Results of interlaboratory tests on a laboratory bituminous mixtures ageing protocol

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ABSTRACT: RILEM TC-ATB- working group TG5, who is in charge of “recycling of bituminous materials” has decided to develop a laboratory procedure with the aim of reproducing the ageing of bituminous materials until the end of their service life (milled product). This procedure is meant to assess the recyclability of mixtures through production of Recycled Asphalt Pavement (RAP) in the laboratory in a realistic way. With the chosen aging protocol (divided into short and long-term ageing), the results of interlaboratory tests are presented. A bituminous material with a 35/50 penetration pure bitumen, taken from an LCPC fatigue carrousel experiment is submitted to the ageing protocol. The recovered bitumen is analysed by means of classical empirical tests (penetration and ring and ball), rheological tests (complex modulus) as well as a physico-chemical test (infra red spectroscopy) to asses the binder evolution throughout ageing. The results coming from the round robin test show that the ageing of the plant mix is similar to the ageing of the mix manufactured in the laboratory and subjected to the short-term ageing protocol, for all the indicators tested (penetration, ring and ball, DSR and FTIR). The mix subjected to long-term ageing evolves until the 9 days of the protocol and seems to lead to a considerable more aged binder compared to the classical ageing tests on binders.

KEY WORDS: lab ageing, mixture, RAP, rheology, chemistry.

1 INTRODUCTION

Task Group 5 (TG5) “recycling of bituminous materials” of the RILEM Technical Committee -Advanced Testing of Bituminous Materials– (ATB) focuses on the increase of the percentages of RAP in new asphalt mixtures. In order to predict and improve the knowledge of the potential to recycle old bituminous materials and the influence of their use on the performance of new bituminous materials, specific laboratory tests have to be developed. TG5 has decided to focus on the possibility to study recyclability of mixtures and RAP production in the laboratory. In order to achieve this goal, a laboratory procedure has been developed, aiming at reproducing in the laboratory the ageing of bituminous materials until the end of their service life (milled product). The testing protocol chosen is shortly introduced (de la roche, 2009) and most of the results of the round robin are presented and discussed in this paper. In more detail the penetratrion/softening point-, DSR- and FTIR results are discussed.

The round robin test was still running at the time of the writing of the paper. 7 of the 12 participating laboratories have reported their results. Therefore some trends can already be drawn from the available results. The complete analysis of the round robin will be published after finalizing all testing and analysis.

2 MIXTURE AGEING PROTOCOL

2.1 Material tested

The mix composition is given in Table 1. Loose mix (750 kg) has been sampled in the paver (this is called Batch 3). Aggregates and binder have been sampled (2950 kg) from the mix plant according to the mix composition in Table 1 (the lab produced mixture is called Batch 1). Homogenization of aggregates was necessary for the lab materials.

Table 1: Components and characteristics of the material tested

Component		Proportions
0-2 mm	Quarry Brefauchet	34 %
2-6.3 mm	Quarry Brefauchet	16 %
5.6-11.2 mm	Quarry Brefauchet	49 %
Fillers		1 %
Pure binder 35/50(external percentage)		5.7 %

2.2 Ageing Protocol and Testing Programme

The mixture ageing protocol chosen for the round robin test takes into account three steps which have to be consistent for all laboratories: the production of the mixture, the ageing of the mixture and the binder extraction and recovery. The protocol is described extensively in (de la roche, 2009). The protocols for short and long-term ageing are:

- For the short term ageing process, the mix is placed in an air-draft ventilated oven for 4 hours at 135°C. Each hour the material is stirred for 1 minute and placed back into the oven. This stirring action is only for homogenisation.
- For the long-term ageing, the short-term aged mixture is placed in an air ventilated oven at 85°C for 9 days. After 2, 5, 7 and 9 days the mixture is stirred before sampling.

The testing program consists of performing rheological (penetration, softening point, DSR) and chemical (FTIR) tests on the recovered bitumen after each ageing step.

In the paper the results for each step (phase) are numbered from day 2 according to the days of ageing. The binder which is used in the asphalt mixture is also aged according to two standard ageing procedures: 1) RTFOT and PAV and 2) RCAT short-term and long-term ageing. The aged binder is also evaluated with penetration, softening point, DSR and FTIR.

3 PENETRATION AND SOFTENING POINT RESULTS

Seven participants have performed penetration and softening point tests for Batch 1 corresponding to laboratory manufacture. For Batch 3, the production plant mixture, six participants have reported penetration and five reported softening point.

3.1 Penetration and Softening Point – Batch 1 (laboratory produced)

In Figure 1 left, an overview is given of the mean results of the penetration tests for Batch 1 without any statistical rejection of outliers. The mean results are calculated as the mean value of 2 samples per step. Laboratory 8 has tested one sample only. In general, the results show a good reproducibility of the production, ageing test and recovery/extraction. A parallel shift between labs is observed throughout the ageing test. Lab 6 shows a continuous offset of 5 for the penetration. The penetration data for lab 10 at step phase 5 and 7 show a large deviation from the mean curve. More detailed observation is necessary.

The softening point results show a good reproducibility (better than for penetration) of the ageing test and the recovery. In general a parallel shift-factor throughout the ageing test can be observed between the labs.

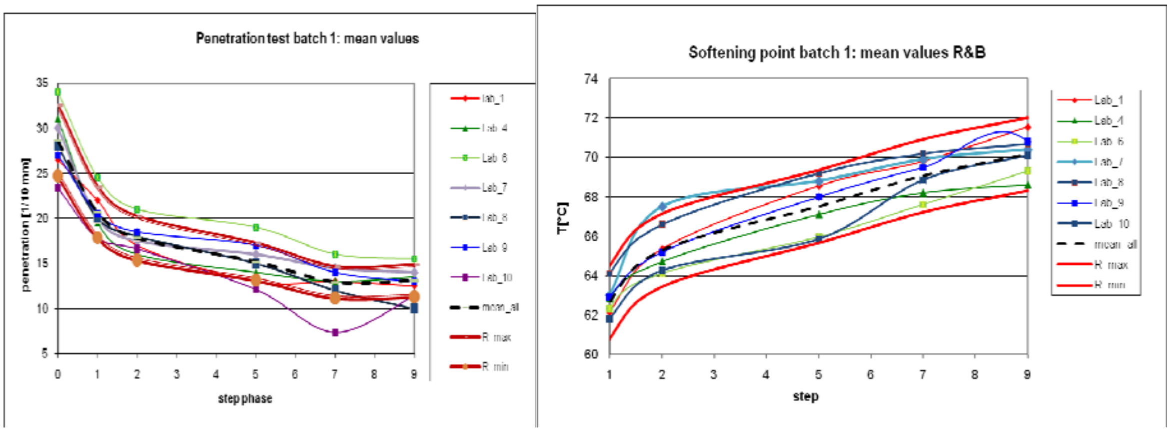

Figure 1: Penetration (left) and Softening Point (right) results of Batch 1 (laboratory manufacturing) for all laboratories

3.2 Statistical Results of Batch 1 and Batch 3

In table 2 an overview is given of the data for penetration of batch 1 with its mean, number of samples, standard deviation and reproducibility interval (R = 0,27x pen). In table 3 the mean data of the softening point are given for each step with its standard devation. For the softening point the results can be considered in the light of the R-value giving the reproducibility limit of the softening point test according to the European standard for binder extraction and recovery: 3.7°C.

In table 4, an overview is given of the mean results of the penetration tests for Batch 3 without any statistical rejection of outliers. It is remarkable that the deviations compared to batch1are larger (plant manufactured mixture). In table 5 the mean data of the softening point are given for each step.

Table 2: Penetration data statistics for Batch 1: all data

	Step	Mean	N	STDEV	R_int	R_min	R_max
virgin binder	-1	35,7	3	0,5	9,9	30,9	40,5
after mixing	0	28,6	13	3,5	7,7	24,7	32,5
after ST	1	20,7	13	2,2	5,6	17,9	23,4
after 2d LT	2	17,8	13	2,1	4,8	15,4	20,2
after 5d LT	5	15,2	13	2,4	4,1	13,1	17,2
after 7d LT	7	12,9	13	2,6	3,5	11,2	14,6
after 9d LT	9	13,1	13	2,4	3,5	11,3	14,8

Table 3: Softening point data statistics for batch 1: all data

	Step	Mean	n	STDEV	R=3,7	R_min	R_max
virgin binder		54,2	3	0,05	3,7	52,3	56,0
after mixing	0	57,5	13	0,9	3,7	55,6	59,3
after ST	1	62,6	13	0,7	3,7	60,8	64,5
after 2d LT	2	65,3	13	1,2	3,7	63,4	67,1
after 5d LT	5	67,5	13	1,3	3,7	65,7	69,4
after 7d LT	7	69,1	13	0,9	3,7	67,2	70,9
after 9d LT	9	70,2	13	1,1	3,7	68,3	72,0

Table 4: Penetration data statistics for batch 3: all data

	step	mean_sing	n	STDEV	R=0,27pen	R_min	R_max
virgin binder		35,7	3	0,5	9,9	30,9	40,5
after mixing	0	21,4	11	2,9	5,8	18,5	24,2
after ST	1	18,2	11	3,0	4,9	15,8	20,7
after 2d LT	2	16,3	11	2,6	4,4	14,1	18,5
after 5d LT	5	13,7	11	3,4	3,7	11,8	15,5
after 7d LT	7	12,9	11	2,8	3,5	11,2	14,7
after 9d LT	9	12,7	11	3,9	3,4	11,0	14,4

Table 5: data softening point batch 1: all data

	step	mean_sing	N	STDEV	R=3,7	R_min	R_max
virgin binder		54,2	3	0,05	3,7	52,3	56,0
after mixing	0	59,8	9	0,4	3,7	57,9	61,6
after ST	1	61,8	9	1,0	3,7	60,0	63,7
after 2d LT	2	63,8	9	1,3	3,7	61,9	65,6
after 5d LT	5	66,0	9	1,6	3,7	64,2	67,9
after 7d LT	7	67,6	9	1,6	3,7	65,8	69,5
after 9d LT	9	68,8	9	1,6	3,7	66,9	70,6

3.3 Tests on binder

Table 6 Results of ageing tests on virgin binders. Penetration and Softening point

	Lab 5		Lab 6	
	Pen(1/10mm)	R&B(°C)	Pen (1/10mm)	R&B (°C)
Virgin binder	35	54.1	36	54.2
Phase 0, batch 1	28	58.8	27	58.1
Phase 1, batch 1	20	64.1	20.5	62.9

Two labs have also tested the penetration and softening point on the virgin binder. In table 6 the virgin binder characteristics are compared to phase 0 (after production) and phase 1 (after short term ageing). Both steps are in the same order of ageing.

4 DSR RESULTS ON RECOVERED BITUMEN

4.1 Complex modulus

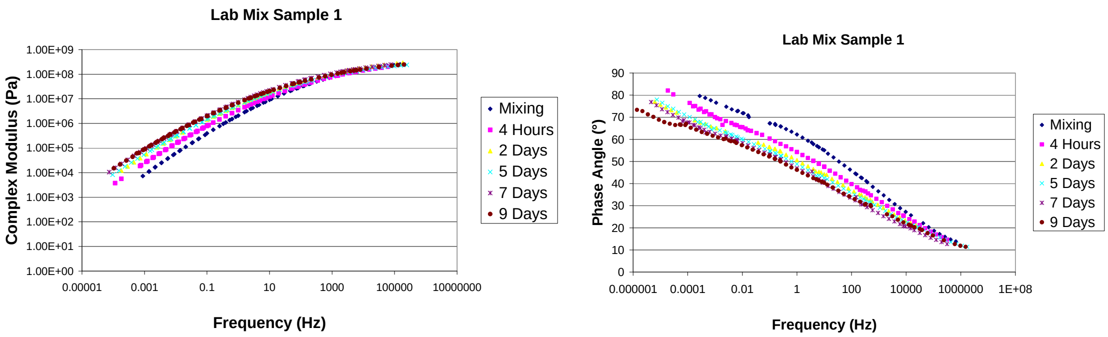

Figure 2: The effect of the ageing steps on the G* (left) and phase angle (right) master curve of Batch 1 at 25 C.

Frequency sweeps were carried out from 0.1 to 10 Hz at 11 different frequencies using DSRs over a range of temperatures (-5°C to 55°C at 10°C intervals). These were carried out in order to be able to construct Master Curves of both Complex Modulus and Phase Angle at specific reference temperatures. Master curves were then constructed using 25°C as the reference temperature.

Figure 2 shows the effect on the Complex Modulus and phase angle Master curves of the ageing steps applied to Batch 1. It can be seen that the G* increases with each ageing step. The Phase Angle is decreasing with each ageing step. A similar trend has been observed for Batch 3 (plant mix).

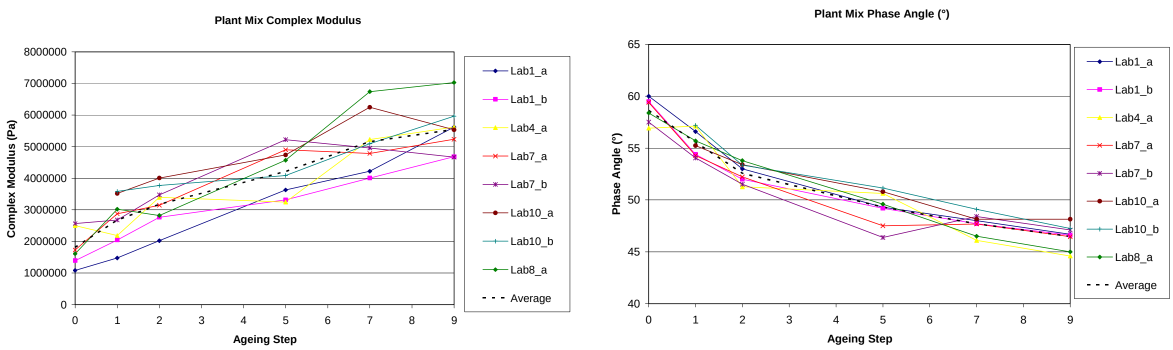

Figure 3: Impression of the Complex modulus and the phase angle at 0.4 Hz and 25 C for Batch 3 of the different labs.

Figure 3 shows the effect of ageing step on the G* and phase angle of Batch 3 at 25 C and 0.4 Hz for all labs. Again it can be seen that the mean G* increases with each ageing step from approximately 1.8 x 10^6 after mixing to 2.7 x 10^6 after short term ageing. The mean value then increases to 3.2 x 10^6, 4.2 x 10^6, 5.1 x 10^6 and 5.5 x 10^6 after 2, 5, 7 and 9 days respectively. The mean value of the Phase Angle is 59° after mixing and drops to 55° after short term ageing. It then decreases to 53, 49, 48 and 46 after 2, 5, 7 and 9 days respectively.

5 INFRARED SPECTROSCOPY RESULTS

In this paper results of 5 laboratories are reported for batch 1(the laboratory mix). Results of 4 laboratories are reported for batch 3 (plant mix). Several tests on the original binder, after RTFOT and after PAV have also been carried out. It has to be noticed that the frequencies used to define the different areas are exactly the same for all the tests. An extensive description of the method is given in (de la Roche, 2009) and (Mouillet, 2009). The results are also different compared to (de la Roche, 2009)

5.1 Results for Batch 1 and Batch 3

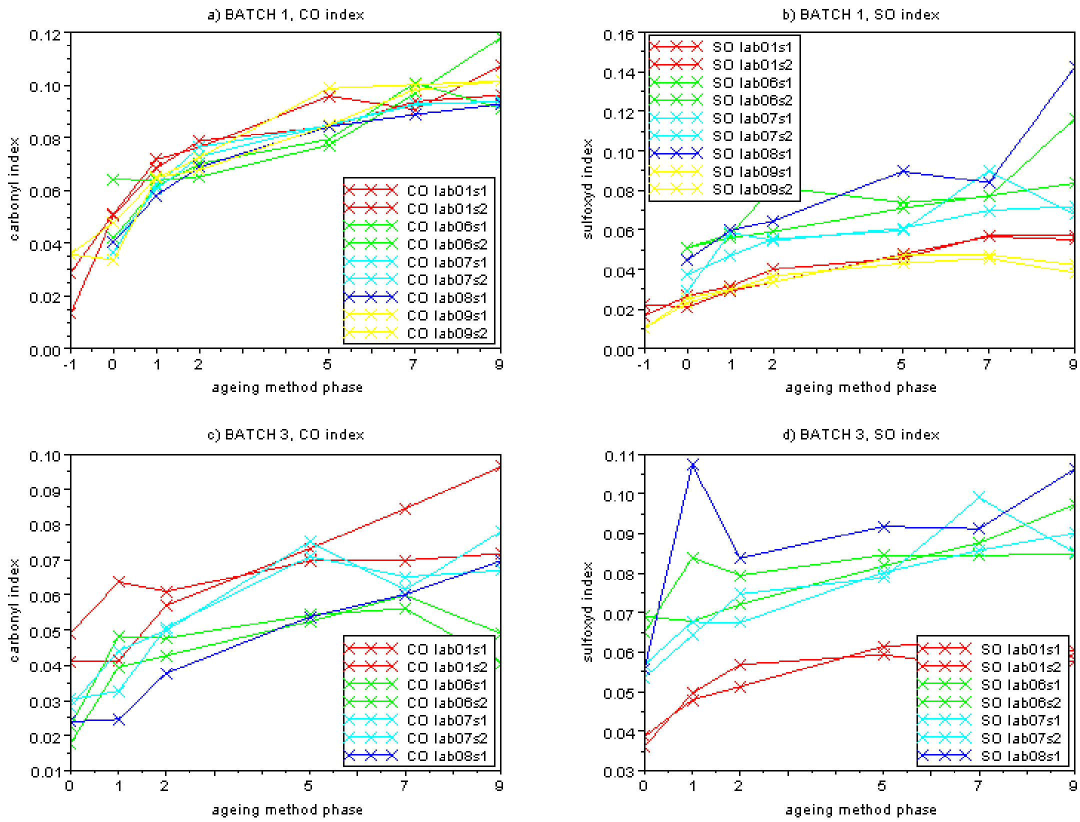

Figure 4: CO and SO indexes, for batch 1 and 3 as a function of the ageing phase

Results of oxidation indices are presented in figure 4. Five Laboratories have carried out tests on the lab mix (batch1). The step “-1” corresponds to tests performed on original binder. In 4a) the carbonyl index (Ico) for batch1 is given during the ageing procedure. For each laboratory, it can be noticed that results of similar samples are very close, highlighting a good repeatability for each lab on this batch and for this index. However, the comparison of all the results shows scattering for the carbonyl index. In figure 4b) the sulfoxide index (Iso) for batch1 is presented. For each lab, the comparison of similar samples also shows a good repeatability, despite some scattered values like: Lab06, steps 2 and 9; Lab07, step 7.

Iso results are less reproducible than Ico results. Fillers in the recovered bitumen may explain the more important scattering, because of its influence on the calculation of the Iso index. The presence of filler inside bitumen may interfere with the calculation of Iso, because sulfoxide and filler areas in spectra are very close. A second explanation may be that there is less sulfoxides creation than carbonyls creation.

Four laboratories have done ageing tests on batch 3. The results are more scattered than for batch1 (see figures 4c and 4d). They show a lower slope in time for the Ico index compared to Batch 1. The Ico curves are not smooth. Lab01 results are not as constant as the other ones. Lab06 and lab07 are quite repeatable despite their non-monotonic evolution. This uneven increase remains hard to explain at this time.

The results for the sulfoxide index are given in figure 4d). They all show increasing values and are repeatable but less reproducible. However, an artificial offset of curves at step 0 would strongly improve the repeatability of results. At step 1, the very high value of Iso for lab08 can be explained by the presence of filler inside bitumen.

6 DISCUSSION

6.1 Penetration and Softening Point

In general it can be concluded that the results are quite consistent taking into account the mixing procedure, ageing procedure, extraction/recovery and test methods. The shifting is probably due to differences in methods.

To show the trends the data are expressed in relative values normalised at 1 for phase 0 corresponding to sampling after manufacturing. In figure 4 (left) an overview is given of the normalized penetration for Batch 1 (lab mix) and in figure 4 (right) an overview of the softening points of batch 3.

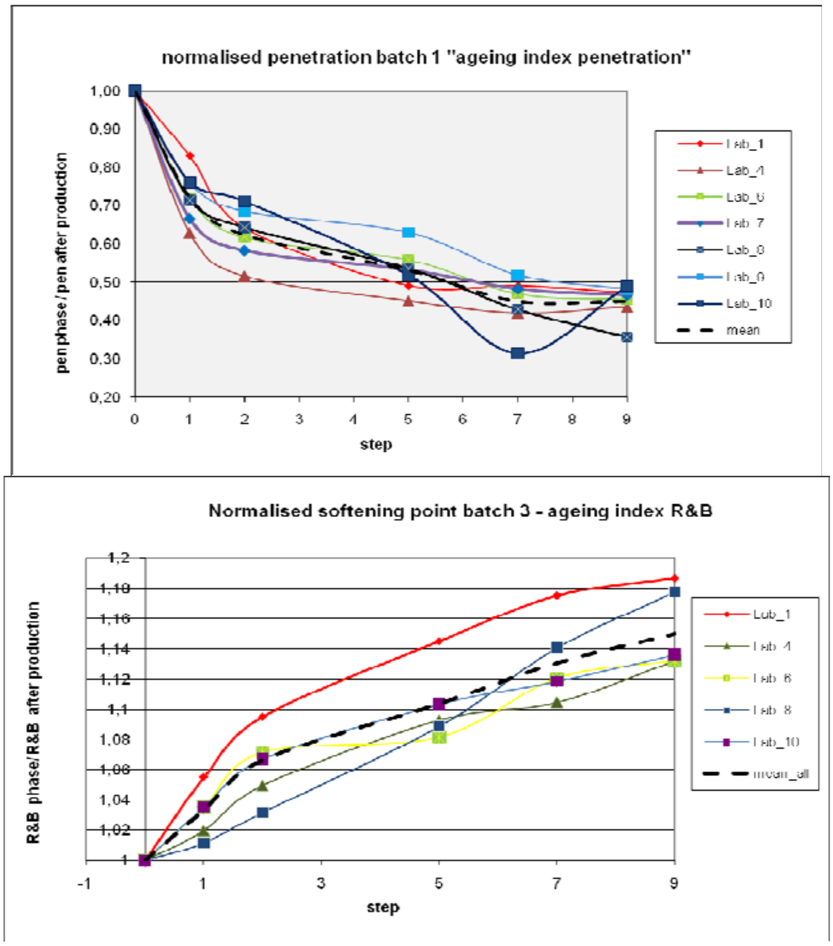

Figure 5: Normalised penetration for batch 1 (top) and softening point for batch 3 (bottom).

The strange points for the penetration in figure 4 need to be checked.

In general the softening point shows a reasonable trend for Batch 3 as can be seen in figure 5 (right). From figure 5 (left) it can be concluded that, based on the mean value it can be concluded that the penetration is not decreasing any more after 7 days. From figure 5 (right) it can be concluded that the softening point shows a constant slope after 2 days and that this slope is still measured after 9 days.

6.2 DSR

The master curves for the complex modulus and the phase angle show logical trends for the results received. In each ageing step the complex modulus increases and the phase angle decreases. For the DSR results a comparison is made between the lab mix, plant mix and the standard ageing tests on bitumen, see figure 6.

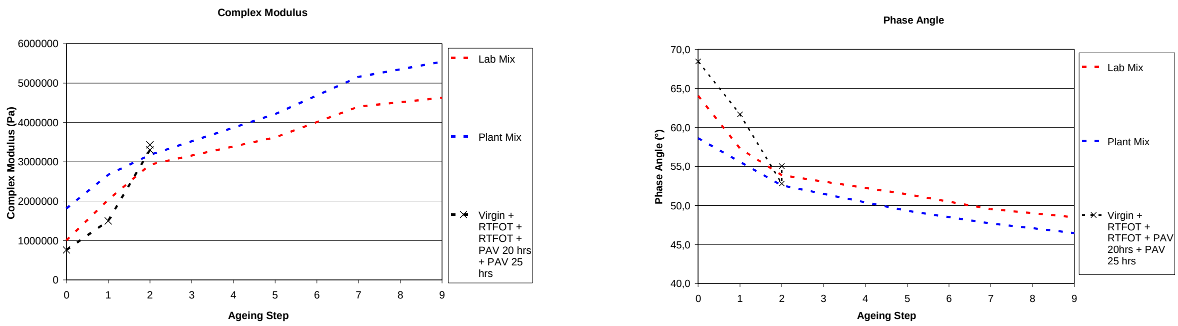

Figure 6: Comparison between ageing results of Batch 1, Batch 3 and pure bitumen at 25 C, 0.4 Hz. Left complex modulus and right phase angle.

From the results it can be seen that Batch 3 shows in all phases a significant higher complex modulus and significant lower phase angele compared to Batch 1. A first indication can also be given of the effect of lab ageing of pure bitumen. It seems that the standard lab ageing can not predict better than 2 days of plant mix ageing and between 3 to 5 days of lab mix ageing. From both graphs it can be seen that the slope of the curves is decreasing considerably after 3 to 5 days, but even after 9 days an increase in complex modulus can be observed.

6.3 FTIR

In figure 7 the results of batch 1 and 3 are compared. Only lab01, lab06, lab07 and lab08 have performed tests on batch 1 and 3. Only mean values for these four labs are presented here.
The results show that the slopes of Ico and Iso indices for Batch 1 are larger than for Batch 3. This is probably caused by the prematurely ageing of Batch 3. However, if we consider the absolute values of the carbonyl index, specimens of batch1 seem to be more aged than specimens of batch3. The absolute value of the Iso index shows exactly the contrary. The explanation for this still has to be discovered and the warming is, be careful.

In figure 8 a first comparison is given with the standard ageing procedures on fresh bitumen based on RTFOT and PAV. At this time, only the results of lab09 have been analyzed. RTFOT results have been arbitrarily located at step 1 (short term ageing step). PAV results (long term ageing) has been fixed at step 2. A first comparison between lab05 and lab09 in terms of RTFOT and PAV results shows a similar trend. At step -1, results of tests performed on the original binder by lab01, lab05 and lab09 can be compared. As the material is supposed to be the same for the different labs, the scattering gives an indication of the reproducibility of FTIR results.

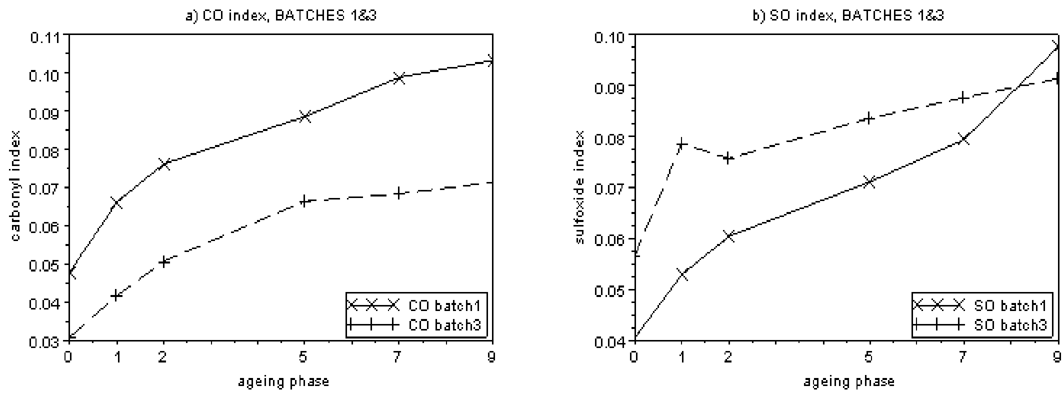

Figure 7: Comparison between results of batch1and batch3

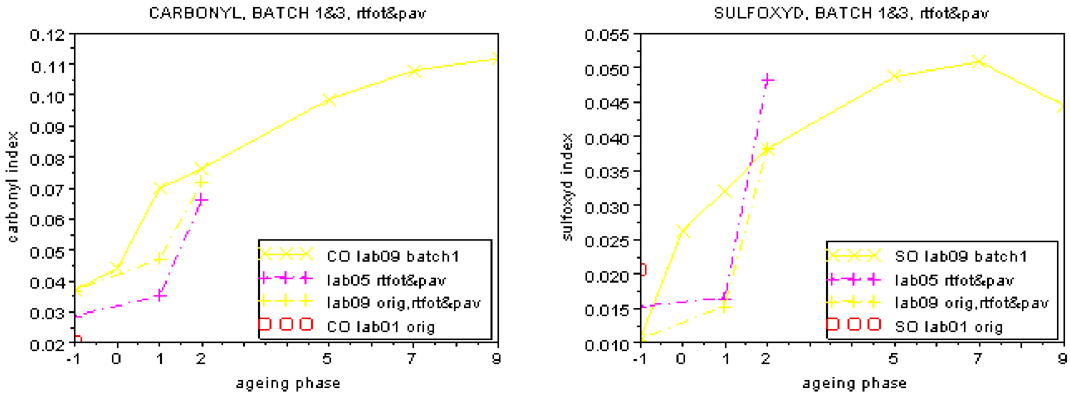

Figure 8: Comparison between batch1lab09 and pav and rtfot from lab09.
comparison between PAV and RTFOT from lab09 and PAV and RTFOT from lab05

7 CONCLUSIONS

At this point of the project with the received data the following preliminary conclusions can be drawn.

The round robin shows clearly tha variation that can be expected when testing an ageing protocol at different laboratories. The variation includes mixing, ageing, recovery of bitumen and test methods.

When the shift at the start is neglected, most test results show a similar trend.

No clear difference can be observed for penetration and softening point of the laboratory mixed and plant mix aged material. After 9 days the penetration value and softening point of the aged samples indicate that the binder still ages from a rheological point of view although the rate of this ageing decreases.

For penetration the reproducibility of the ageing test is not fulfilling the requirements regarding the R-values according to EU norms. As mentioned the deviations in the results are the sum of the deviations for mixing, ageing, the recovery procedure and the penetration test.

From DSR tests it seems that the plant mixed material gives higher stiffness and lower phase angles than the laboratory mixed material.

FTIR results show a good repeatability, but less reproducibility. Ico results are more consistent than Iso results. The comparison of tests performed on the same original binder has shown considerable difference in FTIR results. Concerning the ageing procedure, it seems that reproducibility may be better when using an offset. A comparison of the FTIR results between batch 1 and batch 3 shows that batch 3 seems to be less sensitive to ageing. Also a difference between the levels of Ico and Iso for lab mixed and plant mixed material is observed. Analyses of others result are still being performed and will probably help to explain these results.

For all tests the ageing rates are (strongly) decreasing with time of ageing. It seems that in most cases after 7 to 9 days the rate becomes very slow. For the production of RAP in the laboratory time is very important. This conclusion is related to the bitumen used in this research. It is strongly recommended to perform the ageing protocol also on softer bitumen types.

From the results with DSR and FTIR it seems that standard ageing protocols on virgin binders including RTFOT and PAV relate to approximately 2 days of ageing in the mixture ageing protocol.

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