VAPro – Long-Term Ageing of HMA based on Highly Oxidant Gases

D. Maschauer, D. Steiner, B. Hofko*
Institute of Transportation, TU Wien, Gusshausstrasse 28/E230-3, 1040 Vienna, Austria
**Corresponding author: bernhard.hofko@tuwien.ac.at*

ABSTRACT: This paper presents a recently developed conditioning method for long-term ageing of compacted HMA specimens in the lab. The Viennese Ageing Procedure (VAPro) works at temperatures and pressures that occur in the field on pavement surfaces regularly. To increase the oxidation reaction, highly oxidant gases (ozone and nitric oxides) are led through the specimen. It is expected that the chemical reactions triggered by VAPro are similar to field ageing. The presented study shows the applicability on paving grade and SBS-modified bitumens from different sources. Binders were extracted after VAPro-ageing and tested in DSR. Bitumen samples from different sources show significantly different ageing susceptibility. The increase in |G*| varies from 3 for the least ageing susceptible bitumen to 13 for the samples with highest ageing rate. Significant differences between PAV aged bitumen and bitumen from VAPro-aged samples were found. VAPro can discriminate binders with different ageing susceptibility more sensitively than PAV.

Keywords: bitumen, ageing, oxidation, VAPro, DSR, RTFOT, PAV

1 INTRODUCTION

Bitumen is an organic material derived from crude oil refinery with a highly complex chemical composition (Handle et al. 2016). It exhibits highly temperature and loading-time depending viscoelastic behavior (Eberhardsteiner et al. 2015a). Due to its organic origin, it is prone to oxidation, which changes the mechanical behavior towards higher stiffness and brittleness (Petersen 2009). These changes are usually summarized as ageing. For application in road engineering, ageing is divided into short-term (STA) and long-term ageing (LTA). STA includes all stages in asphalt mix production and laying. It is characterized by high temperatures and thus, fast oxidation processes. LTA includes all subsequent steps after the asphalt mix has been compacted and cooled down. Oxidation rates are much lower for LTA due to the denser material and lower temperatures. Additional effects by UV radiation and moisture can contribute to changes in the material behavior over time. LTA is limited to the asphalt mix close to the surface for dense asphalt mixtures but may reach further into the pavement for open graded mixtures. Bitumen ageing in the field leads to higher risk of low-temperature and fatigue cracking and thus, is a factor that limits the service-life of asphalt mix pavements (Hofko et al. 2014, Steiner et al. 2016).

Due to the highly complex composition of bitumen, the observable macroscopic changes in bitumen due to ageing are still up for scientific discussion (Handle et al. 2017). One theory, based on a detailed study that included microscopic and spectroscopic analysis, as well as mechanical testing and micromechanical modeling is based on the idea that bitumen is a three-phase material (Eberhardsteiner et al. 2015b). It consists of a lowly polar continuous matrix of maltenes and higher polar inclusions (asphaltenes). To realize a working dispersion, the asphaltenes are surrounded by a shell of resins and aromatics. This shell balances out the polarity gap between maltenes and asphaltenes. Since maltenes consist mainly of saturates, the maltene phase is not prone to oxidation. Thus, oxidation starts at the outside of the shell and increases the polarity of the shell steadily. This decreases the quality of dispersion and thus, increases brittleness as it is observed for aged bitumen samples (Hofko et al. 2015).

To assess the long-term quality of a bitumen, standardized conditioning methods are available for

STA and LTA simulation in the lab. For STA, the Rolling Thin Film Oven Test (RTFOT) (CEN 2007) is a commonly used method. For LTA, the Pressure Ageing Vessel (PAV) (CEN 2012) is often used.

The mineral component and mix design can have an impact on ageing as well. Thus, it is important to have a standardized method for LTA of hot mix asphalt (HMA). More than 30 different lab ageing procedures of loose or compacted asphalt mix have been developed in the last decades (Bell et al. 1994). However, most of these methods have to be seen as critical due to the following reasons:

- Methods that age loose HMA before specimen compaction may influence compactability and quality of cohesion and adhesion due to bitumen ageing. Thus, it cannot be determined from subsequent testing whether differences between unaged and aged samples are due to ageing or imperfect compaction.
- High temperatures (> +100°C) that are used in ageing protocols for loose HMA exceed temperatures that normally occur in surface layers. Additional thermal ageing effects (e.g. vaporization for further volatile components) could be activated that cannot occur in the field. High temperatures could also trigger other chemical reaction than in the field, e.g. increased oligomerisation and polymerization with less decomposition reactions. Non oxidative aging effects are triggered above 110°C (Steiner et al. 2018).
- The duration of existing ageing protocols for compacted specimens is too high for some methods (up to several weeks) to be efficient for routine testing. Methods using realistic temperature ranges exceed 2 weeks of testing time. (Board et al. 2017)
- Some protocols apply high pressure (comparable to pressures in the PAV). Again, these conditions could lead to other chemical reactions than those occurring in the field.

Thus, the main objective of ongoing research is to develop an optimized procedure for ageing of compacted HMA specimens in the lab. To ensure realistic and efficient conditioning, the emphasis is put on the following issues:

- Temperature and pressure should not exceed values that are regularly achieved on surface layers in the field.
- Increased oxidation should be achieved by using high concentrations of highly reactant gases that occur in the field as well but in lower concentrations.
- The procedure should not last longer than a couple of days.

Based on these considerations, a conditioning method, the Viennese Ageing Procedure (VAPro), has been developed recently (Steiner et al. 2015).

2 MATERIALS

In this study, an asphalt concrete with a maximum nominal aggregate size of 11 mm (AC 11) was used with porphyritic coarse aggregates and powdered limestone as filler material. Mixes with six different bituminous binders were produced, four paving grade bitumens (70/100 pen) of different origins and two SBS-modified binders (PmB 45/80-65) from the same source but different production years. Table 1Table 1 gives information about basic characteristics of all binders.

The mixes were produced in a laboratory mixer according to EN 12697-35 with a mixing temperature of 170°C for the pen graded bitumens and 185°C for the PmBs, respectively. Asphalt slabs were compacted in a steel roller segment compactor according to EN 12697-33. From these slabs (50x26x4 cm), eight specimens with a diameter of 100 mm were cored. For each mix, three specimens with an air void content of 7.0% ± 1.0% by volume were selected for subsequent ageing and testing.

Table 1. Bitumen characteristics

Bitumen Type	Origin	Penetration in 1/10mm	Dynamic Shear Modulus @ 1.59Hz, 64°C in MPa
70/100	A	88	1.34
70/100	B	79	1.09
70/100	C	64	1.77
70/100	D	55	1.57
PmB 45/80-65	A (2012)	67	5.58
PmB 45/80-65	A (2017)	65	4.97

3 PRINCIPLE OF VAPRO

Figure 1shows a photo and sketch of VAPro .

Compressed air from a central laboratory supply is directed through an ozone generator. The generator enriches the air with ozone and nitric oxides using a dielectric barrier discharge tube. Subsequently, a flow regulator ensures a constant flow rate of 0.9 to 1.1 l/min. The ageing gas is heated up in a heating coil, which is placed in a beaker glass filled with vegetable oil (70°C (T_{liq})) that is placed on a heatable magnetic stirrer. The specimen is placed within a triaxial cell between two filter stones and is covered by an elastic membrane.

A slight overpressure of 80 kPa in the triaxial cell ensures that the membrane is pressed against the specimen so that the gas flows through the specimen and not around it. The triaxial cell and heating coil are housed in a heating cabinet at an air temperature

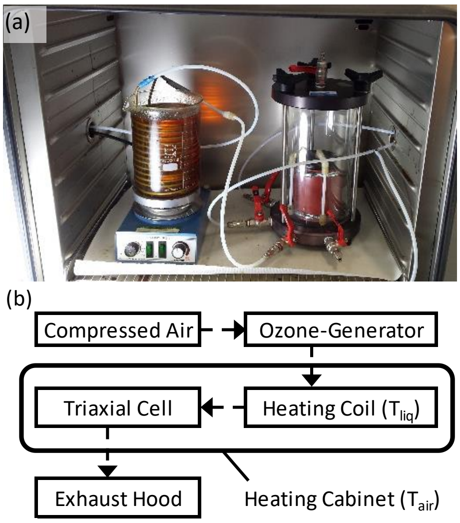

Figure 1. (a) Photo and (b) schematic design of VAPro

of +60°C (T_{air}). Contrary to former VAPro studies (Steiner et al. 2015, Frigio et al. 2016) the standard aging time was reduced since VAPro was continuously improved (Steiner et al. 2018). With the latest device the ageing duration was set to three days, since the binders were in a state of of RTFOT+PAV ageing after three days conditioning.

4 METHODS

HMA specimens were VAPro-aged for three days. Subsequently, the bitumen was extracted with tetrachloroethylene according to EN 12697-3. The solvent was then evaporated by vacuum distillation according to EN 12697-3.

Dynamic Shear Rheometer (DSR) tests were performed on the bitumen obtained from the VAPro-aged specimens (LTA). DSR tests were also carried out on fresh bitumen samples, extracted bitumen samples of slab residues (STA), RTFOT- and RTFOT+PAV aged samples for reference. DSR were carried out at temperatures from +4°C to +82°C in 6°C steps with a frequency sweep from 0.1 Hz to 10.0 Hz.

5 RESULTS

5.1 *Short-term ageing*

In a first analysis, STA due to laboratory HMA production and slab compaction are compared to RTFOT-ageing of the respective binder. Figure 2 shows results from DSR testing at a frequency of 1.59 Hz and 64°C. Similar trends can be observed for other DSR test conditions. In the diagrams, grey bars indicate RTFOT-aged samples and green bars samples extracted from HMA slab residues. Diagram (a) in Figure 2 shows the relative change of the dynamic shear modulus |G*| as a ratio of STA to virgin binder. Diagram (b) in Figure 2 shows the absolute change of the phase angle between STA and virgin binder.

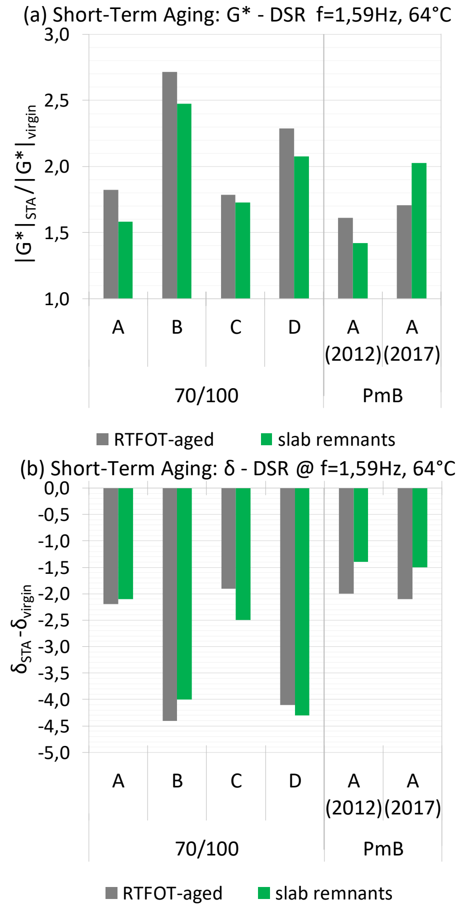

Figure 2. Comparison of STA in terms of (a) relative change of |G*| and (b) absolute change of phase lag (STA to virgin)

From the data, it can be seen that the achieved level of ageing after production are similar to the RTFOT-ageing level. The increase in stiffness ranges from 1.5 to 2.5. In most cases, the aging level of the slab residue is slightly below RTFOT ageing. Knowing that lab production of HMA is usually less severe than plant production in terms of ageing the results are plausible. It is confirmed that RTFOT is a good representation of STA for various binders, paving grade, as well as PmBs. The differences between bitumen samples from different origin can already be seen at the stage of STA. Paving grade bitumen from source B is significantly more susceptible to ageing than all other paving grade samples.

Similar statements can be given for the change in phase lag. Bitumen from slab residues and RTFOT show similar increase in elasticity. The decrease in phase lag ranges between 1.5° and 4.5°. A higher increase in stiffness corresponds to a stronger decrease in phase lag.

5.2 *Long-term ageing*

To compare LTA, RTFOT+PAV-aged samples are compared to binder samples extracted from VAPro-aged specimens. Again, Figure 3 shows results for DSR testing at 1.59 Hz and 64°C. Grey bars represent

RTFOT+PAV aged samples, green bars show binder from VAPro-aged specimens. Diagram (a) contains the relative change in |G*| as a ratio of LTA to STA aged binder samples. Diagram (b) shows the absolute change in phase angle between LTA and STA.

For the grey bars this means RTFOT+PAV to RTFOT and for the green bars VAPro to slab residues. Thus, any impact from STA is removed from the following analysis since the diagrams just show the change from STA to LTA.

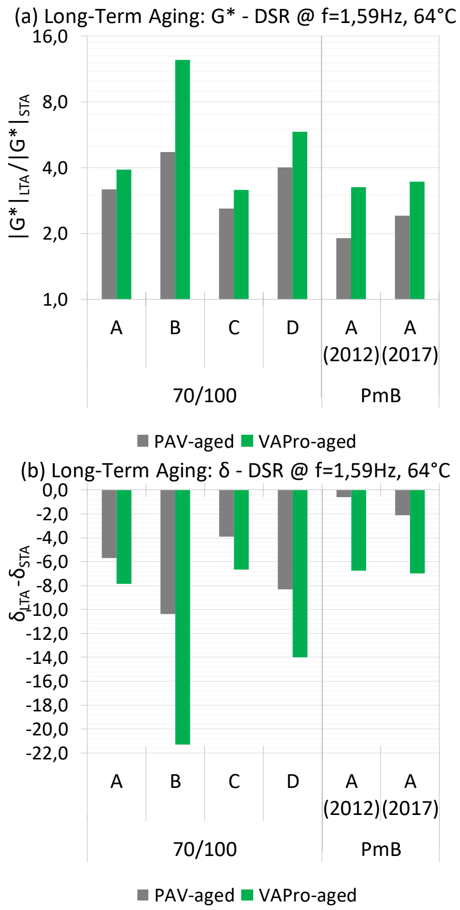

Figure 3. Comparison of LTA in terms of (a) relative change of |G*| and (b) absolute change of phase lag (LTA to STA)

The tendency of differences for LTA is similar as for STA. For the paving grade bitumens, the ageing level in terms of stiffness increase due to VAPro ranges from 3 to 13. Bitumen samples with the lowest increase in stiffness exhibit the highest |G*| in the unaged state and vice versa (cf. Table 1). For both PmBs, the ageing level is similar, which indicates a constant quality of the supplier. PAV-aged samples are below VAPro-aged samples in terms of stiffness increase. In case of bitumen from source B, the differences between PAV and VAPro is significant. While PAV does not discriminate between different bitumen sources well (stiffness increase between 3 and 4), VAPro exhibits that the actual ageing susceptibility is much higher for bitumen from source B compared to all other sources.

The analysis of the phase lag (Diagram (b) in Figure 3) shows that a stronger increase in |G*| is closely linked to a stronger decrease in the phase lag due to LTA. For all cases, the change in phase lag is stronger for all VAPro-aged samples compared to PAV-aged samples. Again, source B exhibits the largest change in phase lag (-21°) compared to all other samples (-6° to -14°).

It should be noted that the PAV-aged PmBs show barely any decrease of the phase angle, whereas VAPro ageing does induce similar increase in elasticity as for the paving grade samples.

6 SUMMARY

This paper presents a recently developed conditioning method for the simulation of LTA on compacted HMA specimens. The method called VAPro is based on the use of highly oxidant gases (ozone and nitric oxides) that exist in the atmosphere as well in lower concentrations. These gases are seen as the main drivers for oxidative LTA of HMA. Temperature (60°C) and pressure in VAPro are similar to what can be expected on a pavement surface in summer in moderate climates (e.g. up to +65°C) (Hofko et al. 2014). This ensures that no chemical reactions are triggered in the lab that cannot occur in the field. The ageing gas flows through a specimen for three days since former studies found that this induces an ageing level that roughly resembles RTFOT+PAV aged binder.

For the study in this paper, changes in the rheological behavior due to ageing of paving grade bitumen from four different sources as well as two SBS-modified binders from the same source but different production years were investigated. Bitumen was extracted from VAPro-aged specimens and tested in the DSR.

The results show that there are clear differences between different bitumen sources, which indicates that the origin of the bitumen has a strong influence on the aging behavior. The level of aging of some VAPro-aged samples differs significantly from the RTFOT+PAV-samples, which suggests that the PAV method does not capture the entire aging processes that occur in the field. Additional results of cyclic indirect tensile test, which are not included in this paper, are available for future discussion. Furthermore, N-VAPro (Steiner et al. 2018) (nitrogen atmosphere conditioning) to separate oxidative from non-oxidative Aging effects were investigated with the presented binder sources.

To investigate the low-temperature behavior of VAPro-aged specimens, the device will be further developed in the near future to age prismatic specimens for TSRST and UTST.

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