

Impact of the Ageing on Viscoelastic Properties of Bitumen with the Liquid Surface Active Agent at Operating Temperatures

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Abstract. The paper presents the influence of the ageing on viscoelastic properties of the bitumen at road pavement operating temperatures. The ageing process of bituminous binders causes changes in physical and mechanical properties of the bitumen. This phenomenon takes place in all stages of bituminous mixtures manufacturing, namely: mixing, storage, transport, placing. Nevertheless, during the service life it occurs the increase in stiffness of asphalt binder that is caused by the physical hardening of bitumen as well as the influence of oxidation. Therefore, it is important to identify the binder properties at a high and low operating temperatures of asphalt pavement after simulation of an ageing process. In the experiment as a reference bitumen, the polymer modified bitumen PMB 45/80-65 was used. The liquid surface active agent FA (fatty amine) was used as a bitumen viscosity-reducing modifier. It was added in the amount of 0,2%, 0,4% and 0,6% by the bitumen mass. All binder properties have been determined before ageing (NEAT) and after long-term ageing simulated by the Pressure Ageing Vessel method (PAV). To determine the binder properties at high temperatures the dynamic viscosity at 60°C was tested. On the basis of test results coming from the dynamic viscosity test it was calculated the binder hardening index. The properties at a low temperature were determined by measuring the creep modulus using Bending Beam Rheometer (BBR) at four temperatures: -10°C, -16°C, -22°C and -28°C. The stiffness creep modulus “S” and parameter “m” were determined. On the basis of dynamic viscosity test it was found that the ageing process caused a slight decrease in a dynamic viscosity. The level of a hardening index considerably increased at 0.6% fatty amine content. The long-term ageing process had a minor effect on stiffening of a polymer modified bitumen with FA additive regardless of a low temperature and an amount of fatty amine content.

1. Introduction

Ever growing traffic volumes and vehicle axle loads require new pavement structure designs, new production technologies and new building materials [1]. Bitumen binder is the primary component of asphalt mixtures designed for road pavements. Its durability is a critical factor. However, during the subsequent stages of production, storage, transport and placement of the mixtures, and at the in-service stage, the composition and chemical structure of bitumen undergo adverse physical and chemical changes called ageing [2].

Gradual loss of viscoelastic properties due to ageing makes bitumen binders harder and brittle [3]. Physical hardening of bitumen can play a major role in thermal cracking of flexible road pavements



[4]. To slow down this process, Warm Mix Asphalt (WMA) [5, 6] technologies are being used more widely for their ability to lower the production and placement temperatures by about 20-30°C compared with conventional Hot Mix Asphalt (HMA) technology [7, 8]. Adequately high workability of the mixture can be attained without the need to heat its constituents to the traditionally used temperatures through the addition of chemical agents lowering the viscosity of bitumen. One of such agents is a liquid surface active agent – fatty amine (FA) [9].

This paper discusses effects of the ageing process on viscoelastic properties of FA modified bitumen at high and low service temperatures. The response of bitumen to high and low temperatures has an important effect on the resistance of asphalt pavements to rutting and cracking. Bitumen binders are referred to as viscoelastic materials whose viscous and elastic properties are very sensitive to temperature. Increased elastic component will improve the high-temperature performance of the asphalt, whereas increased viscous component will improve its low-temperature performance. The viscous to elastic component ratio is important to high- and low-temperature performance of bitumen binders [10].

2. Materials

The base material used in the tests was polymer modified bitumen PMB 45/80-65. High softening point as well as high degree of modification makes it suitable for paving at sites where high elastic recovery, cohesion and resistance to low-temperature cracking are required [11]. But the placement of PMB is difficult under adverse weather conditions (fast stiffening, compaction problems) due to high polymer content and viscosity [11].

To improve the workability of the asphalt mixture and shorten the pavement construction period, the emulsifier – a liquid surface active agent (fatty amine-FA) was added to the base bitumen [12]. The agent lowers the surface tension at the aggregate-bitumen interface [13]. Owing to its asymmetrical and polar structure, the hydrophilic element is turned in the direction of aggregate, whereas the hydrophobic element is dissolved in the bitumen [14]. Better wetting of the aggregate results in the mixture production temperature reduction up to 120°C at lower dosage levels of the fatty amine [12].

3. Test methods

All the parameters were determined before and after long-term ageing (in-service). Prior to the simulation of the in-service ageing, the samples were subjected to short-term ageing through the Rolling Thin Film Oven Test (RTFOT). Due to the use of the modifier and thus possibility to produce the mixture at low temperatures, the samples were conditioned at 135°C (instead of 163°C as set out in PN-EN 12607-1). The long-term ageing of the samples was performed in the Pressure Ageing Vessel (PAV) in compliance with PN-EN 14769 [15].

The following samples were prepared:

- 0% (polymer modified bitumen PMB 45/80-65)
- 0.2% FA (PMB 45/80-65 with an addition of 0.2% FA)
- 0.4% FA (PMB 45/80-65 with an addition of 0.4% FA)
- 0.6% FA (PMB 45/80-65 with an addition of 0.6% FA)
- 0% PAV (PMB 45/80-65 after long-term ageing in PAV)
- 0.2% FA PAV (PMB 45/80-65 with an addition of 0.2% FA after long-term ageing in PAV)
- 0.4% FA PAV (PMB 45/80-65 with an addition of 0.4% FA after long-term ageing in PAV)
- 0.6% FA PAV (PMB 45/80-65 with an addition of 0.6% FA after long-term ageing in PAV).

To determine the viscoelastic properties of the binders at high temperatures, the cone and plate test for dynamic viscosity at 60°C was performed to PN-EN 13702. The plate diameter was 25mm.

The properties of the binders at low (minus) in-service temperatures were determined through the measurement of the flexural creep stiffness by Bending Beam Rheometer (BBR) in compliance with

PN-EN 14771 for four temperatures: -10°C, -16°C, -22°C and -28°C. BBR was adopted to quantify the level of deflection or creep under a 980 ± 50 mN seating load automatically applied on the standard asphalt beam specimens lasting for a total of 240 s [16]. The dimensions of the specimens were 125/12.5/6.25 mm.

4. Results

The initial evaluation of the impact of ageing on the properties of binders at high service temperatures was performed by determining dynamic viscosity η' at 60°C at the load frequency going to zero. In other words, the test aimed at determining the level of zero shear viscosity as an indicator of the behaviour of bitumen in intact condition using the Cross/Sybilski model [17]. Zero shear viscosity is one of the most important parameters for the evaluation of binder response to long-term stress induced by traffic [18]. The results for dynamic viscosity of the PMB made with the surface active agent FA before and after PAV ageing are illustrated in Figure 1.

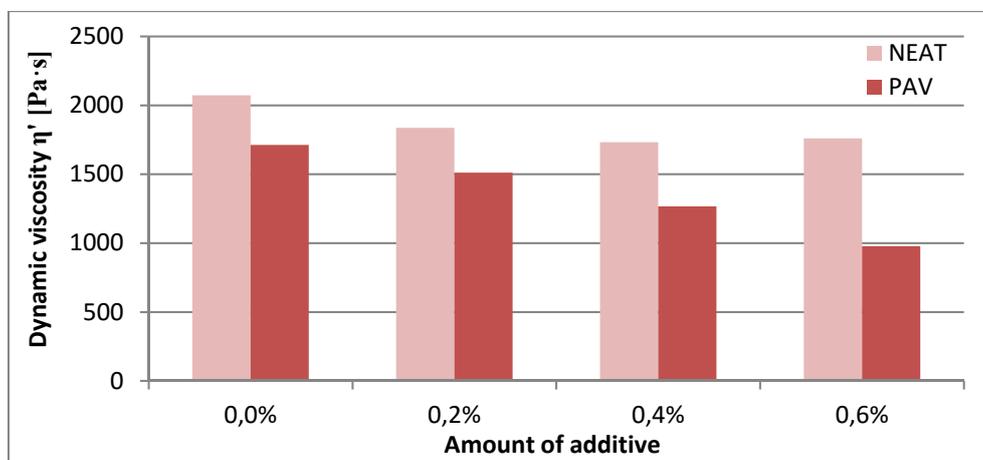


Figure 1. Dynamic viscosity of PMB with FA additive before and after PAV test

Analysis of the results in Figure 1 indicates that FA modification of PMB 45/80-65 leads to a decrease in dynamic viscosity as compared with the base bitumen, regardless of the amount of additive used. This effect is not positive, since the asphalt pavement produced with FA-modified bitumen will be less resistant to deformation than the reference bitumen. Nevertheless, the decrease is low, about 10%. In all the cases under analysis, PMB dynamic viscosity after PAV ageing also decreased in comparison with its value before the test. The highest reduction of dynamic viscosity at 60°C was recorded for the PMB modified with 0.6% FA – over 80%.

To make the assessment of the impact of ageing on the viscosity of bitumen easier, the hardening index (HI) was calculated from the formula:

$$HI = \frac{\eta'_{60}^{PAV}}{\eta'_{60}} \quad (1)$$

where:

HI – hardening index

η'_{60}^{PAV} – dynamic viscosity determined at 60°C after PAV ageing [Pa·s]

η'_{60} – dynamic viscosity determined at 60°C before PAV ageing [Pa·s]

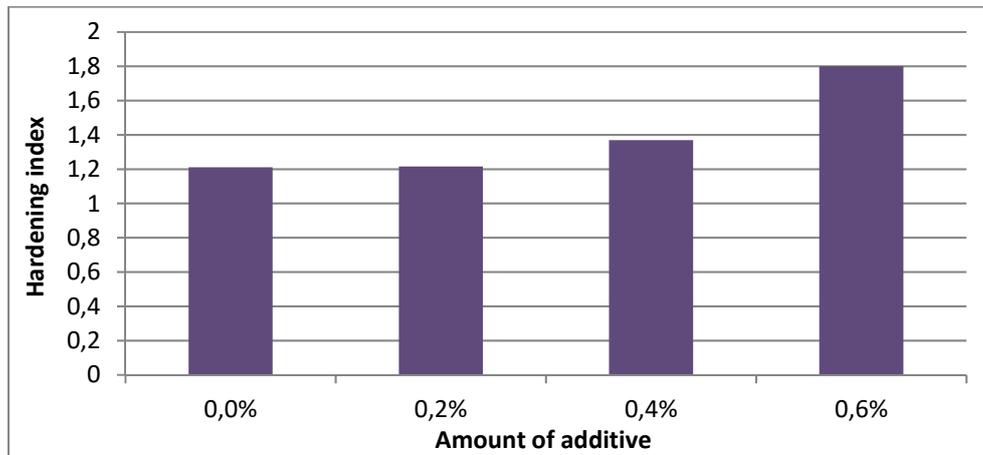


Figure 2. Hardening index of the bitumen with FA after PAV ageing.

The hardening indices calculated from the measured dynamic viscosities before and after long-term PAV ageing, Figure 2, show that an addition of 0.2% and of 0.4% FA to the PMB 45/80-65 had a negligible effect on the increase in zero shear viscosity (ZSV), whereas an addition of 0.6% FA had a significant effect on the hardening index value. The ZSV in modified bitumen is closely dependent on the polymer phase condition in the bitumen. A decrease in ZSV after ageing may be induced by the degradation of polymer chains and by inhomogeneity of the bitumen-polymer system. The presence of the surface active agent had an advantageous effect on hindering SBS chain degradation caused by the ageing process, making the base bitumen more compatible with the polymer [19].

More comprehensive evaluation of the influence of ageing on the properties of bitumen can be performed through its stiffness at low temperatures. Low-temperature properties of bitumen binders before and after long-term ageing (PAV) as per Superpave (USA) methodology were determined in the Bending Beam Rheometer (BBR) test [20]. The measurement of the bitumen stiffness using this method was used to assess the level of stiffness of a bitumen binder sample (creep stiffness S) and to evaluate the creep at static loading and elastic recovery at unloading (the m -value) [21]. The results from the tests for rheological properties of binders at low service temperatures are summarised in Figure 3 and 4 and in Table 1.

Analysis of the results shown in Figure 3 indicates that the decrease in test temperature increases the creep stiffness in each bitumen being tested. The addition of the FA modifier contributed to the decrease in creep stiffness S at each dosage and temperature used. This is a positive effect, as too high stiffness of the bitumen at low temperatures fosters the formation of cracks. The highest value of S after the PAV was recorded for the reference bitumen and the bitumen with an addition of 0.6% FA modifier.

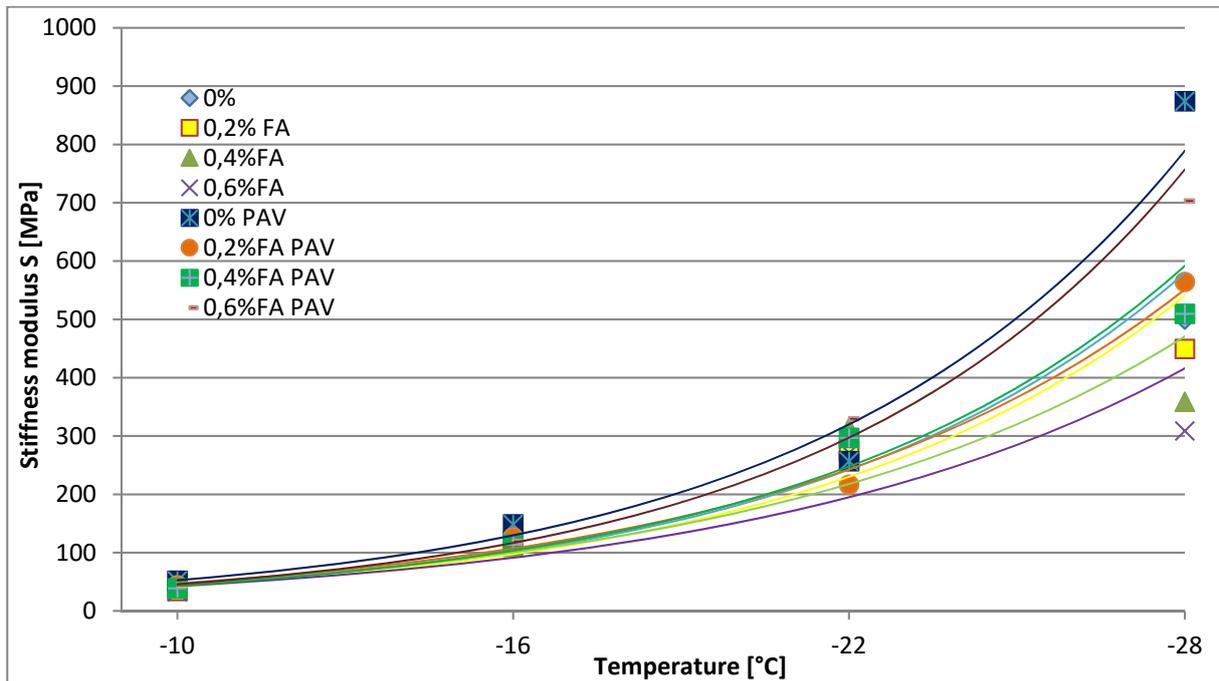


Figure 3. Creep stiffness S of the bitumen with FA before and after PAV long-term ageing

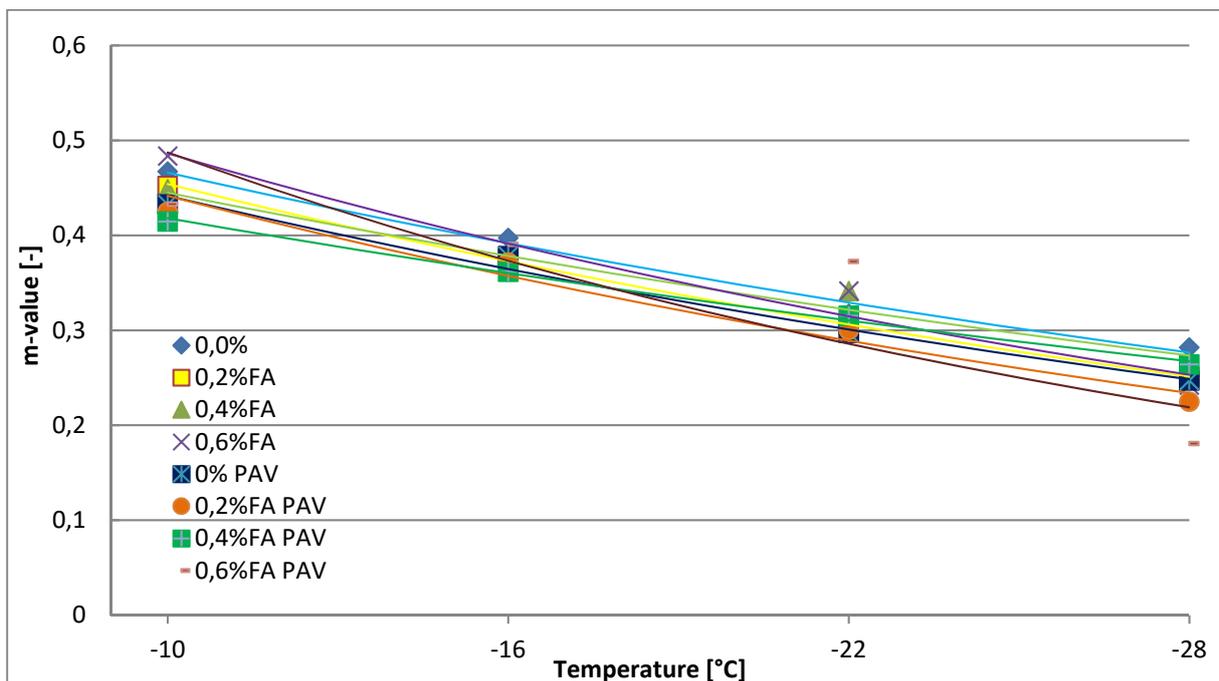


Figure 4. The m-value of the bitumen with FA before and after PAV long-term ageing

The results showing the stiffness change (m-value), Figure 4, indicate that the rate of this change under loading decreases with decreasing test temperature. The addition of 0.2% and of 0.4% of the surface active agent to the PMB 45/80-65 led to the decrease of the m-value relative to the base bitumen. After the long-term ageing in the PAV test, the m-value decreased only slightly in all bitumen under analysis. As this value is within the accuracy limit of the device, thus it can be stated

that modification of the bitumen using fatty amine at a given temperature does not significantly change the m-value.

During the tests the deflection of the beam was recorded continuously. The deflection was used to calculate the creep stiffness S and the change in the creep stiffness – the m-value at 60 sec. [22]. The following formulas were used to calculate the parameters in accordance with PN-EN 14771:

$$S_{60} = \frac{Pl^3}{4bh^3d_{60}} \quad (2)$$

$$m_{60} = \frac{\log(S_{60})}{\log(60)} \quad (3)$$

where:

- S_{60} – creep stiffness S at 60 seconds [MPa]
- P – applied constant seating load = 980mN
- l – span length (test length of specimen) = 102mm
- b – width of beam = 12.5mm
- h – height of beam = 6.25mm
- d_{60} – deflection of beam at 60seconds [mm]
- m_{60} – m-value at 60 seconds [-]
- $\log(S_{60})$ – stiffness S logarithm at 60s

The Superpave requirements take the binder's stiffness $S=300\text{MPa}$ as the limiting value below which bitumen remains viscoelastic and above which it shows elastic behaviour [1]. However, determining the value of S is insufficient due to the need to take into account the behaviour of the bitumen binder resulting from long-term loading. Thus, in addition to the creep stiffness, the rate at which the binder stiffness changed was analysed. The S change rate is regulated through the m-value, which after 60 seconds of loading should be ≥ 0.3 .

Table 1. Summary of stiffness moduli S, m parameter values and critical temperatures per Superpave criteria

Binder	S [MPa]				m [-]				T [°C]		Critical temp. [°C]
	-10°C	-16°C	-22°C	-28°C	-10°C	-16°C	-22°C	-28°C	$S_{60} \leq 300$	$m_{60} \geq 0,3$	
0%	36.7	115.3	283.7	499.6	0.467	0.397	0.317	0.282	-23.4	-25.2	-33.4
0.2%FA	34.2	119.6	274.8	449.1	0.452	0.375	0.306	0.251	-23.8	-22.6	-32.6
0.4%FA	38.8	109.6	313.9	358.4	0.449	0.363	0.341	0.267	-24.4	-24.5	-34.4
0.6%FA	32.7	115.7	270.9	308.6	0.484	0.379	0.342	0.242	-25.5	-23.3	-33.3
0% PAV	51.7	148.2	256.7	873.9	0.433	0.378	0.297	0.247	-21.5	-22.1	-31.5
0.2%FA PAV	43.9	126.2	216.5	564.0	0.425	0.371	0.301	0.225	-23.5	-21.0	-31.0
0.4%FA PAV	38.5	113.6	296.6	509.2	0.415	0.362	0.315	0.264	-23.3	-23.4	-33.3
0.6%FA PAV	43.6	119.1	329.0	702.6	0.433	0.390	0.373	0.181	-22.0	-20.9	-30.9

Analysis of temperature readings at $S_{60} \leq 300\text{MPa}$ and $m_{60} \geq 0.3$ shows that they are similar. The long-term ageing had a minor effect on their increase. According to the Superpave criteria, the higher of the two temperatures obtained for both criteria should be adopted for the evaluation of the bitumen [1].

The creep stiffness S and the m -value can be used to determine the critical temperature per Superpave by selecting the higher temperature minus 10°C at which the requirements $S \leq 300\text{MPa}$ and $m \geq 0.3$ at 60 seconds of loading are met (Table 1).

The results obtained from the BBR test indicate that the bitumen with fatty amine before ageing have comparable critical temperature to that of the input bitumen. After the PAV test, the critical temperature increased slightly (by max. 2.5°C). It can be thus stated that the long-term ageing has a minor effect on the stiffening of FA-based PMB 45/80-65. Absence of a significant increase in the stiffness of the polymer modified bitumen with fatty amine additive suggests that the fatty amine acts as an inhibitor of bitumen ageing. This absence of bitumen stiffness at low temperatures will be very important for the application of the bitumen in asphalt mixtures and for attaining the required low-temperature durability.

5. Conclusions

The following conclusion were formulated based on the test results:

- the ageing process caused a slight decrease in dynamic viscosity,
- the level of the hardening index considerably increased at a 0.6% fatty amine content,
- the long-term ageing process had a minor effect on the stiffening of polymer modified bitumen with FA additive, despite low temperature and regardless of fatty amine content,
- in polymer modified bitumen PMB 45/80-65, the fatty amine acts as an inhibitor of the bitumen binder ageing process.

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