Experimental investigation of blending phenomena in intergranular zones of recycled asphalt mixtures using customized infrared imaging and stage extraction methods

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Abstract

In the recycling road industry, a fundamental knowledge of blending mechanisms between the aged and virgin binders is necessary. An infrared imaging methodology is customized to assess the blending in different recycled asphalt mixtures. Obtained results are compared to those of another stage extraction technique. Both methods show that RA materials behave differently according to the process. Furthermore, the infrared methodology is highly-sensitive to understand blending scenarios: as remobilization is lower in warm-mix processes, the intergranular zone appears less oxidized and more homogeneous. This study encourages mix designers to optimize current blending laws with the introduction of a remobilization degree.

Graphical abstract

Highlights

- A statistical infrared *chemomap* method has been customized on recycled asphalt mixtures
- The solvent-free *chemomap* method is highly sensitive to refine mixing and blending phenomena
- The manufacturing process determines the way reclaimed asphalt affects the binder blending
- Remobilization, homogeneity and oxidation are complementary to assess the binder blending

Keywords

Durable asphalt mixtures, recycling, reclaimed asphalt, degree of blending, remobilization, bitumen, blend, homogeneity, non-destructive, solvent-free method, infrared imaging microscopy

1. Introduction

Nowadays, the main challenge in the road industry is to develop performant and durable road pavement materials. That performance is conditioned by the composite structure of the asphalt mixture where the viscoelastic bituminous binder brings the cohesion to the mineral skeleton. Traditionally, the manufacture of asphalt mixtures occurs at 160°C in a hot-mix process according to the NF EN 12 697-35 standard [1]. But it requires an important consumption in energy [2] and in virgin non-renewable resources including aggregates coming from quarries and bitumen from the crude oil distillation. To optimize energy and raw materials resources, the road industry has also developed innovative manufacturing processes that consume less energy and novel formulation technologies to reduce the supply in the virgin hydrocarbon binder. For instance, the Warm-Mix Asphalt (WMA) technology corresponds to an asphalt mixture manufacture between 110 °C and 140 °C [3] and allows burner fuels savings between 20 and 35 % [4]. Reductions of fumes emissions are also observed in several studies [5]–[7]. On the other hand, significant research in novel formulations technologies has been undertaken to consume less virgin bitumen in materials by studying properties of innovative bitumen/bio-binders mixtures [8]–[11] or by developing alternative binders from biomass to progressively replace the hydrocarbon binder [12]–[14]. Among these ecological solutions, one sustainable technology also lies on the recycling of asphalt mixtures by incorporating in virgin materials stocks of reclaimed asphalt (RA) coming from the destruction of aged pavement layers [15]. Recycling compared to alternative biomass technologies leads to several environmental and economic benefits including the reuse of the aged bitumen from RA materials [16], [17], a lower consumption in natural virgin materials [2], [7] and a reduction in the costs of landfilling road deconstruction waste. Significant reductions in greenhouse gas emissions [18] and energy consumption [19] are observed and the RA aged binder properties are also taken in consideration for the formulation of the novel asphalt mixture. Some studies have namely shown that the RA incorporation in asphalt mixture improves the rutting resistance [20]–[22].

However, asphalt mixtures incorporating more than 40 % of RA materials are hardly developed because several technical questions are raised about the mix design [23] of asphalt mixtures incorporating RA.

The first question of the mix design refers to the virgin binder nature and amount to add to the aged binder to obtain a fixed total binder amount. To answer this question, current methods are based on empirical blending laws (EN 13108-1, Annex A) [24] for which the experimental procedure consists in completely dissolving the RA binder with a chlorinated solvent, the percentage and the physicochemical properties of the aged binder are then determined. The virgin binder nature and amount are then chosen from the penetration values of the aged and total final binders. However, this very commonly used binder extraction procedure assumes that the total hard RA aged binder amount has been completely fluidized and has interacted with the virgin binder [25]–[27]. This is not the case because some studies have shown that partial interactions occurred between binders. In particular manufacturing conditions, the RA material may behave as a “black rock” [26], [28] because no interactions happen between the fluid virgin binder and the hard RA binder. Based on the blending-law dissolution principle, the virgin amount to add is not correct and that may have serious consequences on the total binder content which could be lower than expected leading to a stiffer mix in the asphalt mixture and to advanced under-asphalting damages such as fatigue and thermal cracking [29], [30].

On other hand, a second issue about the mix design is related to the lack of knowledge about blending mechanisms. Particularly, most studies make confusion between the RA aged binder availability in the mix and its blending ability with the virgin binder. In the literature, the “black rock effect” namely refers to a no

blending situation whereas the total interaction correspond to total blending [25], [26]. Some studies also consider that the degree of blending represents the amount of the total RA binder that is mobilized in an asphalt mix [29]–[31]. Using this definition, the degree of blending has also been thoroughly investigated in several studies using various techniques such as infrared spectroscopy [32], [33], microscopy ultraviolet light [34], electron microscopy [35], [36], nano-indentation [37], tomography [35], [37], fluorescence microscopy [31] or the progressive stage extraction method [38]–[41]. By this approach, it is also presumed that the available RA binder amount completely homogenizes with the virgin binder to generate a homogenous mixture [42].

However, the homogeneity degree of the binder mixture is uncertain and may be low due to different chemical compositions of the aged and virgin binders [43]. That constitutes a matter of debate [32]. Thus, other authors suggest another research concept which consists of distinguishing the RA aged binder availability in the mix from the blending efficiency. According to [44], even if the RA binder is mobilized and workable in the mixing drum, it could not reveal that the RA binder has interacted with the virgin binder to form a new colloidal structure. Questions about the degree of mixing, whether mixing or blending [45] are also legitimate and Zaumanis suggests that a good blending would be ensured by making RA binder activated and available for blending with other materials [46]. Therefore the binder blending can be viewed as a homogenization [47] of the mixture produced from the known amounts of the virgin and aged remobilized binders. On Figure 1, Nahar precisely defines the blending as the creation of a new material after mixing two different components [27].

Figure 1 : Scenarii for mixing (A) or blending (B) of microstructural properties of binder mixes [27]

Using the Atomic Force Microscopy, the blend also displays a specific microstructure where no traces of the original material's microstructures are present anymore [27]. This technique has also been used by Zhao [44] to assess the mode of interaction (*i.e* the extent of mixing or blending) between the virgin and the RAS (Reclaimed Asphalt Shingle) binders. Nevertheless, although these promising findings, few studies are still conducted in an effort to address the blending issue and homogenization in the binder mixture. That may be due to the commonly used solvent-based road procedures whereby the solvent selection may affect properties of recovered binders [44], [48]–[50].

Therefore, the objective of this paper is also to better understand blending phenomena in recycled asphalt mixtures using a novel solvent-free infrared imaging method. Recently, an innovative infrared *chemomap* methodology has been developed to assess homogeneity in hydrocarbon mixtures using carbonyl spatial distributions treated with a statistical procedure [51]. This methodology will be customized to determine the blending in recycled asphalt mixtures. In this paper, the degree of blending also refers to the degree of homogenization related to uniformity of micro oxidative properties.

As obtained results will enlarge the discussion about binder interactions, the degree of remobilization (or that of mixing) is also a term which is introduced here. As mixing is a process operation of two components in known amounts [35], [52], the degree of mixing corresponds here to the RA aged binder amount activated and available in the mix because only the manufacturing conditions and the RA binder properties allow the determination of the real activated amount of the aged binder [53].

To validate the novel method used, obtained results of this infrared imaging method will be compared with those of another stage extraction method presented previously [39]. Therefore, the results correlation between the two methods will allow validating the developed *chemomap* method on compacted asphalt mixtures and expand the understanding of blending phenomena in tested hot and warm asphalt mixtures incorporating RA.

2. Materials and methods

2.1. Materials

Some asphalt mixtures incorporating RA materials were prepared using different temperature conditions and different reclaimed asphalt percentages (Table 1): Two first asphalt mixtures, called H40 and W70, were respectively manufactured according to hot and warm mix processes (Table 1). Temperatures of materials were chosen in accordance to the NF EN 12697-35 standard [1] to be in similar temperature conditions than in a power plant. For the H40 mix, a certain amount of virgin aggregates was previously heated at 210°C and added to RA materials. The high temperature of virgin aggregates allows heating reclaimed asphalt at a higher intermediate temperature. About the W70 mix, the heating temperatures of components were also defined according to the NF EN 12697-35 standard [1]. The heating temperatures of virgin aggregates and reclaimed asphalt were fixed to 140°C (Table 1). A warm additive (EVOTHERM® VM30, Ingevity) has been incorporated into the W70 mix. It is composed of fatty amines derivatives which allow improving the wetting between aggregates and the binder. Its percentage corresponds to 0.4 % of the virgin binder.

Two other asphalt mixtures, called W100 and H100, were then produced with 50% of each binder. As the warm additive does not play a significant role in blending phenomena, no warm additive was incorporated to the W100 mix in order to allow a better understanding of blending results between these model mixes. The heating temperatures were also chosen to be comparable between H100 and W100 mixes. The liquid virgin binder was introduced at 160°C for both mixes and the RA heating temperature is respectively fixed at 160°C (for W100) and 120°C (for H100). No virgin aggregates were introduced in these specimens.

Table 1: Manufacture temperatures of tested recycled asphalt mixtures (HMA): Hot Mix Asphalt; (WMA) Warm Mix Asphalt; (H40) Hot 40% RA; (W70) Warm 70% RA, (W100) Warm 100% RA; (H100) Hot 100% RA

Mixture	Process	Specimen	Reclaimed Asphalt (%)	Virgin Aggregates (°C)	Reclaimed Asphalt (°C)	Virgin binder (°C)	Mixing Temperature (°C)
HMA	hot	H40	40.0	210	110	165	170
WMA	warm	W70	70.2	140	140	150	140
WMA	Warm	W100	96.1	-	120	160	120
HMA	hot	H100	96.1	-	160	160	160

Details are then provided about binder used in tested asphalt mixtures incorporating reclaimed asphalt. Thus, the hot H40 mix and the warm W70 mix were precisely manufactured to have a total binder amount of 5.30 ± 0.1 %. They were respectively produced by mixing a 35/50 binder and a 50/70 binder with reclaimed asphalts (Table 2). Properties of binders were characterized before mixing: the carbonyl index of both virgin binders was measured and equals to around 0.0, virgin binders are also non-oxidized. The RA aged binder oxidation was characterized after a binder solvent-extraction. For the analyzed RA stock, the carbonyl index is 8.4, what induces an advanced ageing oxidation.

In comparison to the H40 mix, the W70 higher RA percentage leads to introduce less virgin binder amount (2.47 %) to the aged binder amount (2.90 %) which is already present in the formulation. For this W70 mix, the ratio of binders equals to 1.17 (Table 2). Therefore, two W100 and H100 mixes display the same binder ratio as the W70 mix (ratio: 1.1 ± 0.1 %). The virgin binder used remains identical to the W70 mix and is little oxidized (Ico:0.1) (Table 2).

Table 2: Properties and amounts of virgin and aged binders composing recycled asphalt mixture specimens: (HMA): Hot Mix Asphalt; (WMA) Warm Mix Asphalt; (H40) Hot 40% RA; (W70) Warm 70% RA, (W100) Warm 100% RA; (H100) Hot 100% RA, Pen Index: Penetration Index, Ico: Carbonyl Index

Mixture	Specimen	Virgin binder (%)	Aged binder (%)	Total binder (%)	Ratio of binders (%)	Virgin binder		Aged binder	
						Pen index (1/10 mm)	Ico (-)	Pen Index (1/10 mm)	Ico (-)
HMA	H40	3.60	1.65	5.25	0.46	40	0.0	9	8.4
WMA	W70	2.47	2.90	5.37	1.17	57	0.1	9	8.4
WMA	W100	3.85	3.98	7.83	1.03	57	0.1	9	8.4
HMA	H100	3.85	3.98	7.83	1.03	57	0.1	9	8.4

For each mix, 20 kg of material were prepared in laboratory with a BBmax MLPC® mixer according to NF EN 12697-35 standard [1]. Reclaimed asphalt was used in a 6/10 granular fraction. The virgin binders were previously heated at a temperature comprised between 150 to 165°C (Table 1). For H40 and W70 formulations, virgin granular parts were previously mixed with reclaimed asphalt during 30 seconds $\pm$ 5 seconds. Then, the binder was added and mixed with all other components during 180 seconds $\pm$ 5 seconds.

After the formulations were prepared, 3 kg of each non compacted asphalt mixture specimen were tested by the solvent-based stage progressive method [39]. 12 kg were then used to prepare a compacted slab with $180 \times 50 \times 500$ mm³ dimensions. A BBPAC roller compactor was used in accordance with the NF EN 12367-333 +A1 standard [54]. For the infrared imaging microscopy analysis, slab cuts up to the core were performed under cold water to obtain an asphalt mixture specimen with $20 \times 5 \times 20$ mm³ dimensions. More specially, refined cuts were done on the extremities of the slab height (50 mm³) to obtain a sample thickness of 5 mm. The infrared analysis was performed on the asphalt mixture surface not directly exposed to air.

2.2. Infrared imaging microscopy methodology

An infrared microscope (Spotlight 400, Perkin Elmer) was used with an ATR (Attenuated Total Reflectance) accessory for which the crystal diameter equals to 600 μ m. A complete apparatus description is available in [53], [55]. Briefly, the intimate contact is performed between the compacted asphalt mixture specimen surface and the ATR crystal tip. The FPA (Focal Plane Array) detector allows the simultaneous acquisition of

several thousand infrared spectra on the 2D analysis zone, acquisition parameters are those described in the methodology development on the binder mixtures [51].

For each asphalt mixture formulation, the chemical mapping (Figure 2) was acquired on a refined zone ($200 \times 100 \mu\text{m}^2$) of the intergranular bituminous mastic. It allows spatially identifying the organic (peak around 2800 cm^{-1}) and mineral calcareous (peak around 1400 cm^{-1}) components of the asphalt mixture. The bitumen spatial distribution (Figure 2A) has been firstly obtained by integrating the ethylene function peak absorbance between 2995 and 2760 cm^{-1} (valley to valley). The spatial distribution of the aged oxidized binder is determined by integrating the carbonyl function area between 1720 and 1665 cm^{-1} (valley to valley) (Figure 2B). On Figure 2A, spectra 2 and 3 confirm namely differences in the bitumen concentration (Peak area = $0.042 > 0.003$) whereas spectra 4 and 5 confirm the differences in the carbonyl function concentration (2B) (Peak area = $12.7 \times 10^{-4} > 5.4 \times 10^{-4}$).

Figure 2 : Spatial distributions of the ethylene (A) and the carbonyl (B) functions for the infrared analysis of a H40 compacted asphalt mixture surface [56]

A specific methodology has been developed to assess the bitumen oxidation on the 2D infrared map. As the asphalt mixture is composed of hydrocarbon and mineral parts, oxidation variations were analyzed for the same bitumen concentration. For example, the bitumen map (Figure 2A) was firstly divided into three zones of $20 \times 20 \mu\text{m}^2$ (black dotted lines) displaying a similar ethylene function absorbance (equal to 0.040 ± 0.002). Then these reduced zones were similarly focused on the carbonyl spatial distribution (Figure 2B). It is worth noticing here that this ethylene function absorbance (0.040) is identical for all defined 20×20

μm^2 reduced zones. As tested asphalt mixture specimens display different total binder contents (5.3 or 7.8 % wt [Table 2](#)), this induces no significant influence of the total binder content on the acquisition of the chemical mapping related to the asphalt mixture sample.

In order to compare with reliability color carbonyl spatial distributions, each $20 \times 20 \mu\text{m}^2$ carbonyl chemical mapping was converted into a micro carbonyl absorbance values matrix where each value corresponds to the area absorbance of the carbonyl peak integrated on a spectrum defined on a surface of $1.56 \times 1.56 \mu\text{m}^2$ (pixel size) [51]. A specific statistical treatment was undertaken to quantify these numerical infrared data. As explained in [51], micro carbonyl absorbance values were classified into frequencies intervals defined with a 0.0001 length (in absorbance unit) and obtained appearance frequencies were used to plot statistical curves according to defined absorbance intervals.

For each chemical mapping ($200 \times 200 \mu\text{m}^2$), three statistical curves were obtained. Particular attention was paid to the Gaussian shape indicating a homogeneous distribution of carbonyl functions in asphalt mixture. For instance, [Figure 3](#) shows the curve shape for a homogeneous ([Figure 3A](#)) and heterogeneous ([Figure 3B](#)) asphalt mixture incorporating RA.

Figure 3: Distribution curves for a homogeneous (A, C) and a heterogeneous (B, D) asphalt mixture incorporating reclaimed asphalt

[Figure 3C](#) and [3D](#) illustrate the asphalt mixture incorporating reclaimed asphalt schemes. [Figure 3A](#) shows three Gaussian curves corresponding to three uniform zones (red squares on [3C](#)) in oxidation, thus the asphalt mixture is considered homogeneous. [Figure 3B](#) shows different and enlarged curves on a carbonyl micro absorbance range between 0 and 20×10^{-4} . This statistical graphical is characterized by several oxidation levels which are observed on the three analysis zones (red squares on [3D](#)), thus making the recycled asphalt mixture heterogeneous.

214 Finally, as introduced in [51], CPA (Curves Peaks Average) and CPSD (Curves Peaks Standard Deviation)
 215 blending indicators were determined to translate the positions and looks of statistical curves:

- 216 - The CPA corresponds to the average of all carbonyl abscissa values associated to curves. It provides the
 217 oxidation state of peaks.
- 218 - The CPSD refers to the standard deviation of all carbonyl abscissa values associated to curve peaks. It
 219 translates homogeneity in the mixture from the distance between curves.

220 In a similar way as [51], an asphalt mixture was defined as homogeneous if the CPSD value is less or equals
 221 to 2×10^{-4} , defined as the tolerance.

222 For each asphalt mixture specimen, chemical mappings ($100 \times 200 \mu\text{m}^2$) were also recorded at two
 223 locations which were randomly chosen on each asphalt mixture incorporating RA. Three $20 \times 20 \mu\text{m}^2$ zones
 224 were defined for each carbonyl spatial distribution. The CPA and CPSD indicators were also calculated from
 225 six distribution curves obtained for each asphalt mixture formulation.

226 **2.3. Solvent-based progressive extraction method**

227 The degree of blending of studied samples was also analyzed in parallel in an external laboratory using a
 228 second method [39]. The principle of this method is different and consists of dissolving progressively with a
 229 solvent the total binder film, from outer layers to inner layers. At the analysis end, obtained leachates of
 230 “bitumen+ solvent” were adjusted to have an identical bitumen concentration and leachates were analyzed
 231 by FTIR (Fourier Transform Infra Red spectroscopy) to determine carbonyl indexes. The carbonyl index is
 232 standardized by the introduction of a ICOfref parameter (Figure 4A), corresponding to the ICO/ICOfref ratio
 233 [39]. The oxidation level is then plotted according to the extraction progress (Figure 4B). An extraction
 234 profile is also obtained for each recycled asphalt mixture specimen removed from each 20 kg formulation.
 235 Then, a corresponding index, BR (Blending Ratio) is defined as the ratio between the last oxidation level (for
 236 inner layers) by the first oxidation level (for outer layers) (Figure 4B). This parameter assesses the total
 237 binder homogeneity according to [39]: the closer to 1 the BR parameter is, the more homogeneous the
 238 profile and the total binder film are.

Figure 4: Determination of an extraction profile using the existing solvent-based progressive extraction method (A, B) [39] and introduction of the profile slope indicator (C)

Furthermore, the slope of the curve linear regression is introduced to discuss blending (Figure 4C). A low slope suggests a complete binder blending because it means that oxidative levels (of inner layers) are similar to those of outer layers (Figure 4B). In the next part, the slope and BR indicators will be used to better assess the binder blending (paragraph 3.2) and make possible the comparison with results of the microscopic statistical study (paragraph 4.1).

3. Results

3.1. Infrared imaging *chemomap* results

Table 3 displays the Curves Peaks Average (CPA) and Standard Deviation (CPSD) which have been calculated from statistical distribution curves peaks for each asphalt mixture formulation.

Table 3

Assessment of homogeneity (through CPSD values) and oxidation (through CPA values) for analyzed zones of each asphalt mixture incorporating RA (H40) Hot 40% RA, (W70) Warm 70% RA, (H100) Hot 100% RA, (W100) Warm 100% RA

Specimen	RA content (%)	Curves Peaks Standard Deviation (CPSD) (10^{-4}) (-)	Curves Peaks Average (CPA) (10^{-4}) (-)
H40	40.0	2	8
W70	70.2	4	9
W100	96.1	3	5
H100	96.1	6	13

In Table 3, the H40 asphalt mixture displays a CPSD value of 2×10^{-4} . H40 is considered as a homogeneous asphalt mixture according to the defined tolerance in paragraph 2.2. The CPSD values are increasing with the RA content for other formulations compared to the H40 specimen. The W70 asphalt mixture specimen is less homogeneous (CPSD = 4×10^{-4}) than the H40 one (CPSD = 2×10^{-4}). The CPSD value of the W100 is a little bit lower (3×10^{-4}) than the W70 one (4×10^{-4}), what suggests the W100 is slightly more homogeneous than the W70 one. The H100 formulation is the most heterogeneous asphalt mixture according to the infrared imaging *chemomap* method because its CPSD value is the highest (6×10^{-4}).

About the CPA evolution, the W70 asphalt mixture has a higher CPA value than the W100 one ($9 \times 10^{-4} > 5 \times 10^{-4}$), what means that W70 appears more oxidized than W100. The H100 hot asphalt mixture displays the highest CPA value (of 13×10^{-4}), what makes it the most oxidized recycled asphalt mixture. More variation of CPA values is observed between W100 and H100 mixes. For an identical RA content, the W100 warm mix displays a much lower CPA value in comparison with the H100 mix ($5 \times 10^{-4} < 13 \times 10^{-4}$).

In fact, the combination of CPA and CPSD results shows that the H100 mix is the least homogeneous and the most oxidized of tested samples according to the infrared imaging methodology. For this hot process, the oxidized aged binder amount available in the mix seems to govern the homogeneity level in the recycled asphalt mixture. More details about these blending results will be then thoroughly developed in the discussion.

To validate obtained results of this novel infrared imaging methodology, the same recycled asphalt mixtures formulations were also tested using the solvent-based progressive method.

3.2. Solvent-based progressive extraction method results

Results obtained with the solvent-based progressive extraction method are presented in Figure 5 for four asphalt mixture specimens of the study. The normalized oxidation level is plotted according to the extraction progress.

Figure 5: Comparison of binder extraction profiles of tested recycled asphalt mixtures: (BR) Blending Ratio, (H40) Hot 40% RA, (W70) Warm 70% RA, (H100) Hot 100% RA, (W100) Warm 100% RA

The binder blending is firstly assessed through the BR (Blending Ratio) values. Their comparison shows that the H40 and W100 specimens are more homogeneous than W70 and H100 according to the stage extraction method. The BR parameter decreases from 0.93 to 0.83 (H40 → H100) for the hot-mix process, which indicates a binder blending reduction. For the warm-mix process, the BR parameter evolves from 0.89 to 0.94 (W70 → W100) and suggests a more efficient binder blending.

To complete the binder blending assessment, the profile slope indicator was extracted from the extraction profile. [Figure 5](#) firstly shows a binder surface over-oxidation for the first leachates (extraction progress less than 15 %), which may be explained by laboratory manufacturing conditions or by the sample preparation. As it is present for all formulations, linear regressions of respective profiles have been performed from an extraction progress of 15 % and a big difference in the regression slopes is observed. By considering each manufacturing process on [Figure 5](#), warm-mix process results show that the W100 mix is more homogeneous than the W70 mix (slope: $0.002 < 0.004$). For the hot-mix process, [Figure 5](#) shows that both H40 and H100 mixes are homogeneous whatever the RA incorporated (slope equal to $|0.001|$). Nevertheless, the linear regression coefficients are lower (< 0.80) for H40 and H100 samples than for W70 and W100 samples. Observations about the degree of blending are more reliable for warm mixes than for hot mixes. This aspect will be particularly developed in the discussion.

For an identical RA content, the H100 slope (-0.001) is lower than the W100 slope (0.002), what means that the H100 is more homogeneous than the W100 according to the stage extraction method. This slope comparison is different from the BR evolution which would induce a more efficient binder blending for the W100 specimen (BR: $0.94 > 0.83$).

To better understand blending phenomena occurring in recycled asphalt mixtures, results of both methods are compared and combined.

4. Discussion

The objective was to customize a solvent-free methodology to better understand the blending in asphalt mixtures incorporating reclaimed asphalt. After the methodology was developed, the influence of the reclaimed asphalt content and the manufacturing process was investigated using four different recycled asphalt mixture formulations. Results from the developed microscopic *chemomap* and solvent-based methods were firstly compared.

4.1. Correlation between results of two methods

Using results of the *chemomap* ([Table 3](#)) and solvent-based methods ([Figure 5](#)), similar evolutions have been observed: H40 is more homogeneous than W70 and W100 is more homogeneous than W70. Correlations between both techniques have also been researched and are displayed on [Figure 6](#).

[Figure 6](#) displays the CPSD value (acquired by Imaging ATR) versus the extraction profile slope (obtained by the solvent-based progressive extraction method).

Figure 6: Correlation between blending indicators of two methods: (CPSPD) Curves Peaks Standard Deviation derived from the statistical treatment acquired by Imaging ATR; (extraction profile slope) derived from the progressive extraction method

Results show a first trend (in dotted lines) for the most of samples: the more the CPSPD increases, the more the extraction profile slope increased (Figure 6), indicating a poor binder blending and homogeneity in the total binder film. Even if more samples should be tested to validate the reliability of this trend, the correlation coefficient (of 0.96) remains satisfactory enough.

Concerning recycling issues, the case of LH100 asphalt mixture is an exceptional case here because the extraction profile slope is negative. In fact, the aged binder oxidation is lower than the virgin binder one during the manufacture. For the recycling technologies, this situation should not happen because it assumes that the RA aged binder (in inner layers on Figure 4) is less oxidized than the virgin binder (in outer layers on Figure 4). As the RA stock was identical for all mixes, this result may suggest an oxidation variability in RA stocks, already suggested by [18]. Anyway, this result is representative of real technical issues in plants and should be also taken into account in recycling studies.

Then, results of both methods are combined to determine the influence of each factor (process, RA content) in order to find correlations and explain blending evolutions.

4.2. Towards a better understanding of blending scenarii

Previous comparisons have shown similar tendencies in the binder blending evolutions between recycled asphalt mixtures specimens. The influence of the reclaimed asphalt content can be refined according to the manufacturing process:

- For the hot H40 and H100 mixes (H40 → H100), the CPSPD increases from 2×10^{-4} to 6×10^{-4} (Table 3), indicating a homogeneity reduction in the total binder film. This observation is in good agreement with the evolution of the BR parameter which decreases from 0.93 to 0.83 (Figure 5).
- For the warm W70 and W100 mixes (W70 → W100), the CPSPD decreases from 4×10^{-4} to 3×10^{-4} (Table 3), suggesting a binder blending improvement. This evolution is similar to the solvent-based method results because the BR parameter is higher and closer to 1 for the W100 mix ($0.94 > 0.89$) (Figure 5).

Whatever the method, these two observations show that the laboratory manufacturing process (temperatures, mixing conditions, etc.) determines the way reclaimed asphalt behave at high incorporation rates and affect the binder blending. For the warm process, homogeneity is improved for an RA increase whereas homogeneity is reduced for a hot process. That result is outstanding and similar to Ding's results

which have shown that a highest blending efficiency was observed for warm mix asphalts [33]. Other similarities are also observed on the fatigue resistance evolution because Zhao has namely shown that RA played a positive role on fatigue resistance when introduced in WMA mixes and a negative role when incorporated in HMA mixes [57].

These evolution differences being proved, blending results are then thoroughly discussed between model H100 and W100 mixes to better understand these evolutions and possible blending scenarii.

4.2.1. Influence of the manufacturing process for a 100 % recycled asphalt mixture

Thus, the comparison between H100 and W100 mixes shows that the warm mix process improves homogeneity because the CPSD is reduced (from 6×10^{-4} to 3×10^{-4}) in the *chemomap* method (Table 3). This result is confirmed by the BR parameter which is higher and closer to 1 for the W100 mix ($0.94 > 0.83$) (Figure 5). Both results should allow concluding that homogeneity is improved in the warm manufacturing process for a same RA content (of 100 %).

However, although a same RA content, the carbonyl index average is lower for the W100 mix (ICO average = 3.8) than for the H100 mix (ICO average = 4.4) (Figure 7).

Figure 7: Carbonyl indexes of leachates obtained by the solvent-based progressive extraction method for H100 and W100 asphalt mixture specimens

Although a lower binder over-oxidation in the warm-mix manufacture, the lower oxidation of the W100 specimen may also be explained by an analysis zone mostly composed of the less oxidized virgin binder. The mixing between binders would not be complete in defined laboratory manufacturing conditions, suggesting a lower RA binder remobilization.

This interpretation is possible and validated by the variation of oxidation levels on the extraction profiles (Figure 5). Indeed, for an extraction progress comprised between 20 and 50 %, the oxidation levels are higher for the hot mix process (H100). The oxidation level is centered on 0.86 (Figure 5) but some local oxidation fluctuations are observed between +7 % and - 4 % and impact the regression coefficient. This variation suggests that a larger amount in the aged binder is available in the mix but leads to a lower dispersion in the local zone, as illustrated by the local aged binder agglomerates (Figure 8A). For the W100 specimen, the oxidation levels are lower on the extraction profile (Figure 5) for an extraction progress

comprised between 20 and 50 %. The oxidation level is centered on the 0.83 value but a continued oxidation increase of 7 % is observed up to the 50 % extraction progress (Figure 8B).

Figure 8: Schematic illustration of the total binder homogeneity for H100 and W100 extraction profiles

In two cases, Figures 8A and 8B highlight microscopic chemical heterogeneities (of the one micron observation scale) which are present at different locations of the total binder film. They do not correspond to the optimum homogeneity case which was illustrated in Figure 3C.

Nevertheless, on Figure 7, the carbonyl indexes of H100 and W100 mixes leachates are between the virgin and aged binder ones (green and red curves on Figure 7), indicating that a partial mix between binders occurred for warm and hot asphalt mixtures incorporating 100 % RA. Furthermore, for the manufacture of warm-mix asphalt mixtures incorporating 100 % RA, it is shown that the aged binder remobilization is low because the W100 analysis zone by the *chemomap* method is mostly composed of the less-oxidized virgin binder (low CPA, Table 3). Therefore, the aged binder dispersion also appears as more homogeneous (low CPSD, Table 3) because less aged binder is available in the mix. That explanation is illustrated by Figure 9:

Figure 9: Illustration of blending phenomena for asphalt mixtures incorporating high rates of reclaimed asphalt and produced in laboratory according to hot (160°C) and warm (120°C) manufacturing temperatures

For warm-mix asphalt mixtures incorporating high rates of RA, blending scenarios would happen so that the virgin binder would not succeed in remobilizing totally (during the available time) high amounts of the aged binder. The reclaimed asphalt materials would behave as black rocks where important aged binder quantities would remain at the RA surface. To the detriment of the non-oxidized virgin binder, the aged binder would be less available in the intergranular bituminous mixture (Figure 9). Thus, for asphalt mixtures incorporating high rates of RA, the degree of remobilization (or mixing) would be less complete. This interpretation of a low RA binder remobilization in warm-mix asphalt mixtures is in a good agreement with a previous study [53] performed at the microscopic binder scale which had highlighted that remobilization was lower with the aged binder temperature reduction.

Therefore, a lower aged binder amount in the mix could explain obtained oxidation results (Table 3) such as:

- similar CPA values of W70 compared to H40 (8×10^{-4} and 9×10^{-4})
- lower CPA values of W100 compared to W70 ($5 \times 10^{-4} < 9 \times 10^{-4}$)
- lower CPA values of W100 compared to H100 ($5 \times 10^{-4} < 13 \times 10^{-4}$)

As the final oxidation is inversely dependent on homogeneity for the tested materials in the study, a lower contribution (by weight) of the aged binder in the mixture would lead to a better aged binder homogenization in the continuous medium of the virgin binder (Figure 9).

This explanation also shows that the amount of the RA aged binder activated in the mix has consequences on the resulting homogeneity and oxidation characterized on analysis zones of the asphalt mixture intergranular space. The degree of remobilization in the mix governs the degree of blending. This relation between mixing and blending steps appears clearly and is confirmed by Zaumanis who had affirmed that a good blending should be ensured by making the RA binder activated and available for blending [46].

Thus, as observed in Zhao and Nahar AFM studies [27], [44], the mixing (i.e. remobilization) and blending (i.e. homogeneity) of bituminous binders can be also distinguished thanks to the novel infrared imaging methodology. Nevertheless, compared to the AFM technique, the *chemomap* methodology presents the advantage to be directly applied on compacted asphalt mixtures and not to necessitate any consequent sample preparation procedure of samples as it was performed in [44], [58]. Indeed, compacted recycled asphalt specimens were only cut at required dimensions and chemical mappings were acquired at the tested sample locations where the crystal/sample contact was possible. Furthermore, the infrared imaging method provides a reliable chemical signature of the oxidation state due to the infrared carbonyl peak whereas the AFM topographic features of the asphalt binder and the bees structure may be influenced by external preparation factors and by the solvent extraction [59].

Therefore, this work highlights the validity and high sensitivity of the novel infrared imaging method to *in situ* assess the binder blending in asphalt mixtures incorporating RA. Based on the comparison between model H100 and W100 mixes, results also allow identifying the relation between manufacturing temperature conditions, mixing and hence blending results.

4.2.2. Identification of the temperature role on blending phenomena

Blending CPA and CPSD results (Table 3) have namely shown that the W100 mix was less oxidized and more homogeneous than the H100 mix. As their manufacturing conditions only differ by the RA heating temperature, it reveals that this has an impact on the aged binder amount activated in the mixing which leads to a different homogenization according to the sample. Thus, as RA materials were heated at 120°C for the W100 mix, the RA aged binder layer could not be softened and be satisfactorily released in the mix. It could not interact with the virgin second binder. The heating temperature of RA materials also governs both the degree of remobilization of the aged binder amount and the degree of blending of the whole recycled asphalt mixture.

In this study, the mixing temperature does not play a significant role for W100 and H100 mixes because it was measured and reveals that 96 % RA transfer its heat and temperature to the whole asphalt mixture. In relation to incorporated quantities, the heating RA temperature controls the mixing temperature for highly recycled asphalt mixtures. In addition, the mixing temperature does not play any role for the W70 and W100 mixes because it is identical to the RA heating temperature. W70 also appears more oxidized than W100 because RA materials were heated at a higher temperature (140°C) than W100 (120°C) leading to a higher remobilization of the aged binder in the mix.

This demonstrated influence of the RA heating temperature on the blending is different enough from results of other previous studies which prefer discussing the positive effect of a higher mixing temperature on the degree of blending [60] leading to determine the resistance of RA clusters to break [61].

Nevertheless, Wu has also suggested that the mixing temperature control could be implemented by adjusting the temperatures of in-material components [62]. Furthermore, it was more economical for a recycled mix asphalt mixture to use a higher preheated temperature of RA [62].

Therefore, the RA materials heating temperature should be considered to optimize the mix design and improve remobilization. However, RA materials are generally warmed at low temperatures between 110 °C and 150 °C [63] or at an approximate temperature of 130°C [64], [65] to minimize the binder oxidation. Other alternative solutions must be also researched to optimize the RA aged binder amount in the mix.

4.3. Perspectives of research

Some efforts should be undertaken to increase the remobilization degree referring to the RA aged binder amount available. In this way, the RA aged binder viscosity should be controlled because it plays a significant role on remobilization [53]. An increase in temperature also appears as an important factor to consider in the remobilization process. However, to minimize oxidation and maintain a low energy consumption during the warm-mix process, an alternative solution would be to incorporate chemical additives (such as rejuvenators) allowing sufficient softening of the harder RA binder. As discussed by [46], a specific industrial technique allows spraying a softening additive on RA materials before being mixed with other ingredients in the industrial plant.

On the other hand, the degree of remobilization should be predicted and used in blending laws for a better design of asphalt mixtures incorporating high percentages of RA. Recently, the degree of remobilization has been indirectly evaluated by the calculation of the statistical CPA (Curves Peaks Average), characterizing *in situ* the average oxidation [56]. This work must be pursued in order to calculate a percent of remobilization which should be introduced in blending and other prediction laws. That has partially been introduced by D. Lo Presti [23] who used a Replaced Virgin Binder (RVB) as the real blended percentage of RA binder in the construction of blending charts.

Another research perspective should be also oriented to the assessment of homogeneity in recycled asphalt mixtures because a lack of uniformity may lead to poor high- temperature anti-rutting performance and poor moisture stability of the recycled asphalt mixture [66]. As different blending phases may conduce to crack propagation [67], a poor low-temperature cracking resistance is also possible [66]. Up to now, few studies about the degree of blending (referring to homogenization) exist and are only focused at the binder micro-scale [27], [44]. Future work should be also be pursued to manage the determination of a blending percentage for different recycled asphalt mixtures. Furthermore, the influence of other external manufacturing parameters should be considered in the improvement of the degree of blending. For instance, the increase of the mixing time is an interesting option, as it seems to have a more significant effect on homogeneity of recycled asphalt mixtures [51], [61], [68].

Therefore, much effort must be provided both on the mix design procedures, on the formulation, and on industrial manufacturing conditions in plants. In this industrial multi-factorial issue, more experiments are still necessary to identify other factors such as the aggregate gradation curve, the moisture content, and the mixing energy which could affect the binder blending.

This work also opens novel perspectives to continue developing more complete characterization procedures and defining novel indicators to better assess and optimize properties of sustainable asphalt

mixtures incorporating high rates of RA. In this study, as remobilization, homogeneity, and oxidation constitute three complementary factors to better assess the blending, novel research perspectives should be pursued with infrared imaging to better design the production of homogeneous recycled asphalt mixtures displaying targeted oxidation properties.

Conclusion

Nowadays, the primary challenge for road construction companies is to produce durable asphalt mixtures incorporating high rates of Reclaimed Asphalt (RA). However, questions are still raised about the mix design and a fundamental knowledge of blending phenomena is also required. Based on infrared imaging microscopy, a sustainable statistical chemomap infrared methodology has also been successfully customized to the analysis of some recycled asphalt mixtures displaying different manufacturing processes and reclaimed asphalt content. Obtained results were compared with those of another solvent-based progressive extraction method to better understand blending phenomena. The study shows:

1. A correlation between results from two methods. Evolutions of the binder blending are similar and highlight that the laboratory manufacturing process (temperatures, mixing conditions, etc.) determines the way reclaimed asphalt behave at high incorporation rates and affect the binder blending.
2. The mixing and blending phenomena can be distinguished using the infrared imaging method. Remobilization, homogeneity, and oxidation constitute three complementary factors to better assess the binder blending.
3. For warm-mix asphalt mixtures incorporating high rates of RA, the aged binder remobilization is low, the analysis zone by the infrared *chemomap* method is mostly composed of the less-oxidized virgin binder, hence the aged binder dispersion is detected as more homogeneous in the intergranular space.

Therefore, promising first results about the blending phenomena have been obtained using both the novel infrared imaging methodology and the stage extraction. More recycled asphalt mixtures specimens containing different RA percentages should be nevertheless characterized to confirm the robustness of the methods and the blending evolution trends of this multifactorial issue.

However, this study already demonstrates the necessity to optimize manufacturing conditions in order to improve homogeneity while avoiding any situation of low remobilization in asphalt mixtures incorporating high content of reclaimed asphalt. The introduction of a degree of remobilization constitutes a first promising solution to take into consideration in the mix design improvement.

Acknowledgments

First acknowledgments are dedicated to the financial support, the French National Research Agency (ANR) in the framework of the national IMPROVMURE project (Innovation for Materials and Processes for Improving the Multi-Recycling of Mixes). Authors are also thankful to F. Delfosse and J-A. Decamps from EUROVIA Research Center (Mérignac, France) for the solvent-based progressive extraction method experiments and the help in the extraction profiles interpretation.

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