ON A LABORATORY EXPERIMENTAL PROTOCOL FOR QUALITATIVE CHARACTERIZATION OF HIGH RATE RECYCLED ASPHALT MIXTURES

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ABSTRACT

Nowadays, with the increasing material costs and the environmental needs to reduce the energy consumption and greenhouse gases emission, the technique of asphalt recycling becomes more and more used in road industry. To ensure the performance of recycled asphalt, a quality control of the bituminous mixture is required. The current study is carried out in the framework of a research industrial project in the partnership with EUROVIA Research Center. In this paper, we present an experimental approach to characterize the level of blending of the binder from reclaimed asphalt pavement (RAP) in hot recycled bituminous mixture. First, a series of laboratory tests are performed on asphaltic concrete including 70% of RAP to extract the bituminous binder using an appropriate solvent. The extraction test is done progressively to have three different solutions regularly spaced. Accordingly, the machine parameters are calibrated so that the first solution falls near 30% of the total extraction progress. In addition, bitumen is recovered from the different solutions to compute the associated concentrations obtained during the progressive extraction. Conventional tests are carried out on recovered bitumen to quantify the RAP and the virgin binders during the stripping process. Finally, we use infrared spectroscopy technique to have both quantitative and qualitative information of the bituminous binders used in the mixture.

Keywords: High rate recycling, RAP, bitumen mixture, homogeneity, characterization

1 INTRODUCTION

The asphalt industry is more and more interested by the reuse of high rate reclaimed asphalt pavement (RAP) for the construction of new road pavements. It is well known that the recycling process is very advantageous from environmental and economic point of views and contributes significantly to the sustainable development program. By the same token, in the case of high rate recycling, the recycled asphalt is composed of a high amount of reclaimed asphalt with modified chemical and rheological properties since it was aged in situ by the high load traffic and different climatic changes. As a matter of fact, the reclaimed pavement materials must be well combined with suitable virgin ones to ensure a successful functionality during service life, especially the mix of binders (virgin binder which can be a classic bitumen or a rejuvenator and the old binder derived from RAP). Accordingly, a quality control of the recycled asphalt mixture is required. During the past years, several degradations (cracking, pothole…) were noticed in sites where a high amount of RAP was recycled [1-3]. As a consequence, the homogeneity of the recycled mixture was questioned [4]. For over than thirty years, many research works have been developed to characterize the homogeneity of the mixture of virgin and RAP binders. For instance, [5] suggested a progressive extraction method based on 2 successive soakings of the recycled bituminous mixture in solvent. More recently, [6-8] presented quadruple soaking test and analyzed the recovered blend by chromatography technique using a permeable gel and also by rheological study. Recent research work [4] has discussed this problem and suggested a microscopic approach based on the observation of the spatial distribution of specific tracers in order to evaluate the level of blending of the hot recycled asphalt mixture. More recently, Navaro [9,10] has developed an experimental leaching test based on a continuous dissolution of the bituminous mixture in the tetrachloroethylene solvent by spraying in order to extract progressively the recycled binder. This original experimental system allows to carry out a gradual extraction of binder from a recycled asphalt with ten samples taken into 240 s. The author can determine the kinetic of lixiviation by UV attenuation. Then, a spectroscopy analysis of the different extracted solutions is performed to determine the evolution of the RAP binder content during the stripping process. This experimental system is too complex to be commonly used to control the recycled asphalt quality.
In this context and in the continuity of Navaro's study, a full-year study was carried out by the authors acting on behalf of EUROVIA research Center (Mérignac – France) in order to develop a fast experimental tool to control the recycled asphalt mixture quality. The objective of the present paper deals with the assessment of an experimental laboratory procedure to determine the degree of virgin binder blending in the total binder in recycled asphalt using a classic laboratory machine. This test consists on a discontinuous progressive extraction of the binder from the bituminous mixture at several steps. Two different approaches based on conventional test and FTIR spectroscopy analysis are used in this paper to determine the contents of virgin and RAP binders in extracted solutions.

2 EXPERIMENTAL PROGRAM

2.1 Material properties and Mix design

In this study, a bituminous mixture including 70% of reclaimed asphalt pavement (RAP) was prepared in laboratory. The mix design was considered such as the sieve curve of the recycled mixture is similar to a conventional asphaltic concrete (BBSG 0/10). The particle size distributions of Virgin, reclaimed and recycled aggregates are shown in figure 1. The Rap derives from a recycling platform in the South of France while the virgin aggregate is a 6/10 microdiorite.

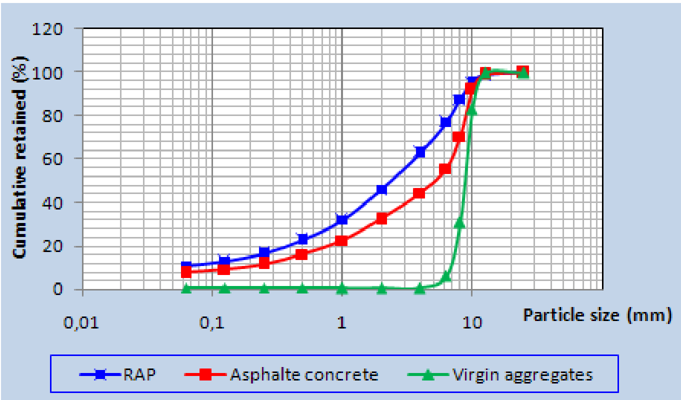

Figure 1: Particle size distributions of used aggregates

The mixing conditions were chosen to obtain a mixture supposed not homogenous : heating RAP at 110°C , virgin aggregates at 105°C, and new bitumen at 135°C, mixing the dry aggregates and RAP during 30 sec then bitumen was added and mixed to aggregates during 40 sec. The mixing temperature is about 110°C.

The richness modulus denoted “K” considered for the recycled mixture is taken equal to 3.6. This French classic parameter is proportional to the binder film thickness coating the aggregates and allows, with others parameters such as density and specific surface, the calculation of the bitumen content “BC” [11].

With our data, the total bitumen content in mixture BCmix is equal to 5.74% (by weight of dry aggregates).

On the other hand, the content binder BC1 of the reclaimed asphalt pavement used in this study is equal to 5.64% (by weight of dry aggregates).

Accordingly, the virgin binder content “BC2” shall be equal to:

BC2 = BCmix – BC1 = 5.74% – (70% x 5.64%) = 1.79% [1]

Given the binder contents of reclaimed and virgin binders, one can easily determine the proportions of RAP and fresh binders of the recycled mixture denoted respectively “a” and “b” by:

a = BC1/(BC1+ BC2) ; b = BC2/(BC1+ BC2) [2]

This leads to a = 68.8%; b = 31.2%.

The bituminous binder was extracted and recovered from the used asphalt pavement according to the European standards [12] and [13]. Then, the virgin binder was selected on the light of the recovered bitumen characteristics.

The mixtures rules applied to penetration and Ring and Ball temperature TRB are used here (Eq. (3)) to determine the grade of the final recycled asphalt. In this study, we aim to obtain a 20/30 penetration grade bitumen for the manufactured recycled asphalt.

$$\begin{cases} \log(pen_{mix}) = a.\log(pen_1) + b.\log(pen_2) \\ T_{R\&B_{mix}} = a.T_{R\&B_1} + b.T_{R\&B_2} \end{cases} \quad [3]$$

In table 1, the conventional properties of RAP and virgin binders used in this study are presented:

Table1: Characteristics of RAP and virgin binders

	“Pen” (1/10 mm)	R&B (°C)
RAP binder	14	72.8
Virgin binder	160	40.4

Given the data of table 1on a hand and the mix law (Eq. 3) on the other hand, the conventional penetration and softening point values of the blend are computed: (Pen)blend = 30 (1/10 mm) and ($T_{R\&B}$)blend = 62.6 °C. As expected, the final binder in the mixture would have similar characteristics than a 20/30 penetration grade bitumen.

2.2 Progressive extraction

In this section, an experimental laboratory procedure is assessed to control the homogeneity quality of the bituminous mixture (Virgin + RAP binders). The asphaltanalysator is a classic tool of road laboratory allowing to check the composition of asphalt or to analyze RAP. It is used to extract bitumen from the asphalt pavement by dissolving it in appropriate solvent. At the end of the extraction, one recovers separately the dry aggregates and the bitumen solution in solvent. This normalized procedure aims to determine the binder content in the asphalt sample. The extraction can be followed by a recovery test using the rotary evaporator to dissociate bitumen from the solvent by vacuum distillation. This operation is useful for the characterization of the binder. The asphaltanalysator parameters have been fixed to determine a procedure of extraction according the European standard [12].

Figure 2: The asphaltanalysator device

To achieve our objective, this conventional extraction method was modified and adapted to extract progressively the binder from the recycled bituminous mixture.

Our objective was to obtain during extraction 3 different solutions regularly spaced (with regards of the bitumen mass). The first one would be the smallest, but values around 30% of total extraction progress are acceptable.

This task has required the preparation of sample and the calibration of both machine program and parameters:

- Pre-heating of asphalt sample (duration and temperature)

- Machine parameters (Number of washing, number of drying, dissolution time, solvent filling time …)

Accordingly, a large series of progressive extractions were performed and followed by a recovery step of the first solution to get an idea on the stripping progress of the recycled asphalt.

Besides, the mixture preheating was also conditioned since it was observed that heating the sample at high temperature accelerates the stripping process. These different trials were useful to fix an experimental program for the asphaltanalysator exploitation.

At the end, it was decided to preheat a mixture sample of nearly 1500 g at 40°C during 2 hours before the progressive extraction. Then, in the program menu, 2 dissolutions of 3 mn are selected. The dissolution step enables a "prewash" of the asphalt mixture so that bitumen can be extracted in a progressive way. The solvent quantity necessary for dissolutions is governed by the spraying delay which was fixed to 20 sec. At the end of each dissolution, the program is stopped to recover the bitumen solution and then a final extraction is planned to strip completely the remained bituminous mixture. The last step of the progressive extraction consists of a classic extraction without dissolution. The selected number of washing and drying cycles are respectively 3 and 5.

At the end of the extraction, the dry aggregates and fillers are weight and subtracted from the overall asphalt sample to calculate the binder content in the mixture.

The complete extraction performed on the overall recycled asphalt sample (1500 g) using the conventional extraction method leads to 81 g of bitumen, this corresponds to the aforementioned BC of 5.4 % (by weight of total mix) considered in mixture preparation. Considering the variations on the binder content of the RAP, the obtained content corresponds to expected content. The application of progressive extraction procedure leads to 3 binder solutions to be analyzed.

2.3 Qualitative analysis of the blend

Two methods were used in this study to determine the homogeneity level of the blend:

1. The conventional Ring &Ball test: this technique requires the recovery of the different solutions obtained by progressive extraction. The recovery process of one solution lasts approximately 2 hours. Besides, the recovery test is very often followed by a spectroscopy analysis to ensure that solvent is completely evaporated from bitumen.

2. The FTIR spectroscopy analysis: this method allows the analysis of different solutions without bitumen recovery. Nevertheless, this technique requires the knowledge of the solutions concentrations and their adjustment to a common concentration.

The organigram of figure 3 summarizes the different steps considered in the experimental program.

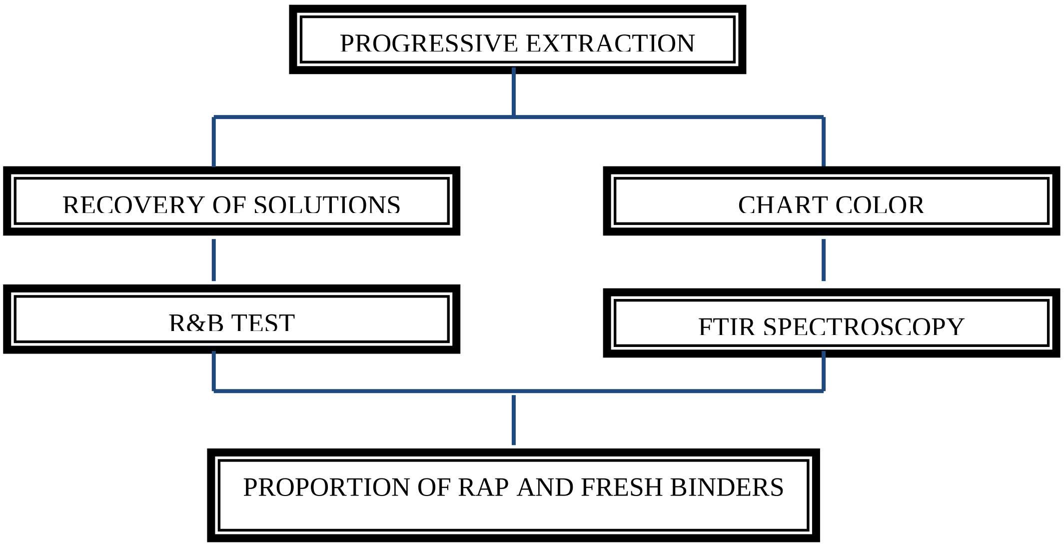

Figure 3: Experimental program

2.3.1 R&B test after recovered process

The binder recovered from the recycled asphalt (B_mix) [13] obtained by the conventional extraction method is characterized to determine its R&B temperature.

The binders recovered from the 3 solutions (B_Sol_i, i=1 to 3) obtained by the progressive extraction are characterized in order to determine the rates of virgin and reclaimed binders in the recycled mixture. Since the recovered binder quantity was not enough to carry out conventional penetration tests, only Ring and Ball tests were performed according to [14]. The results are summarized in table 2.

Table 2: Results of R&B test

	B_mix	B_Sol_ 1	B_Sol_2	B_Sol_3
R&B (°C)	59	52.8	61.4	63

One can note that the weighted average of the softening temperatures of the three solutions is very close to the blend one.

By dividing the mass of each recovered binder by the total bitumen mass in the sample mixture, one can evaluate the progress state of the stripping process.

Using the 2nd equation of system (3) and knowing the softening temperatures of fresh and RAP binders, one can calculate for every solution the proportion of the reclaimed binder in the recycled bitumen. More general, when considering the solution i (i = 1 to 3), the corresponding mass fraction of the RAP binder may be calculated by:

$$a_i = \frac{T_i - T_{VB}}{T_{RAP\_B} - T_{VB}} \qquad [4]$$

Thereby, the proportion of virgin binder in the solution i can be deduced as following: $b_i = 100\% - a_i$

2.3.2 Chart color and Fourier Transform InfraRed (FTIR) spectroscopy

FTIR spectroscopy was performed in this study using Nicolet iS10 FT-IR Spectrometer from thermo scientific. The IR cell made from KBr (Potassium Bromide) is filled with liquid sample. The FTIR test enables a qualitative and quantitative analysis of functional groups in the material. Spectroscopy test was carried out in transmittance mode in order to identify the chemical functionalities as well as ageing indicators in different solutions. As pointed out previously, this technique requires the knowledge of the solutions concentrations in a hand and their adjustment (generally by dilution in solvent) in order to enable the spectra analysis. Accordingly, a chart color with different concentrations has been developed so that solutions can be approximated.

3 RESULTS AND DISCUSSIONS

Using the data of table 2, the RAP binder content can be easily computed using Eq. 4.The results are summarized in table 3.

Table 3: RAP content values during the progressive extraction

	Stripping rate (%)	R&B (°C)	B_RAP content (%)
Sol_1	26	52.8	38.3
Sol_2	61	61.4	64.8
Sol_3	97	63.0	69.7

In addition, it can be shown from the results of table 3 that during the 1st dissolution, virgin binder is stripped more than the RAP one. This can be due to the “bad” remobilization of virgin binder in the blend. In the case of a “perfect” homogenized recycled mixture, the RAP binder content would have been uniform during the extraction and equal to 68.8%.

Furthermore, a comparative study is also carried out using a spectroscopy approach. This technique is advantageous because it preserves the sample integrality and offers the possibility for experiment without altering the material properties.

For the prediction of RAP binder during the stripping process, the Beer-Lambert rule given by Eq. (5) is useful to establish a linear relationship between absorbance and concentration.

$$\mathcal{A}(\lambda) = log\left(\frac{I_0}{I}\right) = \varepsilon(\lambda).C.d \qquad [5]$$

Where A is the absorbance, λ the wave length (m), I_0 (resp. I) the incident light intensity (resp. transmitted light intensity), ε the absorption coefficient, C the solution concentration and d the length of IR cell (0.5 mm).

To ensure the exploitation of Beer's law within the linearity domain, the absorbance maximum value shall be in the range 1-2 [15]. Accordingly, to perform IR measurements, the bituminous solutions derived from the progressive extraction were adjusted to a mass concentration close to 0.8%. This supposes that the mass concentration of a given solution shall be known. To achieve this task, a color chart (Fig. 4) was developed in laboratory by dissolving bitumen into the perchloetylene at different and known proportions. This enables to have different controlled concentrations "C". It was noticed that a solution of 0.8% is very concentrated and this will not make easy the approximation of different solutions. Consequently, the chart was prepared with more diluted solutions (0.08% of maximum concentration).

Then, the solutions obtained by progressive extraction were compared visually to the chart color and their concentrations adjusted (generally by dilution) to be very close to a bituminous known concentration of the color chart. At this primary stage, one can evaluate the real concentration of the bituminous solution by computing the product of C by the dilution factor. Thereby, ones the solution concentration is known, dilution is performed to make the solution at 0.8% for analysis spectroscopy.

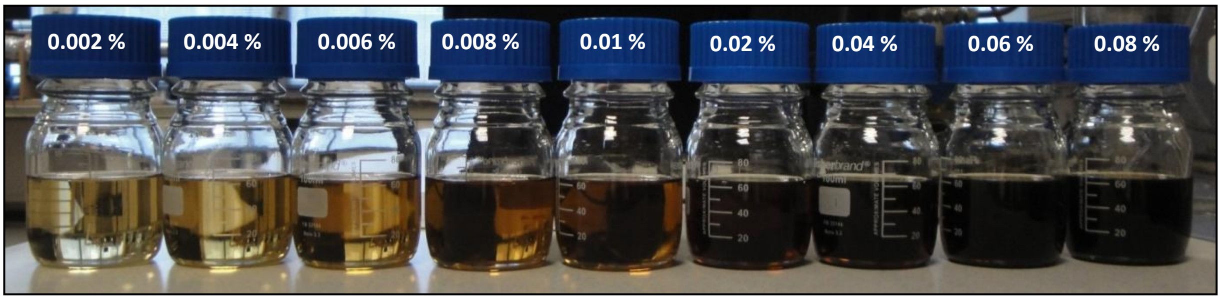

Figure 4: Bitumen chart color

The plots of figure 5 present the concentrations of the three solutions obtained by discontinuous extraction. Note that the second and third solutions are concentrated (Ci > 0.8 %), thereby a dilution was performed in order to adjust them to the wished concentration whereas the first solution was evaporated so that the concentration is increased to nearly 0.8 %.

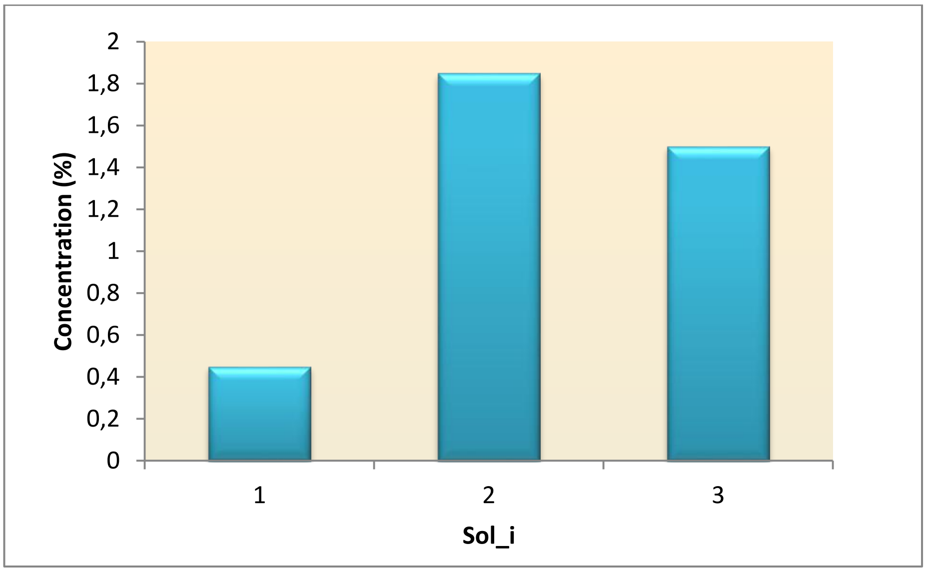

Figure 5: Solutions concentrations

It can be noticed from Figure 6 that the IR Spectra of RAP and virgin binders present intense band at nearly 2950 cm-1 that can be attributed to aliphatic C-H stretching and deformation vibrations. The oxidation of RAP binder is characterized by the apparition of carboxyl (C=O) and sulfoxyd (S=O) chemical functions respectively at the wave numbers 1700 cm-1 and 1045 cm-1. The virgin binder spectrum doesn't exhibit high peak at these wave numbers [16].

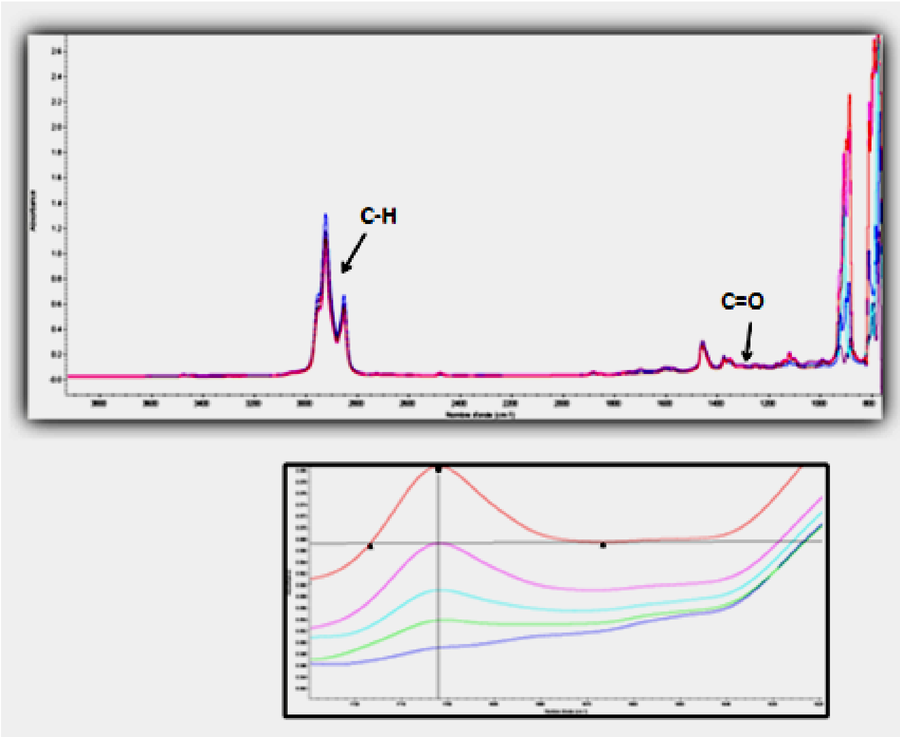

Figure 6: On the top: IR Spectra of virgin binder (blue), RAP binder (red) and 3 solutions with 26%, 45% and 77% of RAP, on the bottom: Zoom of Carbonyl peaks of IR spectra

Furthermore, it was noticed that the 3 solutions had not the same concentrations of 0.8% since the latter were estimated by the approximate visual method of the color chart. In order to compare the different IR measurements, it was decided to normalize the spectra with regards to the C-H peak area. Then, to determine the RAP binder content in the recycled blend, we make use of the called IPAD method developed by [9] and inspired by the previous research works of [16] and [17]. This technique consists on the definition of an ageing index by the calculation of the spectra right slope "IPAD" (Eq. 9) between the wave numbers ν1 and ν2 corresponding respectively to the carboxyl peak (1700 cm-1) and the zero of the first derivative of the RAP solution spectrum (Figure 7).

$$IPAD = \frac{(\Delta\mathcal{A}_{Sol} - \Delta\mathcal{A}_{VB})/\Delta\nu}{(\Delta\mathcal{A}_{RAP} - \Delta\mathcal{A}_{VB})/\Delta\nu} \quad [6]$$

This method offers the advantage to overcome the base line difficulty currently observed in spectroscopy analysis. It was demonstrated that this index represents the RAP binder content in the recycled blend. In fact, 3 solutions were freshly prepared in laboratory with known proportions of RAP binder (26%, 45% and 77%) and virgin binder (Resp. 74%, 55% and 23%). The analysis by FTIR spectroscopy has shown that the IPAD computed values are respectively equal to 24%, 41% and 68%. For further information about this technique, the reader could consult [10].

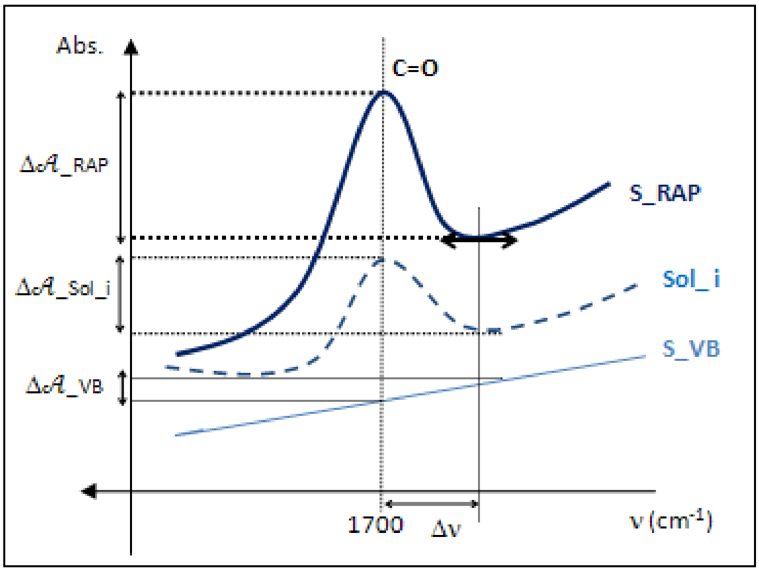

Figure 7: **Schematic representation of IR spectra for the IPAD calculation**

The 3 solutions obtained by progressive extraction were analyzed by FTIR spectroscopy. The content of RAP binder in the recycled blend is calculated by Eq. (9) and the results are given in table 4.

Table 4: **RAP binder content by FTIR spectroscopy**

	Sol _1	Sol_2	Sol_3
B_RAP content (%)	32	58	70

One can notice that the FTIR spectroscopy analysis gives comparable results with regards to the aforementioned R&B conventional approach. The recorded error between the 2 methods is less than 11%.

4 CONCLUSIONS

A progressive extraction test using a classic road laboratory machine is presented in this paper. A calibration step of the extraction machine program parameters is successfully accomplished. Three bituminous solutions are obtained and then recovered to determine the RAP binder content during the extraction. In addition, a spectroscopy analysis is performed to investigate the ageing level of the extracted solutions by the means of chemical oxidation identifiers. In view of the obtained results, it was noticed a heterogeneity in the recycled bitumen mixture as hypothesized at the beginning of this study and especially during the laboratory mixture manufacture. In the future, it would be interesting to investigate the influence of the process mixing on the recycled mixture quality, such as the material preheating temperature, the mixing duration, etc. Additionally, mechanical tests have to be performed to compare homogeneous and heterogeneous mixtures in order to study the influence of heterogeneity aspect on the asphalt rheological properties.

Acknowledgments

The authors would like to thank gratefully Carine FUCHS, Arthur MILLE, Jonas LEGALL and ZACCHARIA ETTAOUIL, engineering students in the ESTP for their active collaboration in this project. The authors acknowledge also Margareta WALFERDEIN from ESTP/IRC for her experimental assistance.

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