SIMULATING AGEING OF EN 12591 70/100 BITUMEN AT LABORATORY CONDITIONING COMPARED TO POROUS ASPHALT

Jeroen Besamusca[1]*, Andre Volkers*[1]*, Jan vd Water*[2]*, Bert Gaarkeuken*[2]
[1]*Kuwait Petroleum Research & Technology*
[2]*Ballast Nedam Materiaalkunde*

ABSTRACT

Short term bitumen ageing conditioning in laboratory is most frequently performed by RTFOT at 163°C for simulating asphalt production process. PAV and sometimes RCAT are used for simulating long term ageing. The introduction of RTFOT conditioning is probably related to a publication of HVEEM, ZUBE and KOCH, AAPT from 1962.

A regularly used graph reflecting the ageing of bitumen during application is the graph with ageing index based on viscosity versus time from the SHELL bitumen handbook. It shows a large impact of the short term ageing and a less severe influence for the long term ageing. Although many publications reference to the graph, hardly any research has been performed to proof the relation with new developed asphalt constructions.

The result from this publication shows that the properties of laboratory short term conditioning on 70/100 bitumen is comparable with road asphalt production. However, the impact of long term ageing in field performance of open grade asphalt is far more severe than the ageing in laboratory conditioning according to European standards.

The most important conclusion is that the bitumen ageing is depending on asphalt components and construction (porous, dense grade, etc.).

Keywords: Ageing, Porous asphalt, Rheology, Performance testing

1. INTRODUCTION

This publication reflects the differences between two existing European conditioning tests for long term laboratory ageing, RTFOT/PAV and RCAT/RCAT. In this study the binder used originate from one single bitumen processing and the same crude oil. By changing time and temperature of the conditioning the limitation of an ageing model for asphalt based on binder conditioning in the laboratory was investigated. Several asphalt cores are used to underline this hypothesis.

The publication is written in the European definitions for road asphalt paving. This means that bitumen is the paving grade binder used for the production of asphalt roads. And asphalt is the road construction material consisting of bitumen with aggregate, filler and, possibly, additives after hot mixing in the asphalt plant and processing on the road. Softening Point is measured in according to EN 1427. Values of softening point reported here are higher than measured according to ASTM D 36 and therefore a calculated pen index value will differ.

In general, all organic material, including the human body, is influenced by the ageing process. The bitumen used in hot mix asphalt roads is organic material, produced from crude oil, and it is therefore susceptible to ageing as well.
During the processing of road asphalt the binder is spread out over the large surface of aggregates and filler and exposed to a high temperature for a limited period of time. Transport and paving of road asphalt contributes to ageing. The changes in properties of the binder during these processes are far more severe than storage in a large bitumen tank with a relatively small surface and often under inert atmosphere at the refinery.
The influence of oxygen, light, thermal expansion, the pressure of tires, etc. is revealed in the ageing of the binder “in service” on the road.

2. CONDITIONING OR SIMULATION

Simulating ageing processes in laboratory conditioning is a complex matter. For most binders one can address reaction with oxygen at 163°C as short term ageing simulating the asphalt processing in the asphalt plant. Based on research [1] the Rolling Thin Film Oven Test, RTFOT, was introduced and regarded suitable for this purpose. But since the introduction of this test in 1962 asphalt constructions and compositions have changed and also the binders used have changed. Warm mix binders are introduced and in Europe hard paving grade binders are more common. The asphalt production temperature for warm mix binders and hard paving grade binders differ almost 100ºC. However, simulation of this process in the laboratory test is defined in EN 12607-1 [2] (and ASTM D 2872 [3]) at 163ºC for 75 minutes. Moreover the results of the binder properties after ageing are used to calculate asphalt design.
Next to the short term ageing is the long term ageing. The oxidation part in long term ageing can be simulated in the laboratory with the Pressure Aging Vessel [4], PAV, or Rolling Cylinder Ageing Test [5], RCAT.
A comprehensive overview of test methods developed was written during the Strategic Highway Research Program (SHRP) in the USA. Report SHRP A-305 [6] shows the different methods used and report SHRP A-368 [7] describes the different chemistry of bitumen including some ageing aspects.
Choquet and Verhasselt [8], and Verhasselt [9] published evaluation of change in properties based on the kinetics of organic material translated to the ageing of binders. The kinetic model they published shows that ageing of bitumen below 100ºC is different compared to ageing above 100ºC. This should be taken into account when research is performed and relation to field performance is mentioned. They used thin slices of dense asphalt cores to show the difference in ageing across the asphalt cores thickness. They confirmed the work of Chipperfield [10], but important to notice is that all this research focussed on investigation of dense asphalt constructions.
The ageing of asphalt is also influenced by (e.g. Ultra Violet) light. But light has limited penetration in dense asphalt layers [11,12].
Optimal comparisons of binders in the laboratory require a standardized conditioning method. To investigate different binders a researcher prefers a short time frame for the conditioning. Conditioning for binders investigation according to SHRP specifications is the combination of RTFOT followed by 20 hours PAV. To gain time and work quicker it seems possible to [13] replace this sequence with only 25 hours PAV showing the same properties as the RTFOT/20h PAV combination. The Danish Road [14] reported in 2009 their findings and concluded that it was not possible to compare results of 25 h PAV and RTFOT/PAV combination.

To our knowledge the optimization of the laboratory simulation of short term and long term binder ageing in asphalt is still in progress. Different binders in one specific laboratory conditioning will reveal only the binder characterisation. The influence of fillers and aggregate is too important to neglect [15,16,17] in the validation of asphalt behaviour.

The common used graph from SHELL Bitumen Handbook [18], see figure 1, reveal that production of asphalt is responsible for 50% of change in properties. Our results show that this reflection is only appropriate with respect to the change in kinematic viscosity values measured at 135°C. For all other properties the change after asphalt production is approximately 25%. The difference in asphalt constructions used for the SHELL graph and this investigation (dense compared to porous, respectively) have to be taken into account. Although an “asphalt model” or a “bitumen model” is widely used it does have its limitations. Not all asphalt behave equal, not all bitumen behave equal. Therefore we investigated asphalt produced with the same bitumen origin as laboratory tests.

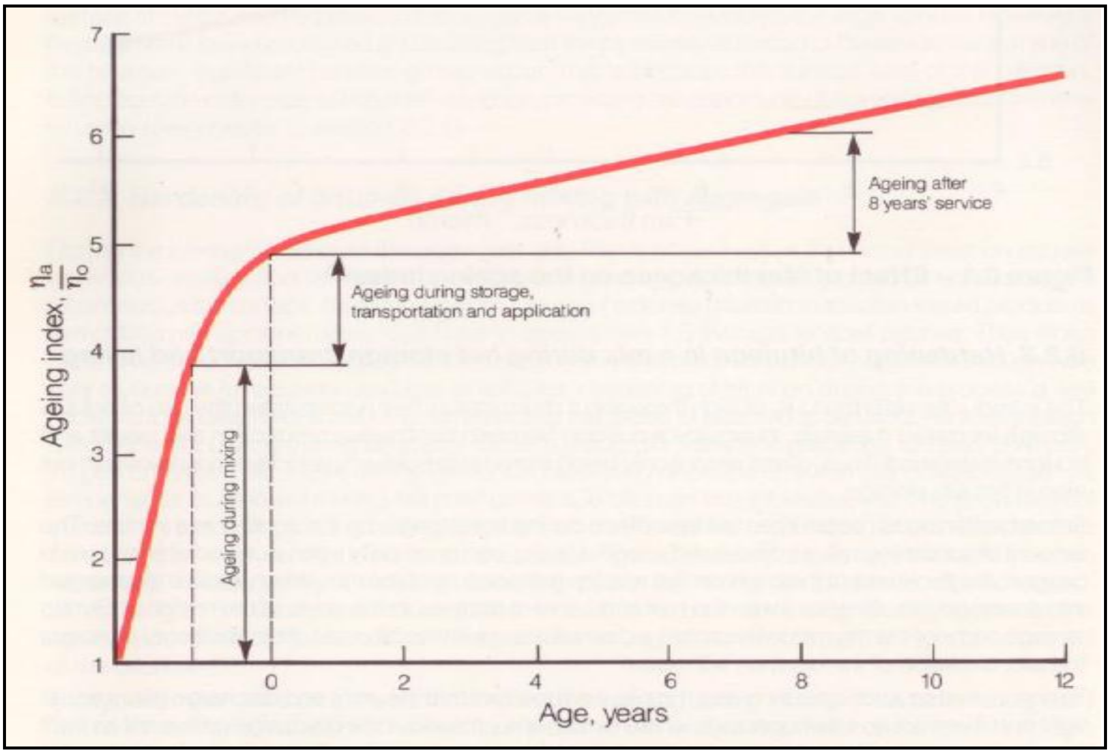

Figure 1. SHELL bitumen Handbook figure demonstrating the high influence of asphalt processing and relatively small influence of asphalt application on degradation of the binder based on the viscosity index.

3. MATERIALS AND CONDITIONING USED

The binder used is the penetration 70/100 paving grade bitumen, complying with all demands of EN 12591-2009 (European product specification for penetration grade paving bitumen), produced at the Kuwait Petroleum refinery in the Netherlands from straight run production. The crude oil is Kuwait Export Crude (KEC) and a constant in the production of bitumen from Kuwait Petroleum Europoort. This bitumen 70/100 is used in all conditioning procedures, see table 1. For optimal comparison the use of one batch for all conditioning procedures was planned. However, the interest in additional experiments, including replicates, resulted in the use of two more batches of bitumen 70/100. The differences in results are shown in the graphs, indicated with the penetration value (e.g. p88 or p82). Wigro 60K is the filler used to investigate the change in properties due to presence of filler.

Table 1. Conditioning of binder in laboratory

	Temp	**Time**	**Standard**	**Abbreviation**
	[ºC / ºC]	**[h / h]**		**(d = duplicate)**
Unaged				Unaged and _p88, p82, p78
Unaged + 5% w/w Wigro 60K				Unaged_W5
RTFOT or RCAT	163	1.25	EN 12607-1	CalcST
RTFOT / PAV	163 / 90	1.25 / 20	EN 12607-1/14769	PAV90
RCAT / RCAT	163 / 90	4 / 140	EN 15323	RCAT90 and CalcLT
RTFOT / PAV	163 / 100	1.25 / 20	EN 12607-1/14769	PAV100
RCAT / RCAT	163 / 100	4 / 140	EN 15323	RCAT100 and _d
RTFOT-EVT / PAV	151 / 100	1.25 / 20	EN 12607 /14769	E_PAV100
RCAT-EVT / RCAT	151 / 100	4 / 140	EN 15323	E_RCAT100 and _d
RCAT / 1.5xRCAT	163 / 100	4 / 210	EN 15323	RCAT100_1.5
RCAT / 2xRCAT	163 / 100	4 / 280	EN 15323	RCAT100_2

Interesting observation is the difference in laboratory practice when comparing RTFOT/PAV with RCAT/RCAT experiments. The RCAT cylinder is rather heavy to work with, but has the advantage of processing enough bitumen material in one run for all desired analyses. The entire conditioning needs 6 days to complete, but only less than 2,5 hours of analyst time. In comparison, the RTFOT/PAV combination results in three runs of RTFOT (each time 8 glasses) followed by one run of PAV using the combined RTFOT processed materials. In practice this means 2 ½ days for the entire experiment but analyst time increased to 6 hours due to conditioning and cleaning the equipment.

The refinery of Kuwait Petroleum produces bitumen from KEC crude for more than 30 years. The database of Kuwait Petroleum Research & Technology is built on all the measured properties of bitumen produced, aged and un-aged [19]. This database is used to calculate a predicted value for the properties for short term aged “Calc ST” and long term aged “Calc LT” to compare with the results from this study.
Not all properties are in the database and therefore some graphs in this paper show no results for calculated values of “Calc ST” and/or “Calc LT”.

The investigated asphalt cores are all produced with 70/100 KEC bitumen. Three different asphalt cores have been used. An asphalt sample just after production was collected at the asphalt plant for this study. From this material, open grade asphalt paving was produced and in service in front of the asphalt plant (transport lane). After 6 weeks asphalt cores were collected and the binder extracted. The last asphalt sample was taken from a highway side lane with an open grade asphalt construction of approximately 10 years old, see table 2.
All binder was extracted in accordance with the EN 12697-1.

Table 2. Asphalt samples produced with 70/100 KEC bitumen.

Asphalt sample	Abbreviation
Production sample, directly after road asphalt production	Prod
Cores taken from open grade asphalt with 70/100 KEC bitumen after 6 weeks of application	6 wks
Cores taken from open grade asphalt with 70/100 KEC bitumen after approximately 10 years of application	10 year

The following tests are performed with the binder materials.

Table 3. Test performed on binder material

Penetration	EN 1426
Softening Point (R&B)	EN 1427
Dynamic viscosity @ 60°C	EN 12596
Kinematic Viscosity @ 135°C	EN 12595
Bending Beam rheometer (@ 5 temperatures)	EN 14771
Dynamic shear rheometer (@ 10 temperatures)	EN 14770
Dynamic viscosity by DSR (@ 5 temperatures)	EN 13702
Fourier Transform Infrared spectroscopy (FTIR) 400 – 4000 cm^{-1}	

4. RESULTS

To show the influence of the conditioning with respect to the properties of the un-aged binder the percentage change is calculated for all properties.
The standard formula showing the change in property used is:

$$\%\ \text{property difference} = \frac{\text{Absolute value [aged binder – unaged binder]}}{\text{unaged binder}} \times 100\ \%$$

For the evaluation of the viscosity @60°C and @135°C the log value was used in the formula.
For BBR and DSR the standard SHRP values are interpolated from the values at different temperatures. Calculation of the DSR values at different temperatures showed the shift of the temperature with respect to the G* value equals 5 MPa for the different binders. The BBR values resulted in the temperature with m-values equal to 0.3 [-].

In all graphs, the order of the samples (x-axis) depends on their individual results in the tests. They are ordered in increased y-values. As expected, all results show the same trend. Only the sulfoxide peak measurements in the FTIR analysis revealed a different trend.
The “prod” sample is in the same order of magnitude as the measured short term ageing and calculated “calc ST”. The PAV 90 is less aged than the RCAT 90. The same applies for the PAV100 compared to the RCAT100. The sample taken after 6 weeks in asphalt application is in the same order of magnitude as RCAT100 material.

The influence of an EVT temperature (151ºC) instead of 163°C, used in the short term conditioning RTFOT and RCAT, did not result in significant changes after the long term aged properties for bitumen 70/100. The 10 years asphalt sample is significantly different from the results of the laboratory aged binders. Even with the RCAT at 100°C long term conditioning extended with double hours (280 h), the ageing is more severe for the 10 years asphalt sample.
In summary, the severity of the conditioning can be expressed as follows:

Unaged	**<**	**Calc ST** **Prod**	**<**	**PAV90**	**<**	**Calc LT** **RCAT90** **PAV100** **EVT-PAV100**	**<**	**RCAT100** **EVT-RCAT100** **6 wks**	**<**	**RCAT100-1.5**	**<**	**RCAT100-2**	**<**	**10 year**

The extension of RCAT100 conditioning up to 280 hours was monitored with samples at the regular 140 hours (RCAT100 _d) and a sample at 210 hours (RCAT_1.5). The samples were too less in quantity to perform all tests and therefore not all graphs show results for RCAT100_d and RCAT100_1.5 samples.
Despite the effort used in the handling of the asphalt cores still some inorganic material (2 to 5 % w/w) was left in the asphalt extracted bitumen. Therefore an unaged bitumen 70/100 sample was mixed with 5% w/w Wigro 60K to investigate the influence of the filler.

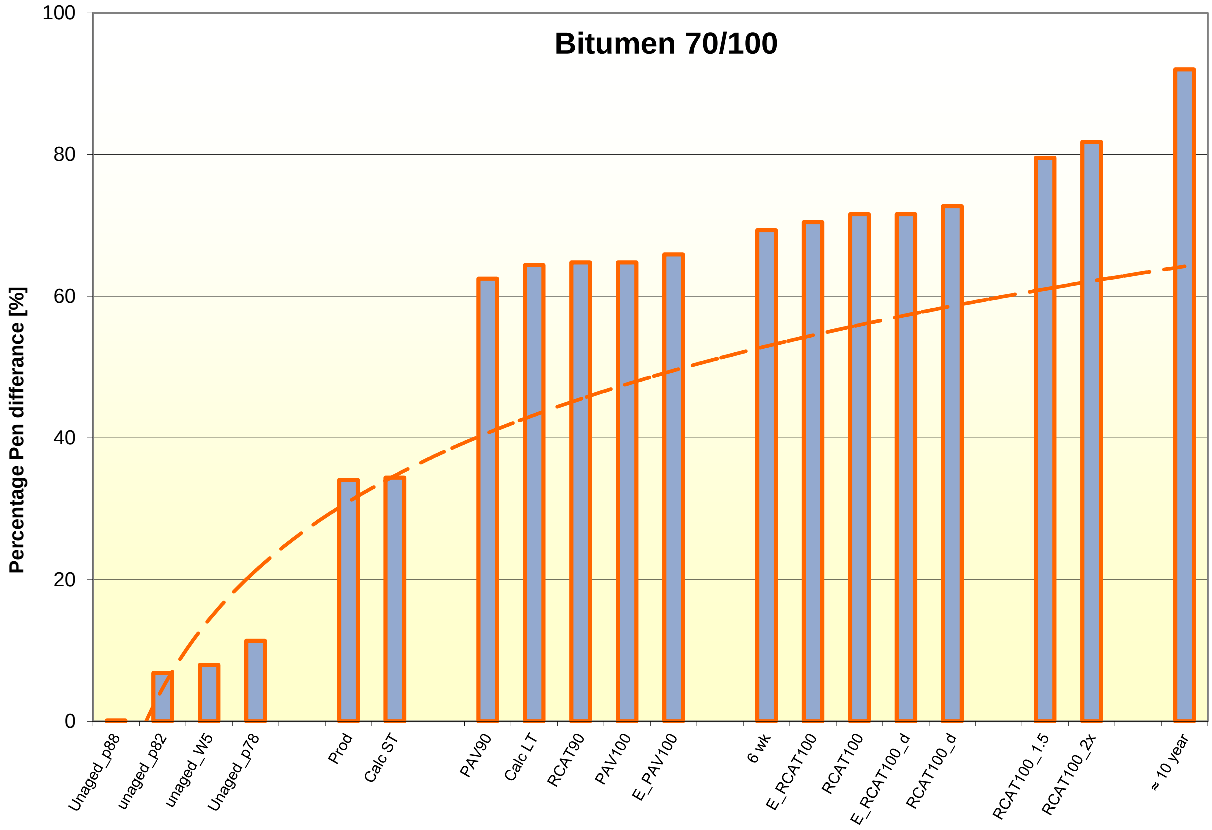

Figure 2. Percentage change in penetration after ageing compared to un-aged.

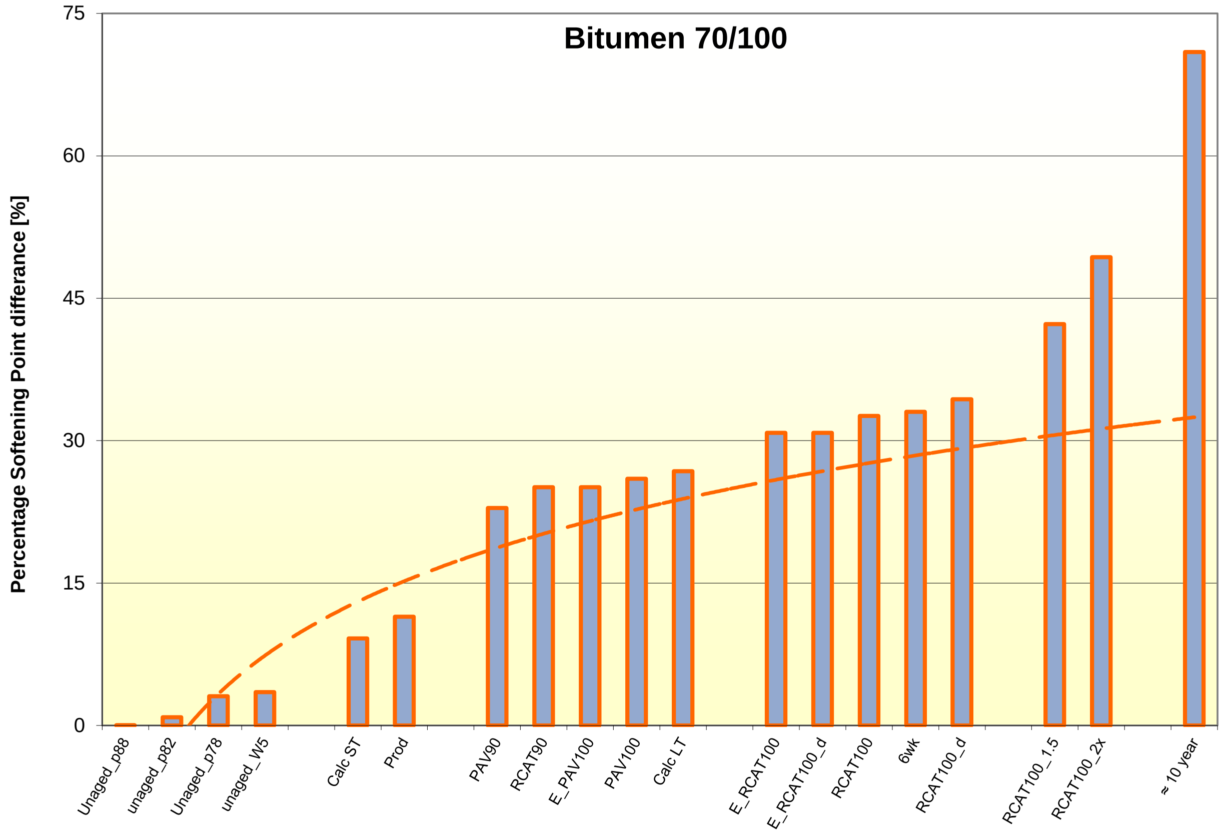

Figure 3. Percentage change in softening point after ageing compared to un-aged.

The graphs for kinematic and dynamic viscosity show the same results.

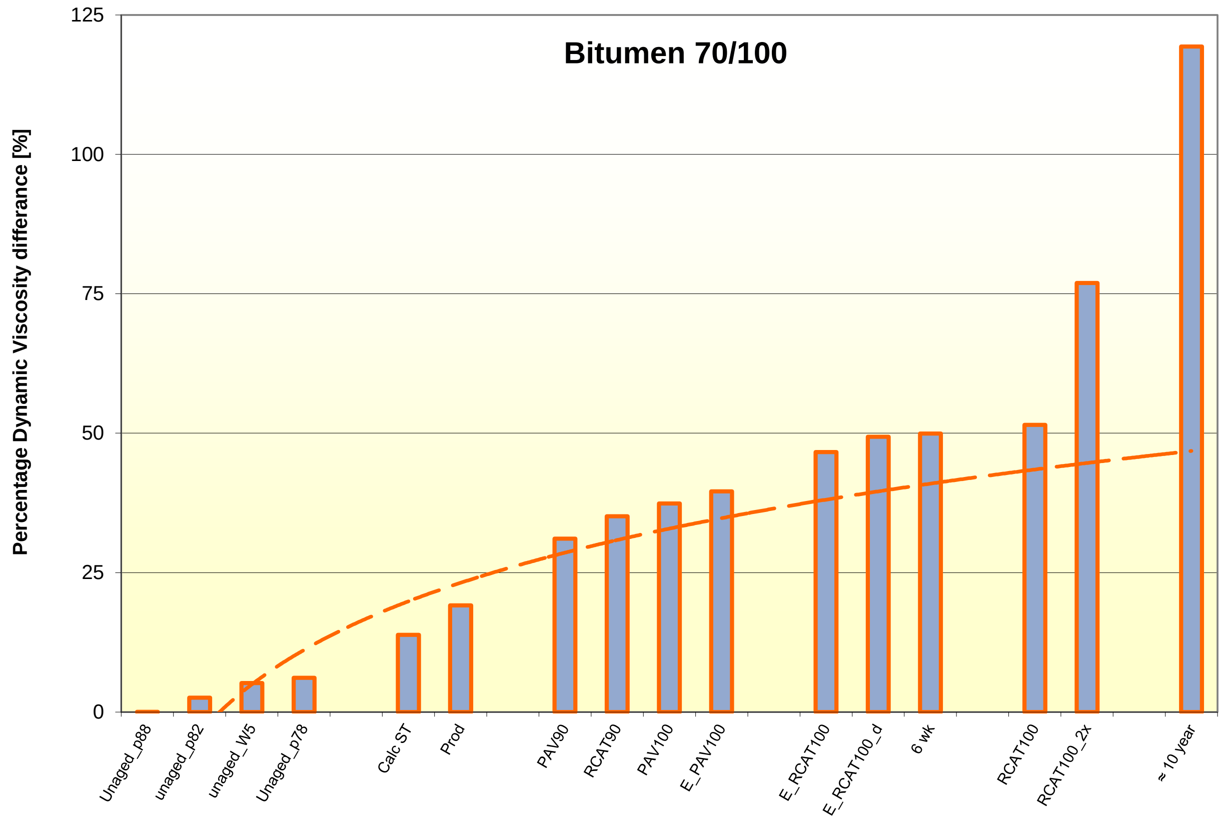

Figure 4. Percentage change in dynamic viscosity at 60°C after ageing compared to un-aged.

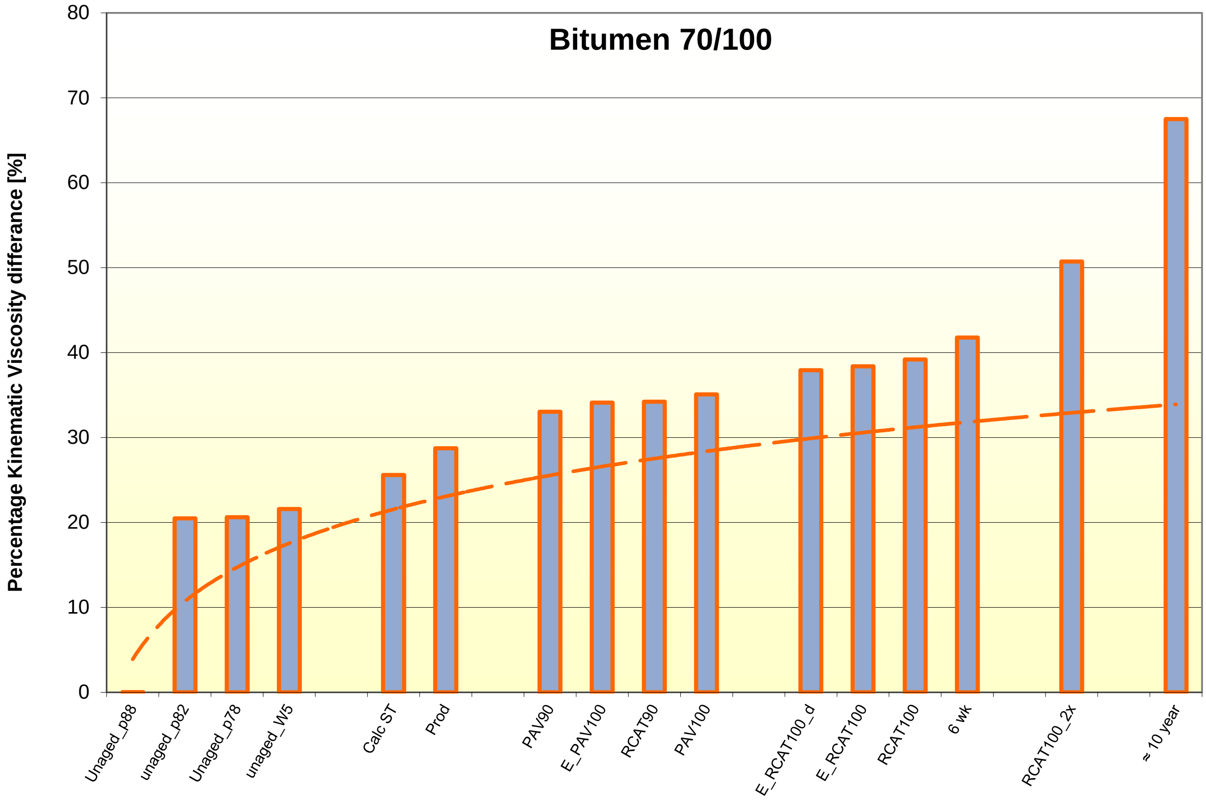

Figure 5. Percentage change in kinematic viscosity at 135°C after ageing compared to un-aged.

To compare BBR values the temperature of the binders with the m-value equal to 0.3 [-] was calculated.

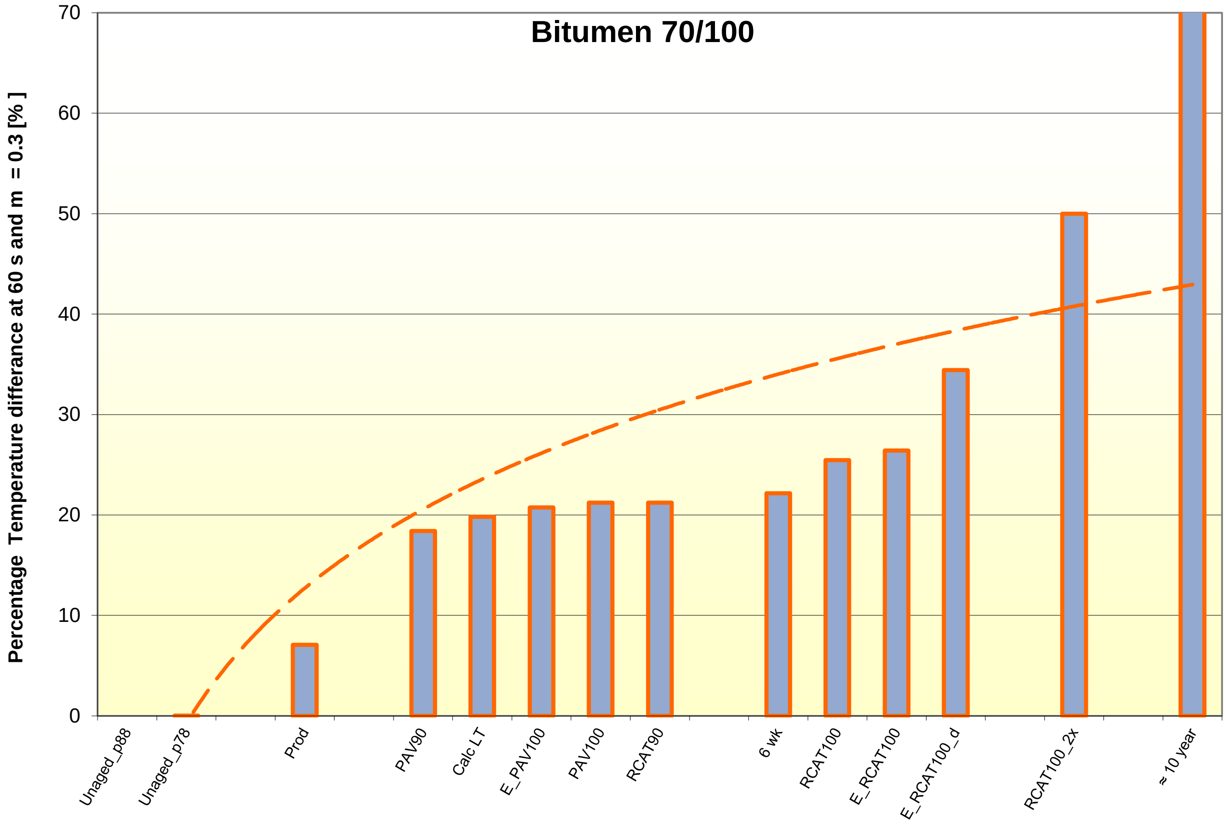

Figure 6. Percentage change in temperature at m = 0.3 after ageing compared to un-aged.

The SHRP fatigue value is calculated at the temperature with G* equal to 5 MPa. We also calculated this value for un-aged binder to compare the difference in the same formula as used for all other results.

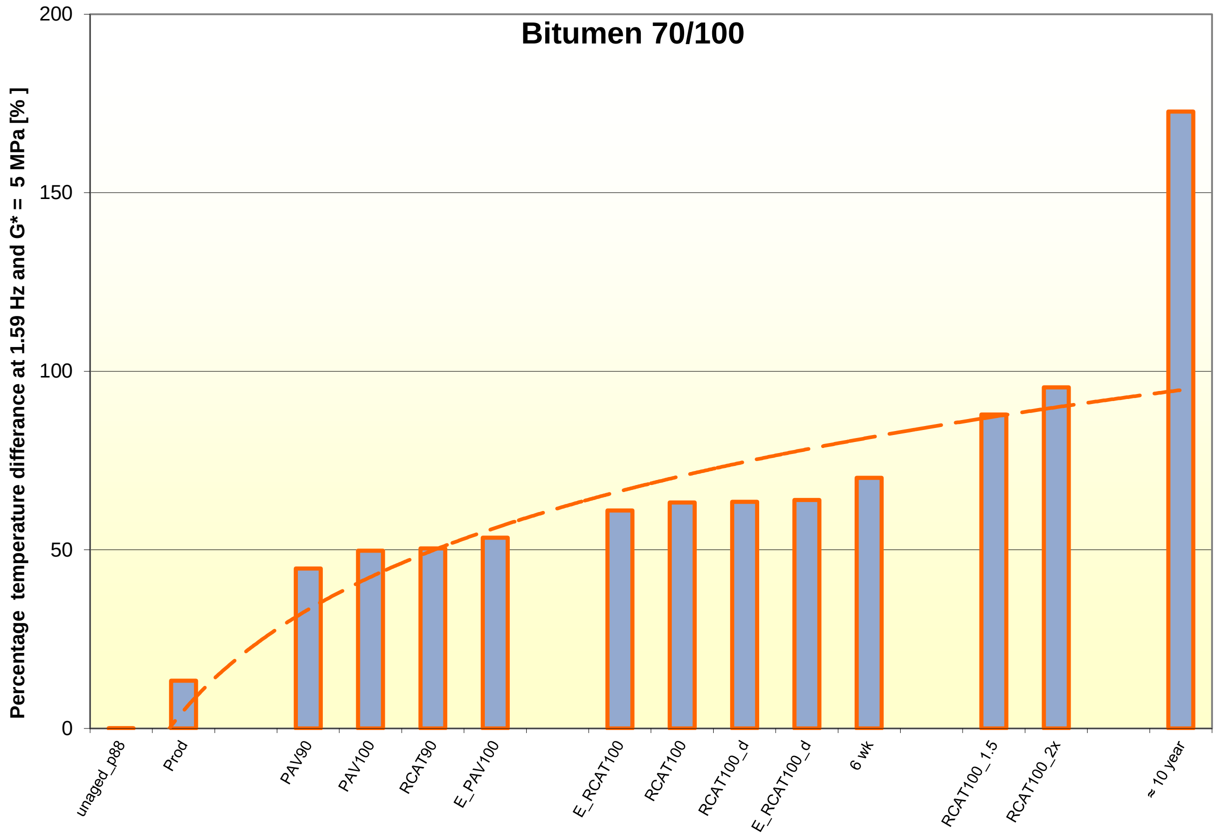

Figure 7. Percentage change in temperature at G* = 5 MPa after ageing compared to un-aged.

FTIR measurements are related to the molecules in the material investigated. The absorbance at wavelength approximately 1700 cm^{-1} is related to the C=O bond. The increase of this peak is correlated with the oxidative ageing of the binder. This was already mentioned in the SHRP reports [6,7] and by Mouillet [20] as a possible indication of the ageing aspects. In our calculations for this paper the FTIR results of the different binders are equalised on the baseline of the fingerprint area, 800 – 400 cm^{-1}.

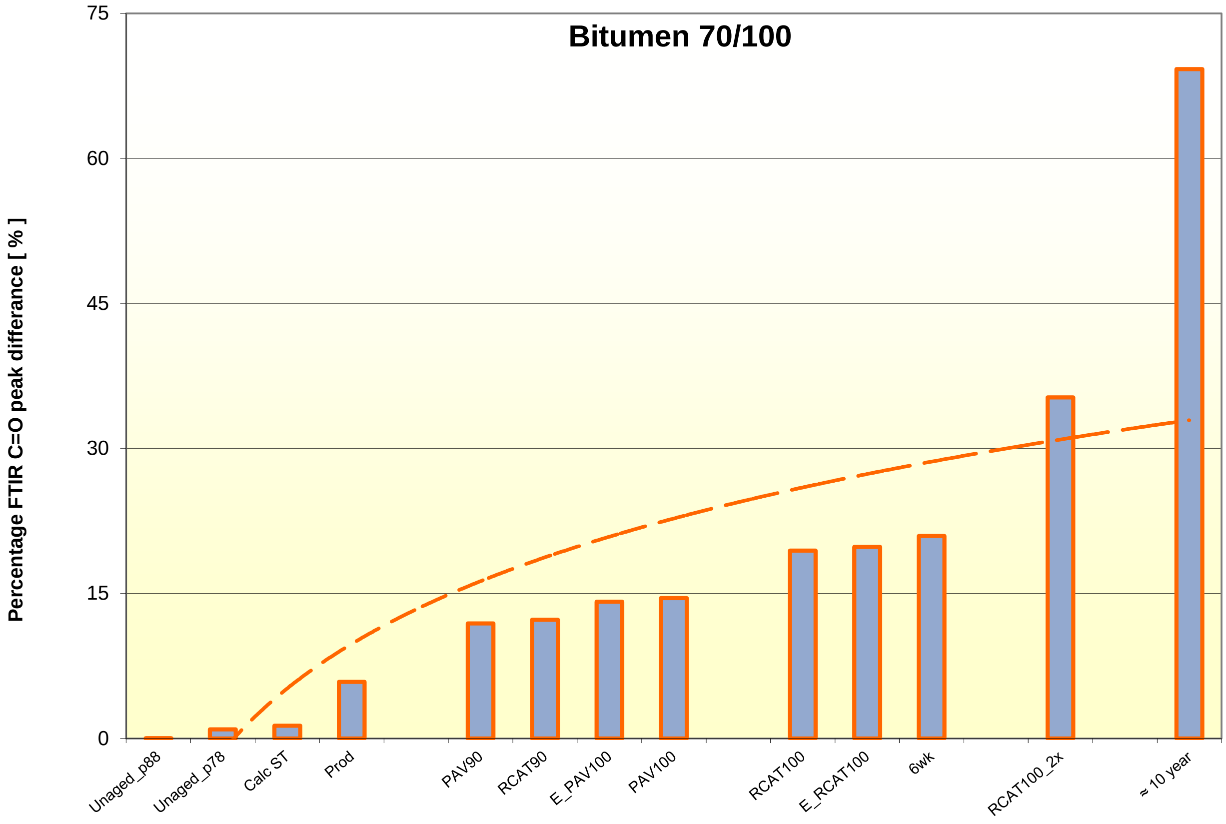

Figure 8. Percentage change in peak height at 1700cm^{-1} after ageing compared to un-aged.

There is only one exception in the common trends shown in the graphs so far. The results for the FTIR of peak measurements for 1030 cm^{-1} related to the S=O, see figure 9, show a different trend. All bitumen extracted from production of asphalt samples clearly show a difference compared to the laboratory aged bitumen.
The small amount of filler material left in the asphalt production samples do not contribute to the S=O peak. The absorbance of the regular used filler, Wigro 60K, was verified and no absorbance was relevant at the investigated wavelengths.

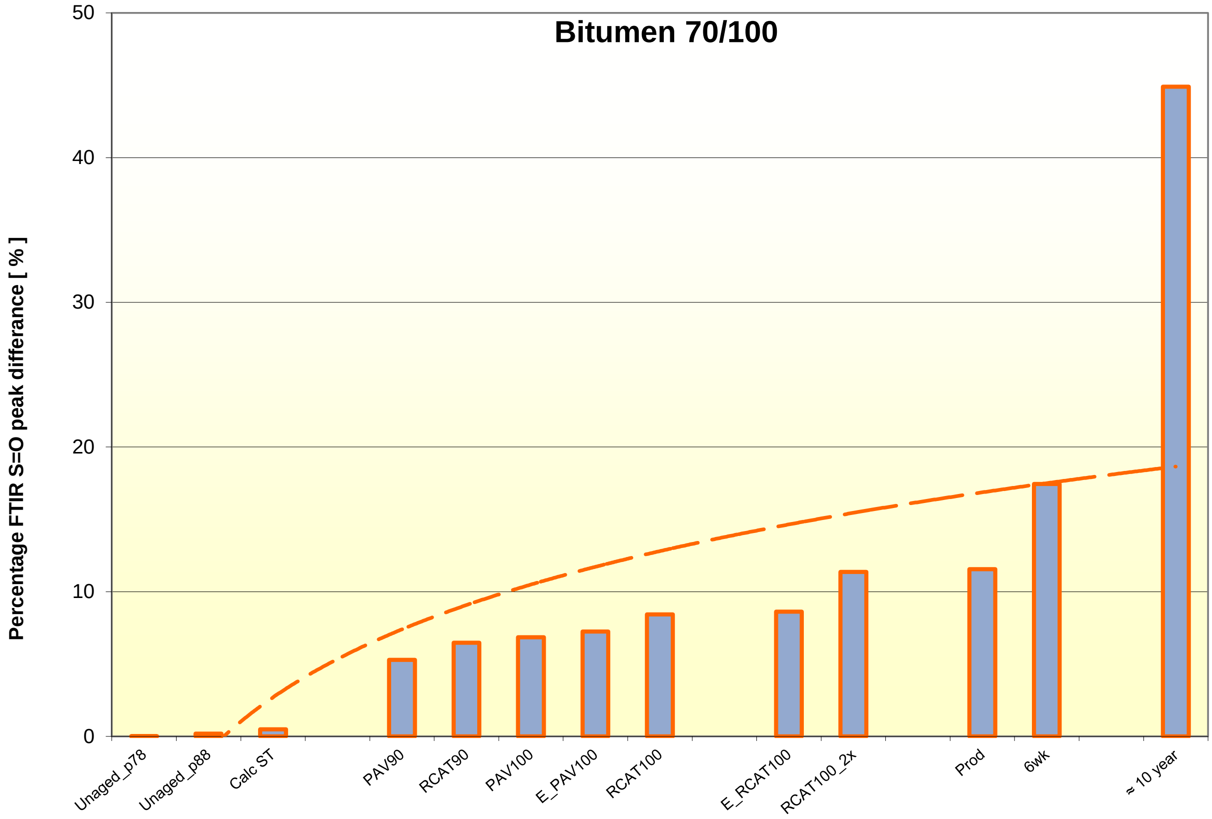

Figure 9. Percentage change in peak height at 1030cm^{-1} after ageing compared to un-aged.

Although the FTIR data of sulfoxide peak measurements could visualise the difference in ageing process, the analysed empirical and rheological properties did not show this influence in any measurements performed. It seems that the use of the sulfoxide absorption to relate ageing of asphalt is restricted to compere asphalt cores. This result confirmed the work of Zang [20].

Thermo gravimetric analysis (TGA) showed that all binders from asphalt samples contained a small amount of inorganic material, from 2% (w/w) to 5% (w/w).
Filler material can influence properties of binder. Verification of the influence of 5% (w/w) Wigro 60K in the binder, sample unaged_W5, on most empirical and rheological properties led to the conclusion that this influence was negligible. Higher levels of filler could potentially influence the properties of the binder as this effect has been reported by Cooley [22] for 55% (w/w) filler in the binder.

TGA analysis also revealed the combustion characteristics of the binder. With TGA we could show that filler was present in the asphalt extracted binders.
The combustion profile looked different between the binders from the asphalt samples compared to the neat bitumen. This subject is under further investigation if this analysis can be used to evaluate the influence of fillers and aggregates.

5. DISCUSSION AND CONCLUSIONS

The results of the characterisation of the laboratory long term aged 70/100 KEC originated bitumen led to the conclusion that the severity of RCAT is slightly more pronounced than the severity of PAV.

The short term conditioning relates well with asphalt production at the asphalt plant. An open grade asphalt constructed with 70/100 bitumen after 6 weeks in service correlates with laboratory conditioning of 4 hours RCAT 163°C followed by 140 hours RCAT 100°C. The open grade asphalt for app. 10 years in service showed much more ageing and did not correlate with any of the laboratory conditionings tested, not even with 4 hours RCAT 163°C followed by 280 hours RCAT 100°C.
The severity of the conditioning for 70/100 bitumen can be expressed in the following sequence:

Unaged < **Short Term**
Prod < **PAV90** < **RCAT90**
PAV100
EVT-PAV100 < **RCAT100**
EVT-RCAT100
6 wks < **RCAT100-1.5** < **RCAT100-2** < **10 year**

The properties of bitumen 70/100 after long term ageing are not influence by the short term conditioning temperature when comparing standard with EVT. Conditioning at 151°C instead of 163°C is hardly noticeable after the combined short term / long term conditioning in the tests performed.

Considering the sulfoxide formation data we have an indication that ageing in the asphalt plant is chemically different from ageing in the laboratory conditioning experiments. We suspect the influence of filler and fine aggregates to act like a surface extender and/or catalyst influencing the ageing process.
Although the chemical characterisation showed differentiation in sulphur oxide formation between the laboratory conditioning and the “in service” road asphalt samples, no empirical or rheological characterisation correlated with the sulphur oxide formation.

The influence of low amount of filler material (maximum 5%) in the binder samples can be neglected with respect to the empirical and rheological characterisations of the binder.

A very interesting conclusion from this study and in contrast with the SHELL bitumen handbook is the low severity of the short term ageing versus high severity of the long term ageing in open grade asphalt and in laboratory conditioning..

6. RECOMMENDATION

This study showed that currently no single laboratory conditioning on bitumen 70/100 relates with years of ageing of open grade asphalt.
This work is performed with 70/100 bitumen and porous asphalt cores. It is not recommended to extend the conclusions towards all penetration grade bitumen, other binders or other constructions.

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