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Publication date
2014

Document Version
Final published version

Published in
Proceedings of the International Conference on Ageing of Materials and Structures

Accelerated ageing protocols for (polymer modified) PA to obtain representative (rheological) properties, mimicking field aged materials

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Abstract: In this work ageing protocols for asphalt and mastics were developed and investigated; i.e representation of the field ageing and their acceleration degree. Here it was aimed to age laboratory prepared specimen analogous to naturally aged materials in the field and compare their material properties. Such simulation of ageing is of importance in order to potentially specify and objectively assess the properties of (novel) binders and asphaltic materials for Porous Aaphalt (PA). Hence, two ageing protocols were developed and investigated; one ageing protocol for asphalt mixtures and one for asphalt mastics. The rheological (DSR) properties of binders from the laboratory aged materials have been evaluated in comparison to binders from field aged specimen.

Keywords: accelerated ageing, porous asphalt, binders, mastics, roads

1 Introduction

The majority of the Dutch national road network is paved with porous asphalt (PA) wearing courses. The performance of PA surface layers is superior with respect to noise reduction and reduction of splash and spray during rainfall, compared to dense surface layers. However, the main disadvantage of PA is the limited durability compared to dense asphalt mixtures. PA is more sensitive to damage because of the open structure compared to dense layers. In general it is assumed that the durability of open asphalt depends on the ageing of the binder and that this process of ageing proceeds faster in these open graded mixtures. [1]The study on ageing of bituminous binders is a widely addressed topic and is particularly interesting for predicting the service life properties of PA. [2 – 5] Research specifically focused on PA was performed on recovered binders and mastics from road sections by using DSR experiments and computer modelling in order to characterize the change of material properties of the binder (specifically the dynamic stiffness and the material phase angle) in time and parallel predict the change in material response as the material ages [6]. Such investigations have been performed aimed at defining a ravelling criterion based on functional (rheological) properties of binders or asphalt mastics. In this work ageing protocols for binders and mastics were developed and investigated; i.e representation of the field ageing and their acceleration degree. In this investigation it was aimed specifically to age laboratory prepared specimen analogue to naturally aged materials in the field and mimic the (dynamic) mechanical properties of the materials. On the one hand short term ageing occurs during production, transport and the laying process due to high asphalt temperatures and on the other hand long term ageing occurs during service life due to climatic

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loading. Ageing is known to influence the mechanical properties of the binders, but these changes may be attributed to both physical and chemical changes in the binder. Hence only a complete picture of binder ageing can be obtained when not only mechanical but also physical/chemical properties are investigated in detail. Results of this still ongoing work [7] are presented here in brief. Such simulation of analogous ageing is of importance in order to potentially specify and objectively assess the properties of (novel) binders and asphalt materials for PA. In order to achieve such simulated-aged materials, in this work two ageing protocols were investigated; one ageing protocol for non-compacted asphalt mixtures and one ageing protocol for asphalt mastics.

2 Experimental design

2.1 Materials

All materials were used as received from suppliers and manufacturers. The raw materials were continuously stored under climate controlled conditions (5°C) until they were used for sample preparation. All materials represent raw materials available on the Dutch market for construction of PA pavements and the chosen compositions generally represent a typical top-layer PA for Two-layer PA structures.. Two type of binders were used: Binder 70/100; Standard paving grade bitumen (penetration ranging from 70 to 100 [0.1 mm]) and Binder 'A': Polymer (SBS) modified binder: typical polymer modified binder used in PA wearing courses. As filler Wigro 60k was employed,: a limestone filler containing at least 25% $Ca(OH)_2$. Grauwacke 4/8 mm was chosen as coarse aggregate to produce the asphalt mixes and crusher sand originating from Bestone aggregate was used as sand fraction.

2.2 Methods

Asphalt Mixture preparation

A PA 8 mixture was prepared, with a mass ratio of binder : filler : sand : coars aggregate equal to 5.7 : 5.7 : 6.6 : 82.1. From this composition 9 kg of asphalt mixture was prepared using a Hobart laboratory mixer. The two type of binders mentioned above were used to make two different asphalt mixtures.

For the Binder 70/100, mixing process the dry aggregates, sands and virgin binder were mixed together for 90 s at 155°C. Subsequently the filler was added and the mixture was mixed for an additional 60s. For Binder A (the polymer modified bitumen), the same mixing process is followed as above, but the mixing temperature was 180°C.

Asphalt Mastic preparation

Two types of asphalt mastic samples were prepared using the two binders and one type of filler. The ratio of bitumen to filler is 1:1 by mass.

The rotating cylinder ageing test (RCAT) apparatus was used to prepare both the mastics at 170°C . For the two type of binders, the same procedure is used and the employed mixing time was 30 min.

2.3 Ageing protocols

In this work two different ageing approaches are proposed and their effect is investigated:

1. Oven ageing of asphalt mixture; under atmospheric air conditions.
2. RCAT ageing of asphalt mastic; under an O_2 (g) atmosphere condition.

By investigating asphalt mixes and mastics the film thickness on the aggregates and fines is more comparable as would be observed in the field. Hence, it is hypothesized that accelerated ageing of binders while being processed in an asphalt system will age in better comparison to the field.

2.3.1 Oven ageing on asphalt mixtures (1)

After mixing, 9 kg of hot loose asphalt mixture is poured from mixing container into a box with a size of 43 x 26 x 8 cm^3. The height of the uncompacted mixture is about 4-6 cm.

Short-term ageing (STA) procedure and sampling

For the short-term ageing process, the mixture (9 kg) is placed in an air-draft ventilated oven for 4 hours at 135°C. Each hour, the material is stirred with a metal spoon for 1 minute and placed back into the oven. This stirring action is employed for homogenization reasons. After this short-term ageing, the mixture is homogenized again by stirring for 1 minute and an aliquot of 1.5 kg is sampled from the mixture.

Long-term ageing (LTA) procedure and sampling

After the short term ageing stage the remaining 7.5 kg of the loose material is mixed and placed in the box located in an air-ventilated oven at 85°C. After 1, 2, 6 and 9 days of long-term ageing 1.5 kg mixture is sampled. Since one side of the box can be moved, the dimensions of the remaining mixture can be controlled to ensure that after sampling the smaller amount of material retains the original thickness of 4-6 cm. The parameters of STA and LTA are summarized in Table 1.

2.3.2 RCAT ageing on asphalt mastics (2)

After the preparation of the mastic, the STA and subsequently the LTA processes are directly carried out in the RCAT apparatus. Parameters for RCAT ageing are also given in Table 1.

Table 1 Ageing parameters for oven ageing (asphalt mixture)

	Asphalt mixture		Asphalt mastic	
Ageing procedure	STA	LTA	STA	LTA
Quantity [kg]	9	-	1	-
Duration	240 min	9 days	235 min	9 days
Gas	Air	Air	Air	Oxygen
Gas flow [l/min]	unknown	unknown	4.0	0.075
Temperature [°C]	135	85	163	90
Pressure [MPa]	0.1	0.1	0.1	0.1
Approx.mixture thickness [mm]	40 – 60	40 – 60	2.5	2.5
Sampling	@ end	@ 1, 2, 6, 9 days	@ end	@ 1, 2, 6, 9 days

2.4 Extraction process

All binders were recovered according to RAW standard 2010 test 110, which refers to NEN-3917. [8] This method uses dichloromethane (CH_2Cl_2) as a solvent to extract the binder from the mixtures and mastics. The binder is then recovered after controlled evaporation of the solvent. All extracted and dried binders were stored under climate controlled conditions (5°C) for a maximum of 10 days prior to further testing. The filler content of the recovered binder was determined by thermo gravimetric analysis of a small quantity (typical weight of 1.5 - 2.0 g) of the recovered binder at 1000°C in air.

2.5 Sample codes

All collected samples are coded and given in Table 2.

Table 2: Codes of each sample

	Asphalt mixture		Asphalt mastic	
Ageing period	Binder 70/100	Binder A	Binder 70/100	Binder A
0 (virgin)	MQ0	MS0	MaQ0	MaS0
1 (STA)	MQ1	MS1	MaQ1	MaS1
2 (ageing 1 day)	MQ2	MS2	MaQ2	MaS2
3 (ageing 2 day)	MQ3	MS3	MaQ3	MaS3
4 (ageing 6 day)	MQ4	MS4	MaQ4	MaS4
5 (ageing 9 day)	MQ5	MS5	MaQ5	MaS5

2.6 Dynamic Shear Rheometry

The dynamic shear rheometer (DSR) tests were performed according to EU specification, EN 14770. A DSR TA instruments AR 2000ex. Prior to the frequency sweep measurements, strain sweeps were performed to determine the linear visco-elastic regions of the binder. Frequency sweeps were performed between 0.1 and 400 rad/s between temperatures of -10°C and 60°C.

3 Results and discussion

All binder samples were extracted and recovered from the mixture or the mastic prior to testing. Despite the standardized extraction procedures and thorough removal of inorganic fines, still some fraction of filler < 63 μm will remain in the recovered binder even after a centrifuging step. In general these filler amounts are relatively low. In our experiments on average we have reported a filler weight ratio varying between 2 and 10% per sample type. Although some filler could remain in the binder after the extraction and ageing could occur during the preparation of mixture and mastic, based on performing control measurements on the fresh binder, these two factors do not influence the rheological properties of binder.

3.1 Effect of accelerated ageing

Two ageing protocols by using an air ventilated oven and the RCAT under O_2(g) were respectively performed on asphalt mixture and mastic. In this section, it is discussed how these two methods affect the rheological properties of binders. Figure 2 shows the master curves of binder samples (MQ0-MQ5 and MS0-MS5), which are extracted and recovered from the asphalt mixture after ageing. It can be observed from the figure that the master curves are gradually changed as the ageing steps proceed, i.e. the complex modulus increases with every ageing stage and the phase angle decreases from MQ0 to MQ5 within a wide frequency range.

As shown in Figure 3, the same change is observed in the complex modulus of the master curves for the binders MS0 to MS5. However, the effect of the presence of SBS polymer makes the change of the phase angle more complex (see Figure 3b). At the frequency above 0.1 rad/s, the phase angle decreases when the ageing degree increases; oppositely, it increases at a frequency lower than 0.001 rad/s. In short, the ageing effect diminishes the characteristic of modified bitumen and the properties appear more and more governed by the properties of the ageing binder.

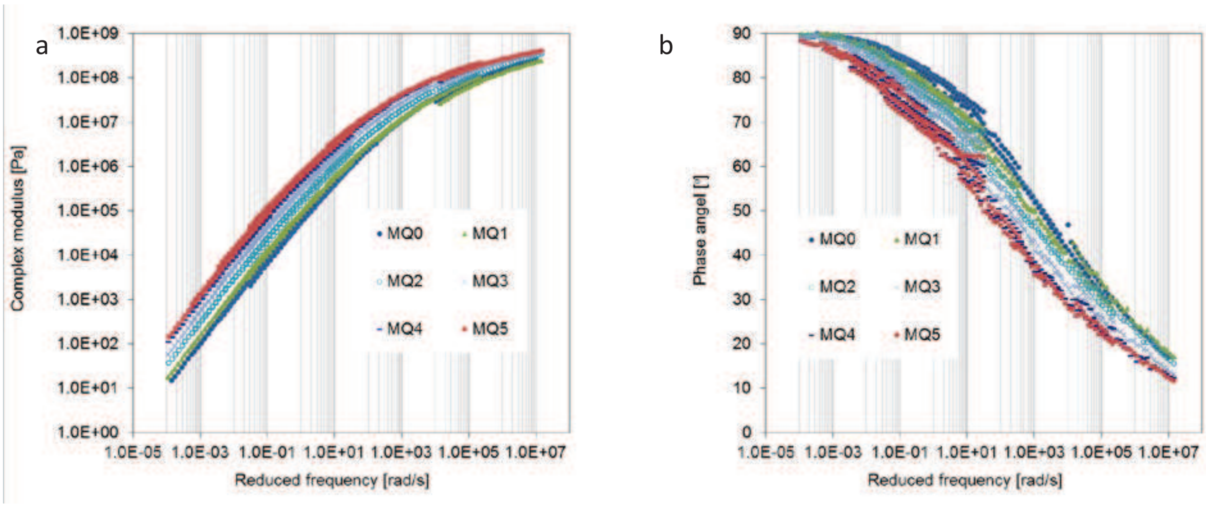

Figure 2: Master curves of (a) complex modulus and (b) phase angle of binder samples MQ0~MQ5

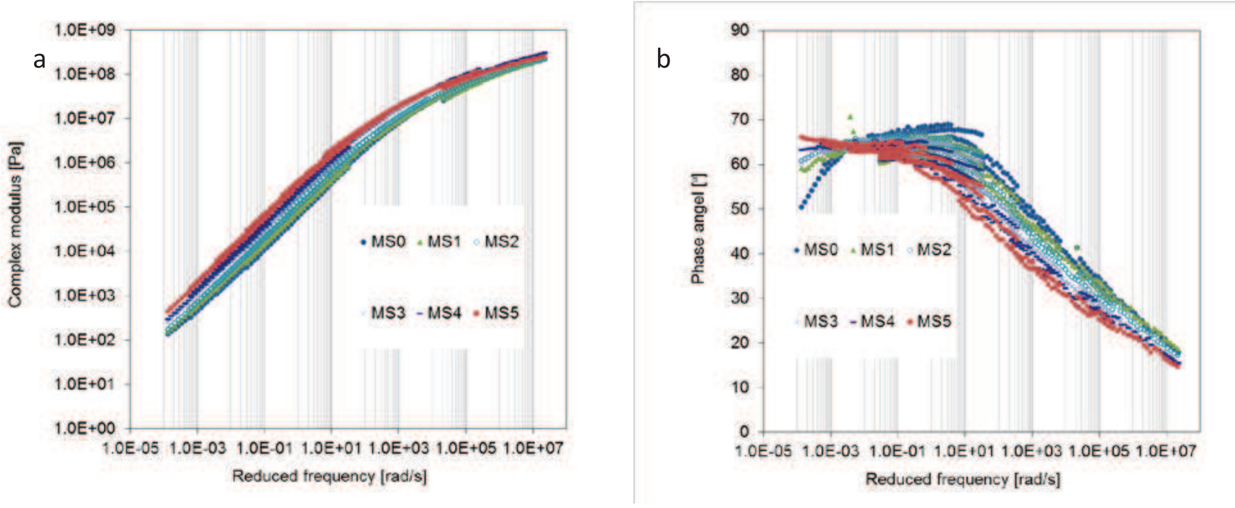

Figure 3: Master curves of (a) complex modulus and (b) phase angle of binder samples MS0~MS5

It is also necessary to note that there is a bigger difference in the complex modulus and phase angles between MQ0 and MQ5 than there can be found between MS0 and MS5. It indicates that the oven ageing acceleration based on the rheological properties is stronger for of paving grade bitumen than for the modified binder. This observation is interesting, but can be explained by the presence of the polymer, which at an equal volume of binder, represents a significant part of the binder that is less sensitive to ageing.

For ageing of the asphalt mastics, with use of the RCAT method, we do not observe serious ageing effect on the binders from MaQ0 to MaQ5 according to their master curves (see Figure 4). With respect to binders MaS0~MaS5 (see Figure 5), the master curves have a similar change trend as for binders MS0~ MS5, but similarly do not manifest a serious ageing effect. A possible reason for this is that the viscous mastic in the cylinder of RCAT hardly flows as the cylinder rotates. Under these conditions only limited oxidation may occur on the surface of the binder during the ageing process. Therefore, based on these observations the RCAT might be a less successful method to age the binder.

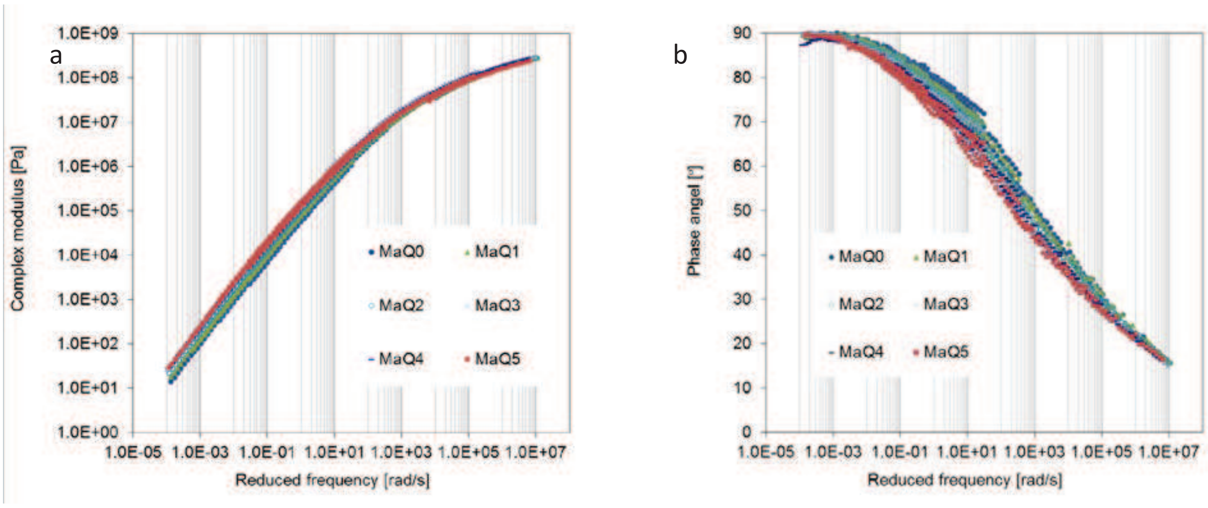

Figure 4: Master curves of (a) complex modulus and (b) phase angle of binder samples MaQ0~MaQ5

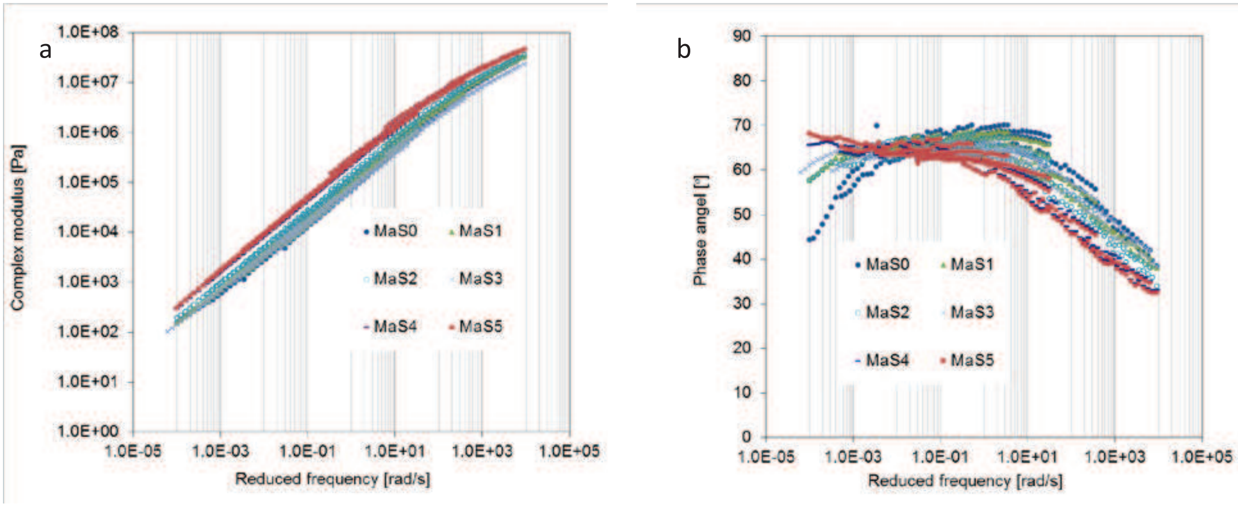

Figure 5: Master curves of (a) complex modulus and (b) phase angle of binder samples MS0~MS5

3.2 Comparison to field ageing

Next, the rheological properties of the laboratory aged materials are compared to existing data from field aged samples with binder 70/100 and polymer modified binder; binder A extracted from the top-layer PA. The comparison allows the evaluation of the field representation of the chosen ageing protocol, as well as the degree of acceleration that was achieved. The way the differently aged materials are compared, is based on evaluating the rheological properties of each material at a given frequency of 10 rad/s and at a reference temperature of 20°C. The values of both G* (complex modulus) and δ (phase angle) at these conditions are then plotted as function of the ageing stage (MX0,..., MX5). These plots are presented as so called Black-Space-diagrams. Similar plots are also presented to evaluate laboratory aged materials in [2]. The selected rheological properties for both the field aged and accelerated laboratory aged materials for 70/100 binder are given in Figure 6a and for binder A in Figure 6b.

Figure 6a contains the measured properties of the recovered binder 70/100 from both the oven (asphalt mix) and RCAT (asphalt mastic) accelerated laboratory ageing protocol in comparison to collected field data. [9] As function of the ageing time, the stiffness (G*) of the binders increases and the phase angle (δ) decreases. Hence flexible binders with high relaxation capabilities can be found in the lower right quadrant of the Black-Space diagram and stiff strongly elastic binders in the upper left. Thus, the presented data for these laboratory aged materials clearly show that these recovered binders after ageing still have higher flexibility and relaxation capacity than the examined field aged materials because they display much lower moduli and are still more viscous.

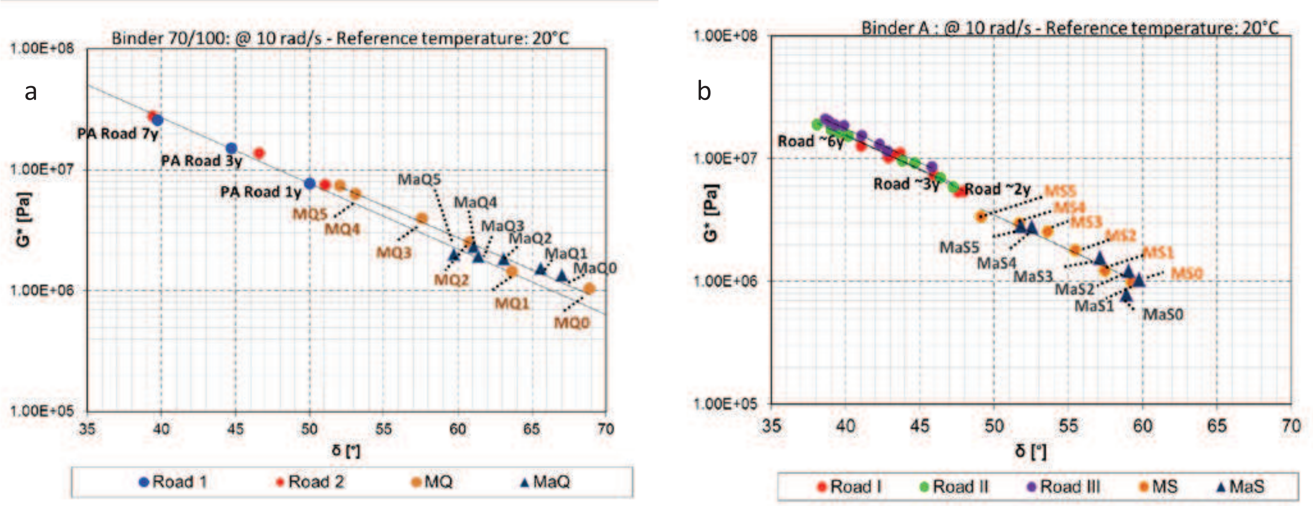

Figure 6: Black-Space diagram - Shearmodulus (G*) as function of the phase angle (δ) for a) binder 70/100 as applied in structures with single-layer PA 16 on several Dutch highways [9] and recovered 70/100 binder from an oven aged asphalt mix (PA 8) and an RCAT aged mastic; b) polymer modified binder "A" as applied in structures with PA 8 as a top=layer of Two-layer PA on several Dutch highway sections[7], recovered binder "A" from an oven aged asphalt mix and the RCAT aged mastic.

When looking at the severity of the accelerated ageing, it can be observed that the oven ageing of the asphalt mix (MQ) was stronger (higher acceleration) in comparison to the RCAT ageing of the asphalt mastic (MaQ). This was is in agreement with the earlier presented results. Although the oven ageing procedure was found to have a much higher accelerating factor, not even the 9 days of ageing represents 1 year of ageing in the field. This implies that much longer durations of ageing are required to simulate ageing in the field. The field representation of the oven ageing protocol for the asphalt mix appears to be good and the gradual increase in stiffness (G*) and increase in elastic behavior (decrease of δ) follows a comparable trend with an equal slope. This is not the case for the RCAT ageing protocol applied for asphalt mastic. Here the observed trend differs from the slope found for the field aged material and representation is disputed. It was already discussed earlier that the RCAT ageing of the asphalt mastic was not as severe as found for the oven ageing of the asphalt mix, but here it is also emphasized that the ageing process is not found analogue to field ageing development.

Similar to Figure 4a, Figure 4b contains both the measured properties of the recovered binder A from the oven (asphalt mixture) accelerated laboratory aged materials as well as from the available field aged materials [7]. The presented data in Figure 4b also show that these recovered binders are still significantly more compliant after ageing than the examined field aged materials because much lower moduli values and higher values for the phase angle are demonstrated. Also for binder A not even the 9 days of ageing has demonstrated to represent 1 year of ageing in the field. The field representation of the oven ageing protocol for the asphalt mix for binder A is found to be as good as for 70/100 binder and again the gradual increase in stiffness (G*) and increase in elastic behavior (decrease of δ) follows the same trend with an equal slope. The oven ageing of the asphalt mix was approximately equally as strong (similar degree of acceleration) in comparison to the oven ageing of the binder 70/100. This is an interesting observation as it implies that the oven ageing protocol potentially could be applied generically to pen grade and modified binders. When comparing both ageing methods for binder A, it can be observed that the oven ageing of the asphalt mix (MaS) was slightly stronger (higher acceleration) in comparison to the RCAT ageing of the asphalt mastic (MaS). This is also in agreement to the earlier presented results. The field representation of the RCAT ageing protocol for binder A in mastic appears to be good and the gradual increase in stiffness (G*) and increase in elastic behavior (decrease of δ) follows a comparable trend to the oven ageing. Here it is observed that the trend for the mastic ageing of binder A is analogue to the field performance and thus differs from the slope found for the RCAT ageing of the binder70/100 paving bitumen. Nevertheless also for binder A the results of both ageing protocols indicate that analogue ageing as in the field can be achieved by thermally/oven ageing the two binders, but that much longer durations of ageing are required to simulate years of ageing in the field. Under the current circumstances ageing periods of 40 – 60 days will at least be required in order to achieve material properties that are in the vicinity of the onset of damage around 4 - 5 years for the examined wearing courses.

4 Conclusions

The developed oven ageing protocol appears suitable for ageing both standard paving grade and SBS modified binders in asphalt mixtures, and results in representative and accelerated ageing when specifically compared to field ageing of PA. The developed RCAT ageing protocol appears less suitable for ageing standard paving grade binders in asphalt mastics and does not always result in representative and accelerated ageing when compared to field ageing. Based on the relative low degree of acceleration found for the oven ageing for binders in asphalt mixtures, the duration of the accelerated ageing will have to be at least 40 – 60 days in order to approximate field aged binder properties.

5 Acknowledgements

The authors would like to thank Mohammed Jafar (TNO) in performing the recovery of the asphalt binders.

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