

Impact of Conditioning Temperature on Thermal and Oxidative Aging of Hot Mix Asphalt

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ABSTRACT

Bitumen is subject to aging due to various environmental impacts. On the macroscale, aging leads to increased stiffness and brittleness, asphalt pavements become more prone to cracking. Thus, the aging behavior of bitumen has a crucial impact on durability, as well as recyclability of pavements. For the simulation of long-term aging of hot mix asphalt (HMA), different approaches have been taken in the last decades mainly based on conditioning at high temperatures to increase oxidation. Thereby, different reactions (chemical and physical) than in the field occur due the higher temperatures. LTA in the field is mainly triggered from slow oxidation processes, whereby at higher temperatures a combination of additional processes are triggered, that cannot be found in field. Since, the additional processes appear always jointly, this is called “thermal aging” within this paper. An alternative approach to age HMA in the laboratory, the Viennese Ageing Procedure (VAPro), has been developed lately. It aims at reducing the conditioning temperature to actual temperatures occurring in the field and increasing the oxidative reaction by using highly oxidative gases. These highly oxidant gases, e.g. ozone or nitric oxides, occur in the field as well in terms of photo smog. This paper investigates the impact of thermal and oxidative effects on long-term aging of asphalt mix specimens in the laboratory and try to separate the effects. Therefore, specimens are placed in a heating cabinet at different temperatures ranging from +60°C to +135°C. One part of the specimens is exposed to the ambient atmosphere, while the other part is stored under nitrogen atmosphere. Thereby, oxidative aging is avoided for the nitrogen stored specimens and the combination of thermal effects can be separated from oxidative effects. DSR tests are carried out on virgin, short-term (RTFOT) and long-term aged bitumen (RTFOT+PAV), as well as on recovered bitumen of both conditioning groups. The results can be used for a better understanding at which conditioning temperature thermal aging becomes a relevant factor for asphalt mixtures. This threshold temperature can also be seen as limit value for lab-aging procedures that should not be exceeded to avoid (thermal) aging effects that cannot occur in the field due to limited maximum temperatures.

Keywords: hot mix asphalt, thermal aging, oxidative aging, nitrogen, oven aging, stiffness, binder

1. INTRODUCTION

Bitumen as an organic material is subject to changes in its behavior throughout its life by a combination of several chemical and physical processes, that are triggered by temperature (oxidative aging). In pavement engineering, aging of bitumen and bituminous bound pavements

1 is divided into short-term aging (STA) in the process of HMA production and compaction within
2 a few hours and long-term aging (LTA) of a pavement during its in-service life within years.
3 STA is triggered by fast chemical oxidation due to high temperatures and a high specific surface
4 contacting with oxidant agents at mix production, as well as an physical effect, the evaporation
5 of remaining volatile components from the bitumen (thermal aging) [1, 2]. LTA is driven by
6 slow oxidation especially of the upper pavement layers by oxidant gases available in the
7 atmosphere (e.g. ozone, nitric oxides) [3]. Bitumen becomes stiffer and more brittle and thus,
8 pavements are more prone to failure by low-temperature and fatigue cracking with increasing
9 aging of the binder [4, 5]. Since bitumen aging affects durability and recyclability of pavements
10 crucially, it is important to assess aging behavior and resistance to aging of binders and mixes at
11 the stage of mix design optimization to achieve cost and energy efficient pavements with low
12 maintenance demands, a long service-life and high recycling potential.

13 The mineral component and mix design of a pavement can have an impact on aging of
14 the mix. Thus, it seems important to have a standardized method for LTA of HMA in the lab as
15 well. Therefore, HMA aging procedures could assist in analyzing changes of HMA material
16 behavior due to aging from changes of binder behavior. More than 30 lab-aging procedure of
17 loose or compacted asphalt mix have been developed in the last decades [6-9]. Most of these
18 methods have to be seen as critical due to high temperatures (+100°C and higher) that are used in
19 aging protocols for loose HMA exceed temperatures that usually occur in surface layers of
20 pavements. Additional thermal effects (e.g. vaporization of further volatile binder components)
21 could be activated that cannot occur in the field. In addition, high temperature could lead to other
22 chemical reactions than in the field, like increased oligomerization and polymerization with less
23 decomposition reactions.

24 Thus, the main objective of this paper is to analyze the influence of thermal aging (by
25 separating thermal from oxidative aging effects) at different temperatures that are commonly
26 used for LTA procedures. The results should be used to recommend limit aging temperatures for
27 laboratory LTA of HMA. These temperatures should stay below values where relevant thermal
28 aging effects occur. Similar studies for STA in the laboratory has been carried out with Nitrogen
29 Rolling Thin Film Oven Tests NRTFOT by Parmeggiani. [10]

30 Preliminary analysis of the thermal effects at LTA procedures were carried out by an
31 aging study at TU Wien [6]. The results showed, that no thermal effects could be observed.
32 HMA specimens where placed in nitrogen atmospheres at +60°C for 4 days. The aging level of
33 the extracted bitumen was comparable with bitumen from lab-produced specimens without
34 further aging procedures.
35

36 **2. MATERIALS, TEST METHODS AND EXPERIMENTAL PROGRAMM**

37 **2.1 Materials**

38
39 For the presented study, an asphalt concrete with a maximum nominal aggregate size of
40 11mm (AC 11) was employed. The coarse aggregates used for the mix is a porphyrite, the filler
41 is powdered limestone. As a binder two (I / II) unmodified 70/100 pen (PG 58-22 / 91 pen and
42 PG 64-22 / 79 pen) were used. The binder content was set for both mixtures (binder I and II) to
43 5.2% by mass with a target void content of 8.0% by volume. The maximum density of the AC 11
44 70/100 was determined to be 2.593 kg/m³ (I) and 2.562 kg/m³ (II). The grading curve is within
45 the borders according to national specifications [11].

1
2 **2.2 Specimen Preparation**
3

4 The mix was prepared in a laboratory reverse-rotation compulsory mixer, according to
5 EN 12697-35, with a mixing temperature of +170°C. HMA slabs (50x26x4 cm) were compacted
6 in a roller compactor according to EN 12697-33. The compactor consists of a roller segment for
7 compacting the slabs, which corresponds to the dimensions of a standard roller compactor used
8 in the field. All slabs were compacted with one lift. From the slabs, eight specimens are cored
9 out with a diameter of 100mm. The air void content of the specimens used for the test program
10 range from 6,6 to 8,3 % by volume for mixture I and from 5,9 to 8,0 % by volume for mixture II.

11 For bitumen testing, bitumen was extracted according to EN 12697-3 with
12 tetrachloroethylene (C2Cl4) as a solvent. The solvent-bitumen solution was distilled according to
13 EN 12697-3 to recover the binder samples.
14

15 **2.3 Experimental Program**
16

17 Table 1 provides an overview of the test program. The study looked into the impact of
18 temperature T_{air} by comparison of the nitrogen and ambient air stored specimen. Therefore, tests
19 on four and three different temperatures $T_{air}= +60 / (+85) / +110 / +135^{\circ}C$ were carried out. The
20 results provide information on how the rheological behavior of the binder (change of $|G^*|$ and
21 phase angle) changes due to thermal and oxidative aging effects. Furthermore, the dynamic shear
22 modulus $|G^*|$ of the same virgin and RTFOT+PAV-aged binder were determined as benchmarks.
23

24 **Table 1: Test Program**
25

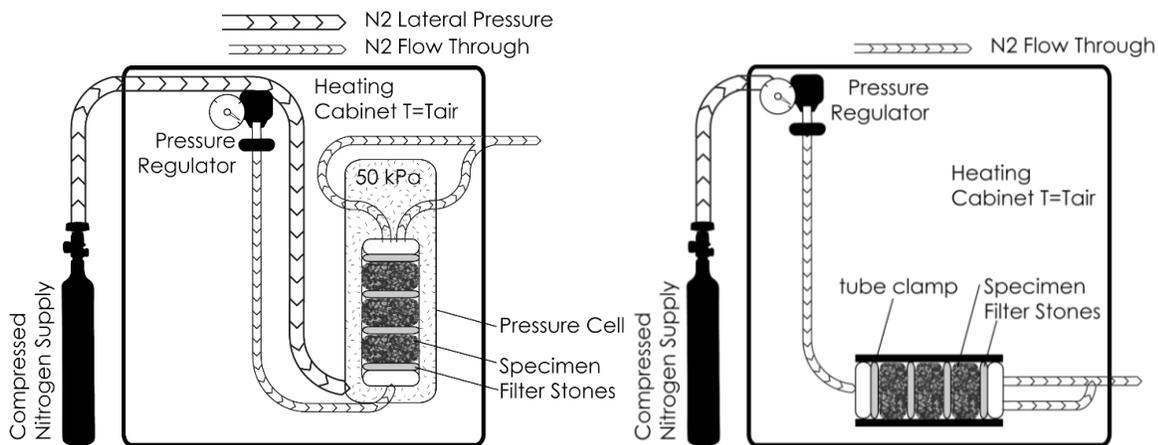
		BINDER I 70/100 PEN PG 58-22				BINDER II 70/100 PEN PG 64-22			
		# of Specimens [-]		# of recovered binder samples and DSR SHRP Tests		# of Specimens [-]		# of recovered binder samples and DSR SHRP Tests	
		Aging duration: 4 days				Aging duration: 3 days			
Temp T_{air} [° C]		N2	Air	N2	Air	N2	Air	N2	Air
+60		3	3	1	1	3	3	3	3
+85		3	3	1	1	-	-	-	-
+110		3	3	1	1	3	3	3	3
+135		3	3	1	1	3	3	3	3

26
27 **2.4 Aging Procedure**
28

29 The left picture in FIGURE 1 shows the setup and equipment which was used for the
30 aging procedure for BINDER I. The equipment and set up is based on VAPro - Viennese Ageing
31 Procedure [6]. HMA specimens are assembled within a triaxial cell between filter stones and are

1 covered by an elastic membrane. The complete system was flooded with nitrogen for a few
 2 minutes to expel any ambient air. An overpressure of 50 kPa was applied within the triaxial cell.
 3 The specimens were also flowed through with nitrogen for a few minutes to saturate all air voids.
 4 The flow pressure was set to 25 kPa. Due to the higher lateral pressure, the elastic membrane is
 5 pressed onto the specimen surface and the gas is forced to flow through the specimen, instead of
 6 passing on the outside. Afterwards all inlets were closed to retain the nitrogen within the pressure
 7 cell and specimen. The triaxial cell is located in a heating cabinet with sealed inlets where
 8 temperature T_{air} was varied for the experimental run for an aging duration of four days. After the
 9 first aging study of binder I, it turned out that a set-up is prone for damages. It consists of a
 10 glass cylinder that is sealed with metal adapters with a slight pressure in axial direction, which
 11 can lead quickly to cracks in the glass cylinder.

12
 13 **FIGURE 1 : Setup for nitrogen Pressure cell aging (left)- BINDER I | Setup for tube**
 14 **clamp aging(right)- BINDER II**
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16
 17
 18 To overcome this downside of setup I, an alternative approach was followed up to carry
 19 out aging on specimens from binder II, which is shown in the right picture in FIGURE 1. A pipe
 20 clamp (DN=100 mm) was used to seal the specimens on the lateral surface. HMA specimens are
 21 also assembled in a row between filter stones and two endplates. The pipe clamp can be perfectly
 22 fitted to the specimen dimensions. This equipment is more durable for aging tests with forced gas
 23 flow at temperatures far higher than +100°C. In this stage, the specimens were continuously
 24 flowed through with nitrogen for an aging duration of three days. In general, conditioning under
 25 nitrogen atmosphere is supposed to prevent any oxidative aging and thus, only trigger thermal
 26 aging effects. In a next step, 3 HMA specimens were placed in the ambient air of the heating
 27 cabinet at each selected aging temperature for four (binder I) or three (binder II) days. This time
 28 the heating cabinet was equipped with a forced convection. The HMA specimens are exposed to
 29 all reactive oxygen species (ROS), which are present in the ambient air. This leads to different
 30 oxidation processes at the varied temperatures T_{air} . To maintain its proper shape, the specimens
 31 are positioned in Marshall compaction molds and wrapped with silicone foil as a release layer.
 32 Ambient air aging is supposed to trigger both, oxidative, as well as thermal aging effects.

33 By comparing results from specimens and recovered binders subjected to nitrogen and
 34 ambient air aging, thermal and oxidative aging effects can be separated.

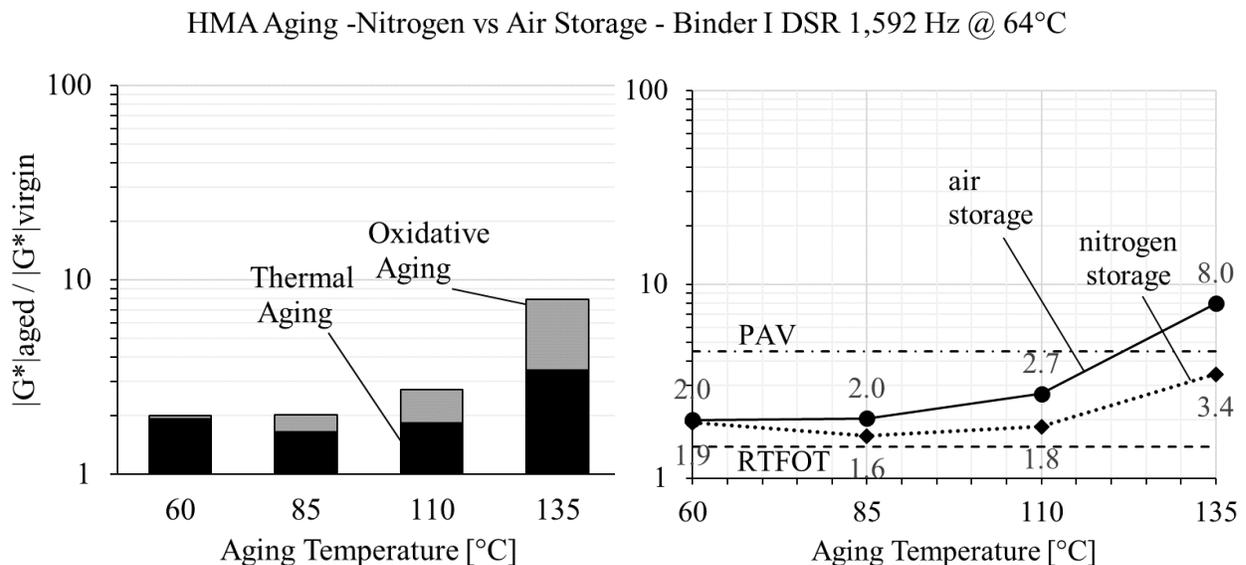
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2 **2.5 Dynamic shear modulus of bitumen**
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4 Dynamic Shear Rheometer (DSR) tests were carried out on bitumen samples recovered
5 from all lab-aged (short-term and long-term nitrogen vs. ambient air) HMA specimens. The test
6 conditions were chosen according to the SHRP procedure [1] and EN 14770 with a temperature
7 sweep from +46°C to +82°C using the large plate (diameter: 25 mm) and a 1 mm gap. A
8 frequency of 1.592 Hz is employed. From test data the dynamic shear modulus $|G^*|$ and the
9 phase angle ϕ vs. frequency are determined.
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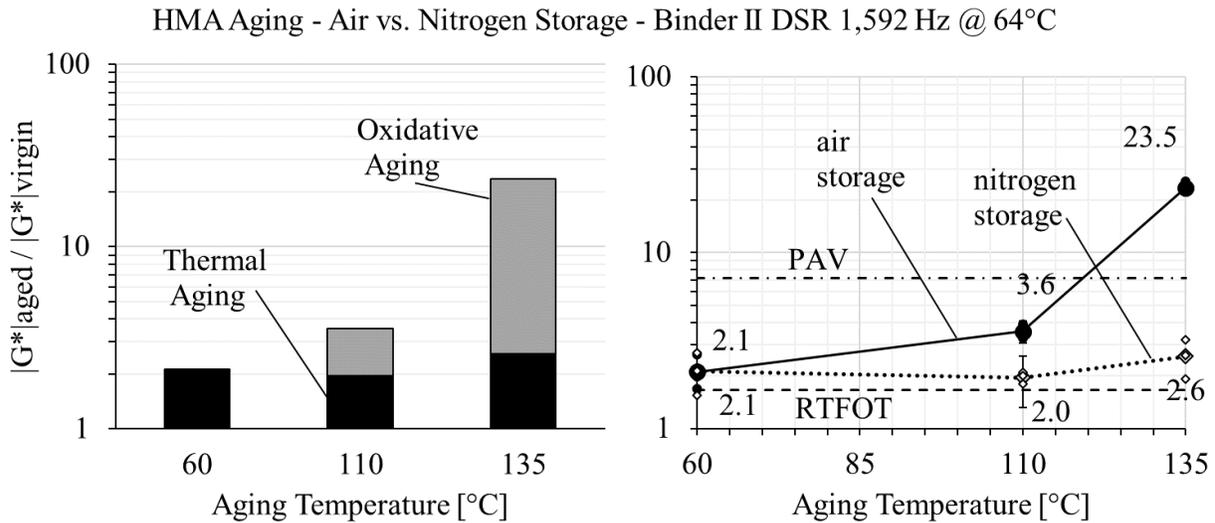
11 **3. DISCUSSION AND LABARATORY TEST RESULTS**

12 To investigate potential thermal effects from temperature, bitumen from all nitrogen stored and
13 ambient air stored samples were extracted and recovered from the HMA. Analysis of changes of
14 the viscoelastic behavior were carried out. In addition, STA bitumen by RTFOT and LTA
15 bitumen by RTFOT+PAV was tested as well to compare standardized bitumen aging procedures
16 to the presented procedures. The results are presented in Figure 2. It shows the relative change in
17 dynamic shear modulus $|G^*|$ from recovered bitumen samples vs. virgin bitumen. The dotted
18 lines represent data from the RTFOT (B I 1.45 / B II 1.66 [-]) and RTFOT+PAV (B I 4.49 / B II
19 7.17 [-]) aged bitumen. For both binders, the changes in the $|G^*|$ vs. the aging temperature of the
20 different procedures are shown. The results of binder I as well as binder II indicate no significant
21 changes in the mechanical behavior for the nitrogen-stored samples below 110°C. At 135°C, $|G^*|$
22 significantly increased for both binders in the nitrogen atmosphere. If it is assumed that the
23 mechanical changes at the temperatures of 110°C and below are only due the STA at the slab
24 production, it can be concluded that aging effects that can be attributed to thermal aging are
25 starting at temperatures between 110°C and 135°C. Between +60°C and +110°C all binders are
26 1.6 -2.0 times stiffer than the virgin binder, whereas the +135°C nitrogen samples are 2.6 and 3.4
27 times stiffer.
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29 **FIGURE 2 : Change in dynamic shear modulus $|G^*|$ of bitumen recovered from lab-**
30 **aged HMA specimen (4 days binder I – 3 days binder II) to virgin bitumen sample**
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Looking at the data from the ambient air aged samples, almost no effects occur until 85°C. Starting at 110°C significant changes can be seen. The oxidative species of the ambient air are reactive enough to trigger oxidative aging only above 110°C. The difference between nitrogen stored and ambient air stored samples can be attributed to oxidative aging effects. Both binders follow the same trend in relation to the aging temperature. Comparing both binder, it must be stated, that binder I was conditioned for 4 days, while binder II was aged for 3 days. Despite this, binder II is more prone to aging. The increase of stiffness is higher for RTFOT+PAV aged samples and for samples aged as HMA in ambient air in comparison to samples from binder I.

14 4. CONCLUSIONS

15 The main drive for the research project presented within this paper is to investigate the potential
16 of ambient air conditioning as realistic LTA procedures and to isolate thermal aging effects from
17 oxidative aging effects. Considering realistic field conditions (max. 65°C on asphalt surface),
18 thermal effects should not be triggered by laboratory LTA procedures since they are not expected
19 to occur in the field. To prevent thermal aging effects, conditioning temperatures must kept
20 below 110°C as it is shown in this paper. At the same time, ambient air conditioning for HMA
21 specimens is only suitable as an LTA procedure for temperatures above 110°C. Below this
22 threshold, the conditioning hardly triggers any oxidative effects in a short amount of time and is
23 therefore inefficient. Additionally, at temperatures above 110°C, the viscosity of some binder
24 grades is so low that permanent changes in the structure of compacted HMA specimens are
25 likely to appear upon conditioning for an extended period of time. Results from mechanical tests
26 of such samples are therefore biased.

27 Summarizing, to achieve both, realistic and efficient LTA procedures for HMA, temperatures
28 have to stay well below 110°C and oxidative aging effects have to be increased by other means,
29 e.g. the use of more reactive gases that are present in the atmosphere as well.

30 The data presented in this paper will be extended with stiffness data from recovered binder from
31 control HMA specimens that were not subjected to any aging after production. By using these

1 data, any STA effects from mixing and slab production can be taken into account and separated
2 from the analysis of LTA effects.

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