mm^3; (c) Impregnated and polished ESM sample.

3. Results and discussion.

3.1. Asphalt binder characterization.

3.1.1. Determination of dynamic viscosity of bituminous binder.

The rotational viscosity test was used to evaluate the grade of modification of the bitumen properties (specifically the dynamic viscosity) caused by the incorporation of CR after different conditioning times. The results for different CR concentrations are shown in Figure 5 and Figure 6.

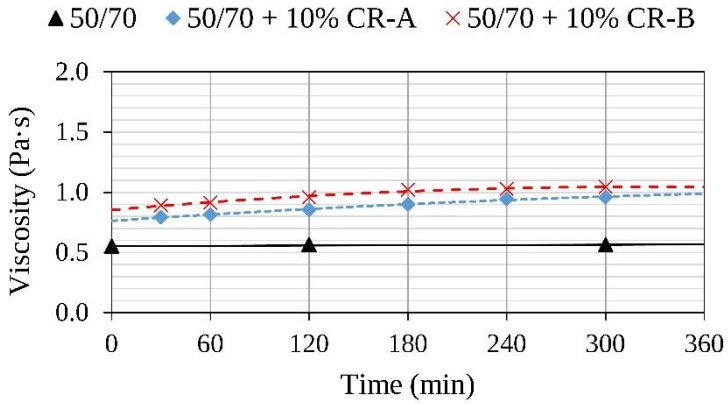

Figure 5. Rotational viscosity test results from 10% CR content samples

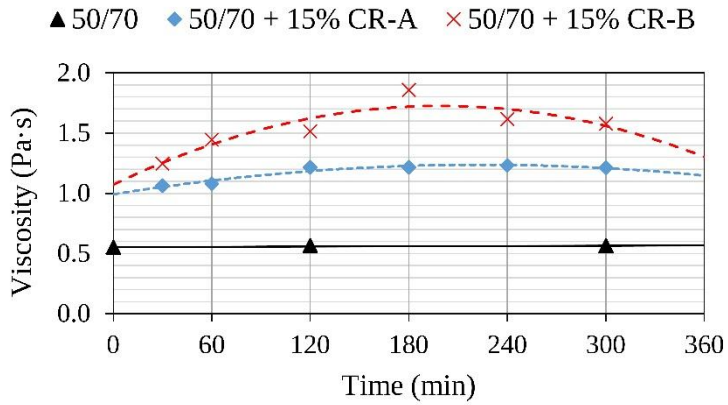

Figure 6. Rotational viscosity test results from 15% CR content samples

The interaction with CR clearly causes a variation in the bitumen viscosity, probably indicating that the CR digestion does occur at conditioning temperature (165 ºC). An increase in the viscosity of the binder was more significant when CR-B particles were used. These results indicated a better interaction between CR-B particles and the binder compared to the CR-A particles. The use of 15% CR by mass of binder resulted in a higher viscosity than the use of 10% for both CR types and in all the conditioning times tested. There were no changes in the bitumen viscosity for the 50/70 binder (reference samples) after exposure to temperature for the conditioning times used. Therefore, thermal aging cannot be significantly influencing the results of this test, and any changes can be attributed to the CR bitumen interaction. For all the samples, the viscosity tends to increase with conditioning time and then decreases, indicating the effect of an initial swelling phase followed by the degradation process discussed above. To evaluate this behaviour, the results were fit to a 2nd order polynomial trend line, as shown in equation 1. Using the trend line, the maximum viscosity value was calculated for each sample and shown in Table 1.

$$\mu = C_1 \times t^2 + C_2 \times t + C_3 \quad (1)$$

$\mu \equiv Dynamic\ viscosity\ (Pa \cdot s)$

$t \equiv Digestion\ time\ (min)$

$C_1, C_2, C_3 \equiv Polynomial\ constants$

Table 1. Regression analysis of rotational viscosity test results.

Sample	R^2	C_1	C_2	C_3	μ_{max}	Time μ_{max}
10% CR-B	0.98	-1.82E-06	1.18E-03	0.853	1.05 Pa·s	326 min
10% CR-A	0.99	-8.15E-07	9.17E-04	0.763	1.02 Pa·s	562 min
15% CR-B	0.80	-1.65E-05	6.56E-03	1.071	1.73 Pa·s	199 min
15% CR-A	0.93	-4.89E-06	2.19E-03	0.992	1.24 Pa·s	224 min

Both samples with 15% CR content reached the maximum viscosity in the range of conditioning time tested in the laboratory (199 and 224 minutes for CR-B and CR-A respectively). Sample modified with CR-B, apart from the higher increment in the viscosity previously described, required less conditioning time to reach the maximum in the viscosity value than the sample with CR-A. Specifically, the sample with CR-B increased its viscosity 0.65 Pa·s until it reached a maximum value of 1.73 Pa·s in 199 minutes, while the sample with CR-A increased 0.25 Pa·s until it reached a maximum value of 1.24 Pa·s in 224 min. In addition, the reduction in the viscosity values after reaching the maximum was more rapid in the samples with CR-B.

As the addition of crumb rubber has a direct effect on viscosity [4–6], it is assumed that samples with 10% CR content reached the maximum viscosity outside of the range of conditioning time tested in this study. It is assumed that the behaviour of these samples follows that reported in the literature [14,27] and is similar to the samples with 15% CR content and can also be adjusted by an order 2 polynomial trend line. By making this assumption, it is possible to hypothesize that the samples with CR-B required less conditioning time to reach the maximum viscosity value than samples with CR-A (326 minutes instead of 562 min). However, in this case, both samples reached similar viscosity values (ca. 1.0 Pa·s).

These results indicate that there are differences in the CR-bitumen interaction as a function of the conditioning time, depending on the type and content of CR. Samples with 15% of CR-B showed a higher variation in the bitumen viscosity than samples with 15% of CR-A. The results are an indication that CR-B interacts faster with the bitumen; that is, the process of swelling and degradation occurs faster. As in the published works of Dantas Neto *et al.* [27] and, more recently, Loderer *et al.* [14], the results of viscosity tests suggest that CR-B particles have a higher specific surface than CR-A particles and therefore, a higher capacity to interact with the binder. Despite both CR types were produced using the ambient grinding and no obvious differences in particle texture and shape (Figure 2), possible differences in the grain size distribution and the influence due to the polymer coated treatment applied to CR-A could be the reasons that justify these differences.

Also, not surprisingly, the amount of crumb rubber has a significant influence on the results. Changes in viscosity were higher and occurred more rapidly for samples with 15% than for samples with 10% of CR-B. These results indicate that CR content also has a significant influence. These results corroborate with Lo Presti and Airey [28]. In this study, the authors evaluate the influence of CR content on the bitumen properties and conclude that 15% rubber content seems to be the minimum to get a tire rubber modified binder with significant improvements.

3.1.2. Chemical and micro-morphology surface characterization of asphalt binder samples (AFM-IR).

The AFM-IR technique was used to show the effect of conditioning time on the local surface chemistry of the bitumen CR blends, and the results of two example cases of the modified bitumen are shown in Figure 7. The figure shows bitumen 50/70 with 15% by weight of CR-B after 30 minutes of conditioning (Figure 7

(a-d)) and after 180 minutes of the conditioning time (Figure 7 (e-h)). Typically three primary microstructures develop on the bitumen surface at about room temperature after annealing of bitumen from its melting temperature. Wrinkled areas have been named catana domain (bee structures), islands around the wrinkled domains are called peri domain, and para is the smoother domain in the neighbouring of the peri domains.

The obtained results show a change in the respective microstructures as well as the local chemistry of these microstructures. The topography and phase images in Figure 7 (a, e) and Figure 7 (b, f) respectively show a difference in the catana domain or the so-called bee structures sizes. Catana domains are smaller and harder to distinguish on the 180 minutes conditioned samples (Figure 7 (e)) compared to the case of after 30 minutes (Figure 7 (a)) conditioning time. IR spectra collected from a point in the peri and a point in the para domain marked on the AFM phase scans (Figure 7 (b, f)) are plotted in Figure 7 (i). IR maps show the distribution of a favourable functional group exited by a specific wavenumber.

IR mapping is conducted in parallel to the AFM scans: Figure 7 (c, g) is that of sulfoxide groups at a wavenumber of 1030 cm^{-1} and Figure 7 (d, h) IR mapping of carboxyl functional groups at a wavenumber of 1700 cm^{-1}, each corresponding to the AFM scans of the same location on the two samples. The local IR spectra are shown in Figure 7 (i) indicates a difference in the local chemistry of the peri and para domains. According to noisy IR spectra as a result of the nature of the AFM-IR in tapping mode, the AFM-IR spectra were smoothened using the Savitzy-Golay function by fitting a polynomial order of five on six neighbouring points. It can be seen that after the 180 minutes conditioning, certain functional groups diminish in both domains. For example, the red peak of the peri domain at 1456 cm^{-1} is reduced in the yellow spectra (180 minutes). The same occurs on the para domain, comparing the blue spectra with the green. However, IR map panels of Figure 7 (d, h) show the relatively non-uniform distribution of carbonyl functional groups for the case of 30 minutes of conditioning time compared to the 180 minutes.

At the same time, detected peaks of functional groups remained the same for different conditioning times on the peri domain, but the main chemical change took place in the para domain. This change in para domain chemistry potentially leads to changes in the interaction with the surrounding peri domain and the bulk material. The reduction of the bee structures attributed to the wrinkling of the surface is an indication of this change in mechanical properties, which contributes to changes in bulk mechanical properties of the CR modified bitumen in different conditioning times.

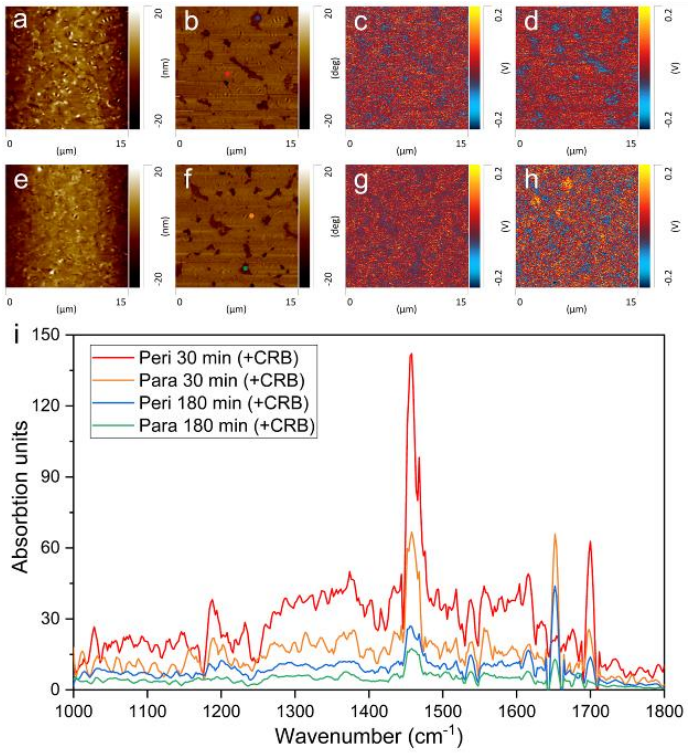

Figure 7. AFM-IR results for 30 minutes (a-d) and 180 minutes (e-h) conditioning time. (a, e) Topography; (b, f) Phase; (c, g) IR maps of sulfoxide groups at the wavenumber of 1030 cm^{-1}; (d, h) IR maps of carboxyl functional groups 1700 cm^{-1}; (i) IR spectra

3.2. Asphalt mixtures evaluation.

3.2.1. Volumetric properties.

As described previously, the effect of conditioning time on the mechanical performance of the CR asphalt mixtures has been evaluated through laboratory tests. Volumetric properties are shown in Figure 8. Regarding air voids content, all the mixtures meet the requirement established by the Swiss standards for this kind of mixtures (10-14%). The control mixture showed higher air voids content than the experimental mixtures. This is expected since the mix design was not adjusted to account for the addition of the CR and the consequent swelling process. In this case, the composition of the aggregates and the binder content were the same for all the mixtures. It is assumed that the added CR particles were partially filling the air voids of the modified mixtures, resulting in asphalt mixtures with lower air void content.

It has been observed that there are differences in terms of volumetric properties between mixtures with different types of CR. Mixtures with CR-B showed higher air voids content than mixtures with CR-A. It's important to notice that the type of CR was the only difference between these two mixtures. These differences could be related to the treatment of the two types of CR. The polymer coating of CR-A could affect the bitumen light fraction absorption. As discussed in the previous section, the interaction of the CR-B with the binder led to a higher viscosity compared to that of the CR-A. This viscosity difference could be the reason for the higher air voids content in the mixture with CR-B. However, differences in the particle size distribution of the CR or in the final effective binder content could be another reason behind the obtained results. Nevertheless, there are no significant variations regarding the conditioning time applied.

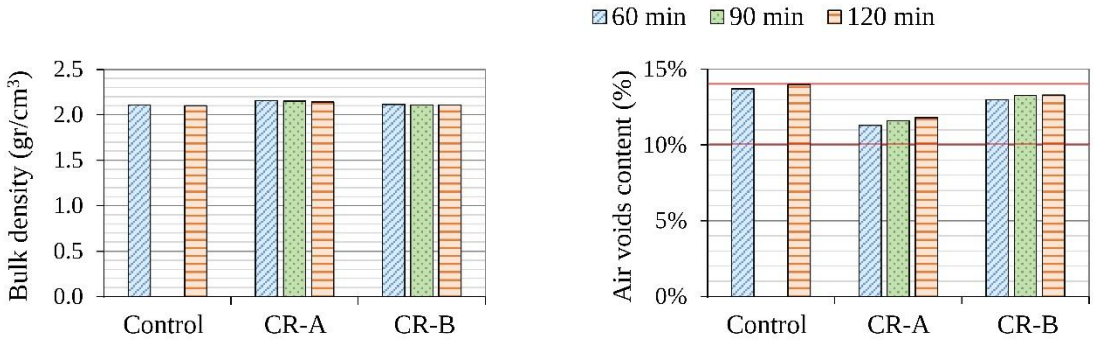

Figure 8. Volumetric properties of SDA mixtures

3.2.2. Marshall test.

Stability and flow, including the confidence interval (α=0.05), and Marshall Quotient (MQ) values are shown in Figure 9. For SDA mixtures, the Marshall test is not a requirement in Swiss standards. However, this test is a good indicator of the expected mechanical performance. MQ, the ratio of stability to flow, is recognized as an indicator of the resistance of the material to shear stress and is also related to the mixture's resistance to permanent deformation. Thus, high MQ values indicate that the asphalt mixture will present high stiffness and greater ability to transmit the loads and to resist creep deformation [29].

The results in Figure 9 show that the conditioning time has a significant influence on the Marshall test results, and this also depends on the type of CR used. Regarding Marshall Stability, all CR modified mixtures showed an increase in the stability as the conditioning time increases. Both CR modified mixtures showed higher stability increment in comparison to the control mixture, which was more influenced by the effect of exposure to higher temperatures, after 120 minutes of conditioning time. The increase in stability value can be attributed to the stiffening of the binder as a result of exposure to temperature and by the stiffening as a result of CR modification. Furthermore, the increase of stability could be attributed to the effect of CR and its role as an elastic aggregate. By comparing types of CR, this increase is higher in the mixture with CR-B than in the mixture with CR-A.

In terms of Marshall flow, it was observed that the results depend on the CR used as well. At longer conditioning times, mixtures with CR-B resulted in lower flow values. However, there were no significant changes in mixtures with CR-A. For instance, samples with 120 minutes of conditioning time showed a slight increase in this value. Converted to MQ values, mixtures with CR-A showed constant values for all of the conditioning times tested (around 4.5 kN/mm). For mixtures with CR-B, the MQ increases as conditioning time increases, from 3.6 kN/mm with 60 minutes of conditioning time to 4.9 kN/mm with 120 min. As described before, the increase in the MQ showed by the mixtures with CR-B could indicate an increase in the mixture stiffness and an improvement in the rutting performance. As seen in the viscosity test results, the Marshall results indicate that mixtures with CR-B are more affected by the conditioning time than mixtures modified CR-A with a surface polymer coating treatment.

Compared with control mixtures, the CR-modified mixtures showed higher MQ, especially samples with 120 minutes of conditioning time. This is mainly caused by the lower flow values shown by the experimental mixtures. Therefore, it is expected that these mixtures to be stiffer than a conventional polymer modified asphalt mixture.

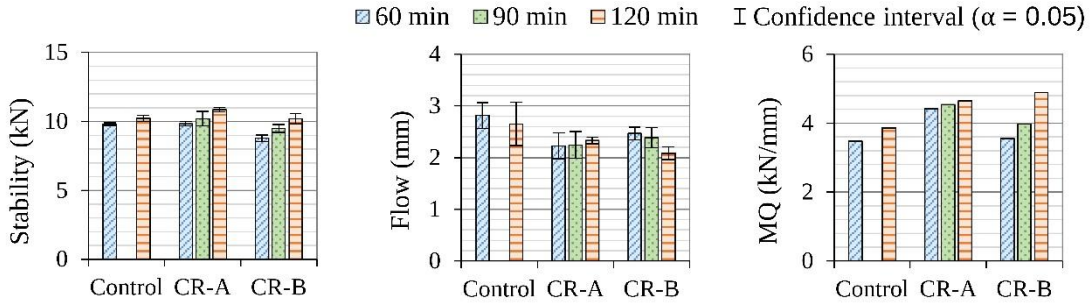

Figure 9. Marshall test results

3.2.3. Water sensitivity test.

The water sensitivity test results are shown in Figure 10. As can be seen in this figure, conditioning time did not cause a significant influence in terms of water sensitivity. Samples with higher conditioning time showed slightly higher values of the indirect tensile strength (ITS) in both dry and wet groups of samples and both types of CR. Conditioning time did not cause significant changes in the indirect tensile strength ratio (ITSR). The strength (ITS) obtained in dry and wet state of CR-B modified mixtures surpassed that of the reference. However, the experimental mixtures resulted in ITSR values similar to those of the control mixture. Although close, it seems that these experimental mixtures would not meet the requirement defined in the Swiss standards for this type of SDA mixtures (ITSR > 70%), and further work would be required to optimize the modified mixtures. The authors would recommend the use of additives to improve the adhesion between binder and aggregates or a modification in the mixture recipe in order to improve its performance. Nevertheless, it is essential to note the differences between the binders used for the different mixtures. While the experimental mixtures used a conventional binder, the control mixture used a polymer modified binder.

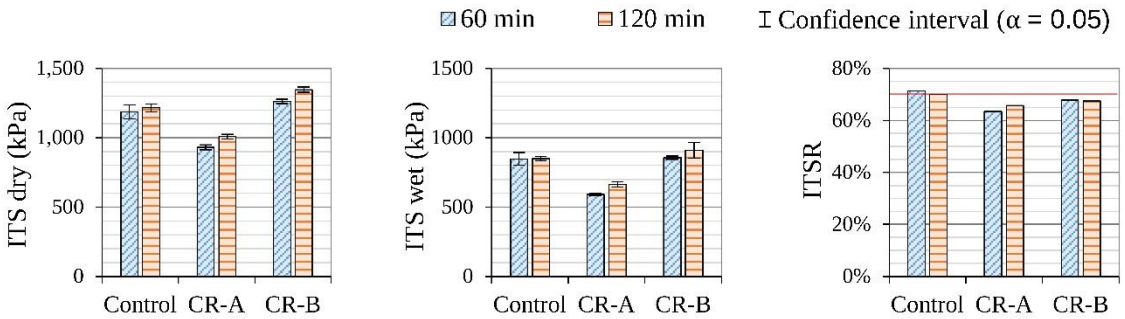

Figure 10. Water sensitivity test results

3.2.4. Imaging analysis of asphalt mixtures (ESEM).

Figure 11 and Figure 12 show two examples of CR-B modified mixtures at 60 minutes of conditioning time and 120 minutes of conditioning time, respectively. It can be observed that after 60 minutes, the rubber particles can still be distinguished, whereas after 120 minutes, they could no longer be distinguished from the mastic, indicating that they had changed their morphology and were no longer intact. This could be due to a partial digestion or a better grade of blending consequence of the longer interaction process at high temperature. However, some fibres from the CR could still be seen.

These results corroborate the findings of Thives *et al.* [30] that investigated the effect of conditioning times on binders modified with CR. They showed that at shorter conditioning times, many particles of rubber were present in the asphalt rubber binder; however, these particles disappeared after the higher

conditioning times. Specifically, they showed how two types of crumb rubber show different contact areas when added to the neat binder. Thus, it is not surprising that the interaction between the asphalt and different kind of crumb rubber will cause different degrees of blending among the binder mixture. For instance, in the specific case of that study, the use of crumb rubber to modify the asphalt binder results in an interaction with a more substantial rubber surface, and thus, more rubber will be used in the modification of the asphalt binder. In contrast, for the crumb rubber produced by the cryogenic method resulting in a different surface structure, they found the opposite effect with less crumb rubber in contact with the bitumen. This fact finally led to a worse modification of the asphalt binder in comparison with that obtained with the ambient crumb rubber.

Within the framework of the present research, it is foreseen how ESEM can be considered a useful tool when studying the blending between rubber particles and asphalt binders. Specifically, it is shown how higher conditioning time caused a better degree of blending between crumb rubber particles and the asphalt binder.

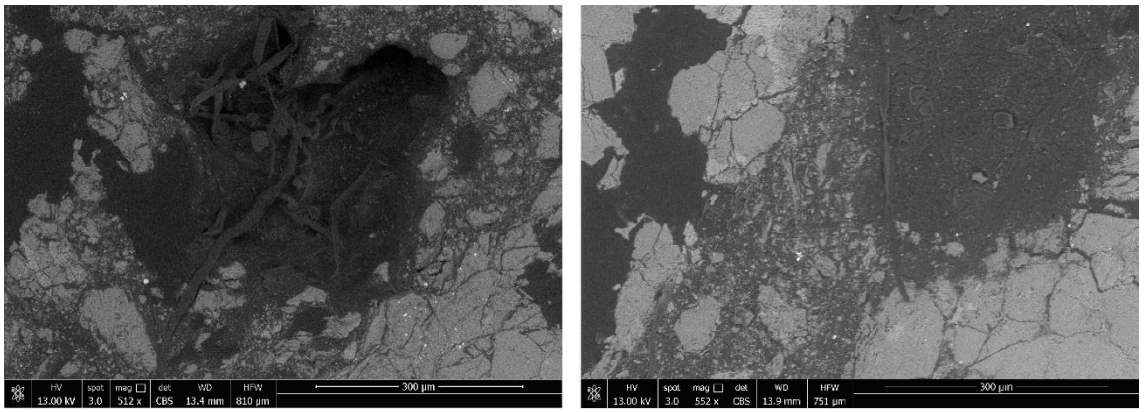

Figure 11. ESEM micrographs of asphalt mixtures samples with 60 minutes of conditioning time (scale bar = 300 μm)

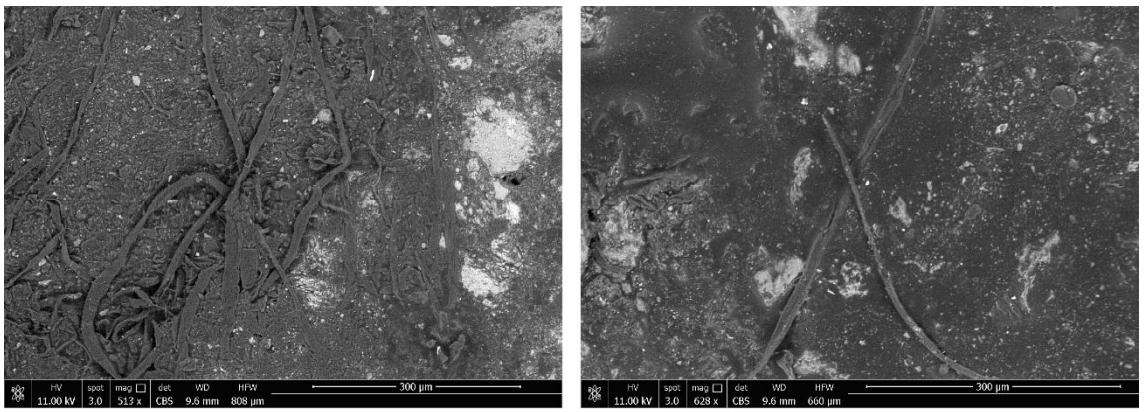

Figure 12. ESEM micrographs of asphalt mixtures samples with 120 minutes of conditioning time (scale bar = 300 μm)

4. Conclusions.

In this study, the influence of conditioning time on the modification of asphalt mixtures with CR by the dry process was evaluated. Two different types of CR were incorporated into SDA mixtures, and the results were compared to the standard SDA mixture with polymer modified binder. Mechanical tests and microstructural analyses were performed in the laboratory. The following conclusions can be drawn:

- The conditioning time has a significant influence on the asphalt mixture performance, depending on the type of CR. In this case, the use of CR modified with a polymer coating resulted in a similar mechanical performance as the reference mixture, which was independent of the conditioning time. Nevertheless, conditioning time caused significant differences in mixtures with straight run none modified CR.

- The interaction between CR and bitumen caused a modification in the bitumen viscosity. The CR-modified bitumen had a higher viscosity than the virgin bitumen. Moreover, higher CR content resulted in higher viscosity values for both types of CR.
- The bitumen viscosity tends to increase with conditioning time until it reached a maximum and then decreased. This behaviour depends on the type and content of CR. In this case, the interaction between bitumen and non-modified CR-B is faster and reach higher maximum viscosity values than the interaction with polymer modified CR-A. Also, changes in viscosity are higher and occur more rapidly for samples with 15% than for samples with 10% of CR.
- The influence of conditioning time on mechanical performance depended on the properties of CR, and, in some cases (e.g., Marshall test), this influence could have a significant effect in the mixture performance. In this case, the conditioning time had influenced the mixtures modified with CR-B more, in agreement with the viscosity test. Therefore, for a proper mixtures design, it is important to determine the optimal conditioning time by considering the particular conditions for each case (e.g., materials, type of mixture, transport time to workplace, etc.).
- From the ESEM analysis, it was observed that microstructure changes are closely linked to the conditioning time. In this sense, rubber particles can be clearly distinguished after 60 minutes but, after 120 minutes, they could no longer be distinguished from the mastic, indicating that they had changed their morphology.
- The AFM-IR results for two conditioning times of 30 minutes and 180 minutes show that after the longer conditioning time, there was less developed microstructure (bee). The main chemical change of the functional groups took place in the para domain.

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