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Effect of ageing kinetics on the rheological properties of bituminous binders and mixes: experimental study and multi-scale modeling

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Abstract

The oxidative ageing of hot mix asphalt (HMA) and foamed warm mix asphalt (WMA), performed through laboratory accelerated ageing processes is investigated in this study. 50/70 pen-grade bitumen was used to produce the mixes. Four ageing durations (0, 3, 6 and 9 days) were considered to investigate the kinetics of long-term ageing based on a RILEM procedure. The binders of the aged mixes were extracted and characterized. Moreover, RTFOT and PAV tests were carried out on fresh bitumen to simulate the long-term ageing of HMA. These procedures were adapted to simulate the long-term ageing of WMA for which there is not yet a standardized ageing procedure. Rheological tests were performed to determine the stiffness modulus of binders and mixes. The results show that the stiffness modulus increases with the ageing kinetic for both binders and mixes. This increase is more pronounced for WMA and their binders than HMA. Furthermore, the equivalence between the RTFOT and RTFOT+PAV ageing procedures on bitumen and the RILEM procedure on mixes was discussed for both HMA and WMA. Finally, a multi-scale model was implemented to predict stiffness moduli and phase angles of the mixes based on bitumens' and aggregates' properties. The results show a relatively good agreement between the multi-scale model and the experimental data curve-fitted with the well-known modified Huet-Sayegh model.

keywords: bituminous mixture, ageing, stiffness modulus, multi-scale modeling

1 Introduction

Bituminous mixes are viscoelastic complex composite materials build up from a mixture of aggregates, bitumen, fillers and additives. As such, their end performances entirely depend on their components percentage and properties, their interaction with each other and the evolution of their properties over time. Many parameters have a significant influence, such as aggregate size and grading, filler size distribution, bitumen properties and the interaction between bitumen and aggregates. Understanding the influence of all these parameters is a challenging task and requires a multi-scale modeling to assess the effect of each component [1],[2]. Moreover, the ageing induced by the manufacturing conditions and the field climatic modifies the chemical composition of the binder **that makes the mixes' performances to evolve**. Indeed, the ageing of mixes occurs in two main stages: The short-term ageing **is** caused by the manufacturing in the mix plant, the transport and the paving during which the ageing process is **mainly** associated to a departure of the volatile compounds of bitumen and the long-term ageing during pavement life **is** influenced by climatic conditions, and road traffic and **is** related to steric hardening, oxidation and UV rays which affects only the 12 mm upper layer of the roads.

In order to simulate **the** ageing that has taken place over several years, accelerated ageing methods were developed in laboratory. Among these methods, one can distinguish the ageing processes on bitumen RTFOT (Rolling Thin Film Oven Test) **followed by** PAV (Pressure Aging Vessel) which **is claimed** to reproduce 7 to 10 years natural ageing of an asphalt pavement [3]) and those carried out on loose mixes. The prediction of field ageing based on laboratory ageing protocols is still questionable and imprecise (from

5 to 10 years). In general, the predictions are more or less based on the analysis of one of the following indicators: carbonyl or sulfoxide indexes [4], rheological properties, mechanical performance but never all indicators together. More than twenty ageing procedures on the mixes can be found in the literature but to our knowledge there is no equivalence between these latter and the RTFOT+PAV ageing methods that can be performed on bitumens. For WMA the ageing protocols are still under investigations.

This study has three main objectives: Firstly it aims to compare HMA and WMA in term of ageing by characterizing the rheological properties of the mixes and the extracted binders taking into account several ageing kinetics. Secondly, it aims to implement a multi-scale model capable to predict the rheological properties of both HMA and WMA based on their constituents (bitumens, aggregates, air voids) properties. Incidentally, it also allows to compare the RILEM ageing protocol on loose mixes (described CEN TS 12697-52) and the RTFOT+PAV method on bitumen (described in EN 12607-1 (RTFOT) and EN 14769 (PAV)) developed for HMA and proposes an adaptation of these ageing protocols to foamed WMA.

2 Materials and mix design

2.1 Materials properties

The mixes were produced with a paving grade bitumen, whose properties of needle penetration (EN 1426) and softening temperature (EN 1427) are respectively 57×0.1 mm and 49.6 °C, and porphyry aggregates which Los Angeles (EN 1097-2) and Microdeval (EN 1097-1) properties are 17% and 8%. The mixture composition and the constituents densities are given in Table 1.

Constituent	Mass content (in %)	Density (in $kg.m^{-3}$)
Limestone filler	2.96	2700
0/2	27.59	
2/6.3	25.05	2720
6.3/10	44.4	
Bitumen	5.4	1040
Cecabase for WMA	1% (bitumen mass)	900

Table 1: Mix composition

2.2 Ageing of asphalt mixture

Ageing of hot mix asphalt (HMA): The hot mixes were manufactured respectively at $165^{\circ}C$ and then subjected to different ageing durations from 0 to 9 days. After the mixing, the loose HMA mixture was cool down to $135^{\circ}C$ and spread in thin layer (not more than 2.5 cm) into a tray and placed in a ventilated oven during 4 hours to simulate the short term ageing. Then, it was cool down to $85^{\circ}C$ and kept in the oven for 3, 6 or 9 days to reproduce the long term ageing. At the end of the ageing procedure the loose mixes were reheated at $135^{\circ}C$ during 2 hours then compacted. The reference hot mix was compacted without long-term ageing and designated H0 while those subjected to long-term ageing were labeled H3, H6, H9 corresponding to the 3, 6 or 9 days ageing.

Ageing of foamed warm mix asphalt (WMA): The foamed bitumen was produced with a mixture of hot bitumen, 1% of Cecabase additive and 5.5% of added water which allows to obtain maximum expansion. The WMA were manufactured at $135^{\circ}C$ then immediately spread in thin layer. The short and long term ageing and compaction procedures applied are similar to that of HMA. The mixes manufactured were labeled W0, W3, W6, W9 for reference, 3, 6 or 9 days ageing respectively.

2.3 Ageing of bitumen

Simulation of HMA field ageing by RTFOT and PAV protocols: The RTFOT and PAV ageing protocols were carried out on fresh bitumen to simulate respectively the short-term and long-term ageing that a binder undergoes during HMA production until paving and its natural ageing during the service life. The RTFOT was carried out at $163^{\circ}C$ during 75 min according to EN 12607-1. The long-term ageing of HMA was simulated using the pressure ageing vessel (PAV) test in accordance to the EN 14769.

Simulation of WMA field ageing by RTFOT and PAV protocols: The short term ageing of WMA was

simulated by the RTFOT performed at a temperature of 140°C other parameters remaining unchanged. [This temperature has been selected in order to be the closest to the manufacturing temperature of WMA](#)

The RTFOT and RTFOT+PAV aged bitumens and those recovered from the mixes (H0 to H9, W0 to W9) were tested with dynamic shear rheometer (DSR).

3 Rheological tests

3.1 Mixes rheological tests

The complex modulus E^* of the mixes was carried out on a prismatic cantilever beam in accordance to EN 12697-26 (Annex A: 2P-PR) varying the temperature from -10°C to 50°C and the frequency from 3 Hz to 40 Hz. The master curves of $|E^*|$ and the phase angle δ were built by shifting horizontally the isotherms as described in reference [5], [6]. Figs. 1 show that the stiffness modulus increases logically with the long term ageing duration for both HMA and WMA. This increase in the modulus induces a decrease in the phase angle as shown in Figs. 2. Moreover, the experimental data of complex modulus have been curve-fitted with the [modified Huet-Sayegh model](#) model described in [7], [8], [9] at a reference temperature of 10°C. The parameters are given in Table 2 then used to validate the multiscale model presented in section 4.

Mixes	E_0 (MPa)	E_∞ (MPa)	δ	τ	k	h	β
W_0	88.7	2.21e+04	2.78	1.45	0.231	0.771	1000
W_3	44.9	2.29e+04	2.69	5.27	0.173	0.573	1000
W_6	2.10	2.72e+04	2.51	24.5	0.148	0.478	1000
W_9	23.8	2.51e+04	2.45	704	0.159	0.515	1000
H_0	1.53	4.85e+04	2.34	2.34e-02	0.1	0.446	7902
H_3	484	5.00e+04	3.64	9.75e-02	0.107	0.480	404
H_6	0.03	3.34e+04	2.30	4.65e-01	0.127	0.425	200
H_9	35.5	4.24e+04	3.12	3.07e-01	0.108	0.457	1000
Bitumen	G_0	G_∞	δ	τ	k	h	β
B_0	1.e-05	3.00e+03	10.9	5.94e-05	0.238	0.660	124
$B(RTFOT163)$	1.e-05	3.63e+02	2.50	4.67e-03	0.350	0.664	64.4
$B(RTFOT163 + PAV)$	1.e-05	4.32e+02	4.82	1.38e-02	0.331	0.646	160
$B(H_0)$	1.e-05	2.40e+03	10.3	2.42e-04	0.268	0.656	173
$B(H_3)$	1.e-05	5.47e+02	3.82	2.37e-03	0.296	0.643	114
$B(H_9)$	1.e-05	1.45e+03	9.92	1.06e-03	0.279	0.653	211
$B(RTFOT140)$	1.e-05	3.00e+03	10.3	6.37e-05	0.246	0.645	237
$B(RTFOT140 + PAV)$	1.e-05	1.02e+03	8.12	1.47e-03	0.263	0.626	346
$B(W_0)$	1.e-05	3.00e+03	17.4	4.08e-04	0.230	0.623	269
$B(W_3)$	1.e-05	4.50e+02	7.20	3.31e-02	0.278	0.627	193
$B(W_6)$	1.e-05	2.61e+03	23.7	2.61e-03	0.237	0.593	1680

Table 2: [Modified H-S](#) model parameters for binders and mixes curve-fitted at $T_{ref} = 10^\circ\text{C}$

3.2 Properties of bitumen

Dynamic shear rheometer (DSR) test was performed to characterize the linear viscoelastic properties (*l.v.e.*) of the asphalt binders. It was performed over a temperature range from 10°C to 80°C and over the frequency sweep from 0.681 to 21.54 Hz. Applying the same approach as before for mixes, the master curves of the [norm of the shear modulus](#) $|G^*|$ and phase angle δ were built and represented in Figs. 2. Figs. 2 show that the shear modulus increases with the long term ageing duration for all binders recovered from HMA and WMA. This increase in the modulus induces a decrease in the phase angle as shown in Figs. 2. In addition, one can observe [the temperature imposed in RTFOT test at 163 °C](#) test simulates quite well the short term ageing of the HMA (H_0) production [as already shown by literature \[10\]](#). However, the long term ageing simulated by the RTFOT at 163 °C + PAV is more severe [than](#) the RILEM long term ageing procedure at 9 days on loose mixes. Regarding the WMA, the RILEM ageing procedure on loose mixes is more severe than the RTFOT at 140°C and RTFOT at 140°C+PAV. The $|G^*|$ of the bitumen extracted from short term aged mixture W_0 is similar to that of the RTFOT at 140°C+PAV which is supposed to simulate the long term ageing. In addition, it should be noted that the complex modulus of the bitumen recovered

from W9 was not tested because this latter was so hard. In addition, these data were also curve-fitted with the modified Huet-Sayegh model and the obtained parameters are given in Table 2.

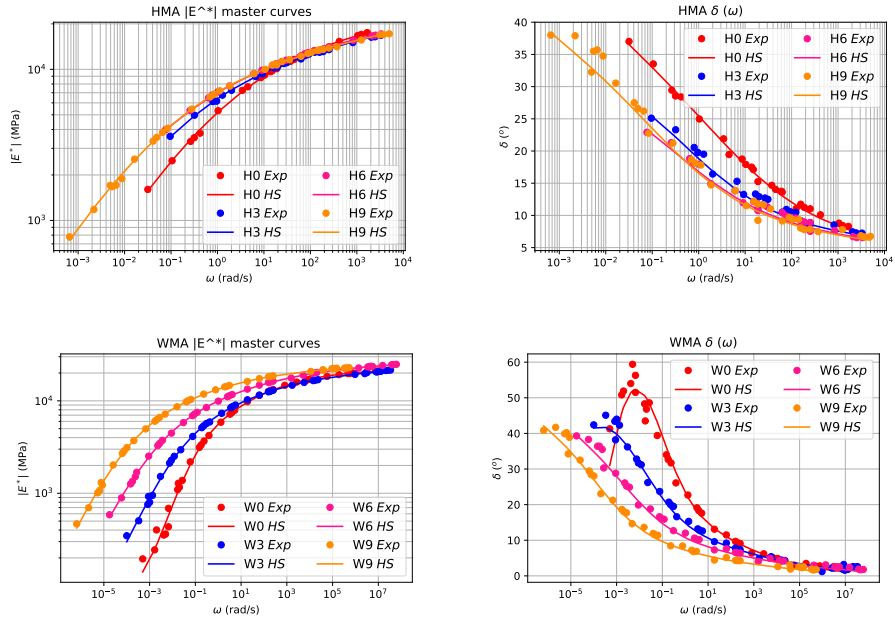


Figure 1: Master curves of complex phase angle of mixes at $T_{ref} = 10^\circ\text{C}$

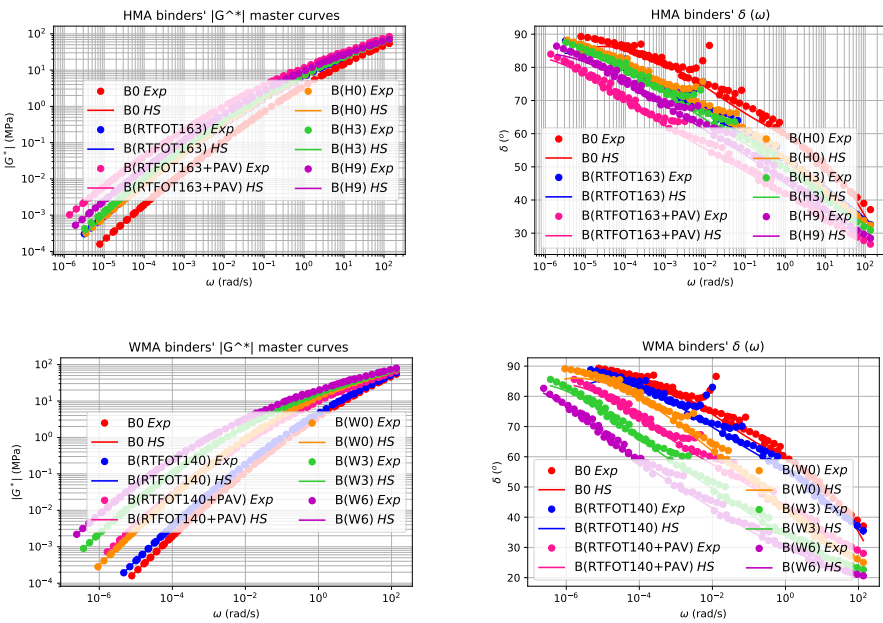


Figure 2: Master curves of binders at $T_{ref} = 10^\circ\text{C}$

4 Multiscale modeling

The AC is a multiscale material that we propose to decompose into three scales (see Fig.3):

- the macroscale is made of coarse aggregates coated with bitumen, a mortar phase partially filling the intergranular space and air voids (voids content estimated to be $5.6\% \pm 0.8\%$ for the tested mixes),
- at the mesoscale, the mortar is composed of a mastic matrix embedding sand grains,
- at the microscale, the mastic is composed of a bitumen matrix embedding fillers.

The aggregates are assumed spherical coated with soft bitumen layer whose thickness e is proportional to the aggregate diameter d according to the relation inspired from Duriez' relation $e = e_0 \times (d/d_0)^{0.8}$, where $d_0 = 1$ mm, e_0 is a setting parameter which varies between 20 and 28 μm according to Duriez [11]. At the mastic and mortar scales, the Mori-Tanaka (matrix / inclusions) scheme was used while the generalized self consistent scheme was applied at the mixture scale as in ref. [1]. At low frequency and high temperature, the bitumen behaves like a Newtonian fluid therefore, the model based on coated aggregate by thin bitumen layer fails to predict the complex modulus of the mix as previously mentioned [12]. Moreover, stiffness is needed to account the contact between aggregates when the binder becomes liquid. Thus, according to an idea proposed in [12], the contact is modeled by a juxtaposition of bitumen thin layer (of surface fraction χ) and actual grain contacts (of surface fraction $(1 - \chi)$). The grain contact does not allow interpenetration of grains but only relative sliding so that it is characterized by a linear spring model relating the local shear stress to the tangential displacement jumps thanks to a shear stiffness k_t . For $\chi = 1$, the grains are entirely surrounded by a thin layer of bitumen, which implies a total loss of stiffness at high temperature or low frequency. On the contrary if $\chi < 1$ such a model is consistent with the idea of connexity of the granular skeleton even at high temperature or low frequency. The aggregates linear isotropic elastic properties are $E = 62$ GPa, $\nu = 0.2$. The bitumen bulk modulus K was assumed constant while the shear modulus $\mu^* = \frac{3KE^*}{9K - E^*}$ depends on ω . The parameters k_t , χ and α have been determined by minimizing the objective function between the multiscale prediction and the data fitted with the modified H-S model whose parameters are given in Table 2. Figs. 4 a comparison between the multiscale and the modified H-S models. The results show a relatively good agreement for k_t between 10 and 410 MPa.mm^{-1} from softest to stiffest mixes, $\chi \simeq 0.1$ and $0.1 \leq e_0 \leq 0.4 \mu\text{m}$. For readability purposes, only a few results have been represented.

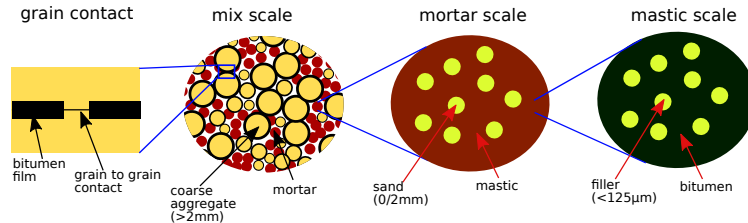


Figure 3: Multiscale model for asphalt mix

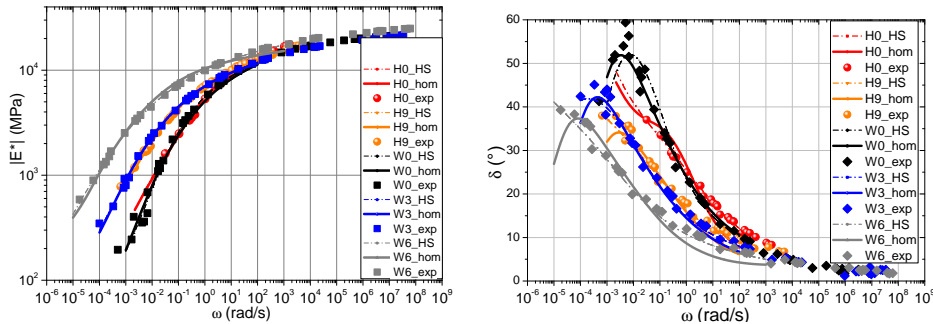


Figure 4: Comparison between multi-scale model (*hom*), modified H-S model and experiments (*exp*)

5 Conclusion

The effect of HMA and WMA ageing kinetic on the *l.v.e.* properties of binders and mixes has been investigated. The results show that the mixes stiffness increases highly with the ageing duration specially for WMA. Moreover, RTFOT and PAV ageing procedure have been also performed on fresh bitumen to simulate HMA short term ageing then their *l.v.e.* characterized. Furthermore, RTFOT and PAV ageing procedure were adapted to reproduce ageing of WMA. The results showed an increase of the binders' stiffness with the ageing duration, consistent with the mixes properties. In addition, a multiscale model has been implemented to predict the *l.v.e.* of the mixes based on those of the constituents. The model predict relatively well the *l.v.e.* properties of the mixes, by means of good calibration of the contact parameters k_t , χ and α . These latter can be well calibrated by performing laboratory experiments in a wide range of temperatures. Finally, in this study the bitumen Poisson's ratio ν is assumed following the modified HS model and bounded at 0.4 and ≈ 0.5 . As a perspective, an experimental measurement of ν can allow improving the multiscale model.

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