There is more to say about fatigue and healing of Polymer Modified Asphalt

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Digital Object Identifier (DOI): dx.doi.org/10.14311/EE.2016.025

ABSTRACT

The list of extensive studies on asphalt fatigue and associated healing aspects is long and impressive. In general there seems to be consensus on a few topics: in nearly all tests carried out a moderate to significant fatigue resistance improvement is observed when mechanisms that allow healing are incorporated in the test procedure, while at a similar level of confidence it has been shown that SBS-modified asphalt shows better fatigue resistance than their unmodified equivalents.

However, where the consensus ends and the confusion starts is when it comes to healing aspects with Polymer Modified Asphalt. Negative effects on the healing factors for straight asphalt mixtures and/or binders have been observed with harder bitumen, lower temperatures and ageing. Hence a correlation with viscosity and thus ease of flow, wetting and diffusion is plausible and often postulated. So it is counter-intuitive that numerous researchers have found positive effects on healing with Polymer Modified Asphalt. Compared to their unmodified base binders they do not have a lower viscosity at the prevailing temperatures and diffusion is rather hampered than improved by the high molecular weight species. For some such an outcome is so counterintuitive that they deny these results and that may lead to an unnecessary thick pavement design in cases where a lower pavement thickness could be implemented. For example the Dutch road authorities use a factor of 4 for unmodified asphalt and use a factor of 1 for PMA, unless proven otherwise.

This paper will provide information on the potential mechanism behind improved healing performance up and above the improved fatigue characteristics for SBS-modified asphalt.

Keywords: Durability, Fatigue Cracking, Healing, Modified Binders, Polymers

1. INTRODUCTION

The desire for materials to restore their consistency after tearing or cracking by a healing process has generated quite some research and potential solutions. In the polymer industry new approaches like click chemistry and micro-encapsulated healing agents have already made inroads and based on these developments so-called 'self-healing' developments have also started in the asphalt industry. Focus in these latest investigations is on how to restore cracked asphalt pavements and potential solutions have been found in adding iron (wool) to asphalt and reheat it after cracking through induction. However, this particular form of healing is different from what traditionally has been addressed when using this term: the ability of an asphalt mix to retard crack propagation in fatigue testing by the introduction of rest periods. The latter is an important parameter to be taken into account in the design of pavements. In principle the two phenomena captured under the same name do not only require different test methods, but have also polluted the discussion as conclusions that were drawn for one phenomenon have erroneously been declared valid for the other phenomenon and vice versa.

Another fundamental question is: are we going to design roads that are prone to cracking, but we build in something that should allow these cracks to be healed at some point in time, or are we going to build roads that will have no or minimal cracking over the foreseeable future. In this paper we will focus on cracking prevention and in that context we will also focus on the healing aspects of SBS-modified binders and will provide a plausible explanation why these binders can lead to better healing factors than their unmodified counterparts.

2. BACKGROUND

2.1 Traffic loading pattern

Contrary to what is normally done in fatigue testing, the pavement has an intermittent loading pattern. Even the distance between the wheels is significantly bigger than the tire contact let alone the distance between two trucks. Hence when one looks at a test method simulating practice closest, it should be fatigue testing with intermittent loading, while the load-free period should be long enough to mimic what happens on the road. As an average one could advocate a load-free period of 25 times the load, but a 9 : 1 ratio also seems reasonable to provide good indications. Opting for fatigue tests with unloaded storage periods is in that respect more remote from practice, while fracture healing tests are not relevant for design purposes: macro cracks have already risen, which should have been avoided.

2.2 Literature

There is no point in reiterating on the huge number of papers that have been published on this topic, but we can refer to the excellent overview given in Qiu's thesis [1] and highlight from his section on binder effects on healing. He found positive results on PMB healing reported by Bahia [2] (on binders), Lee [3] and Carpenter [4] on asphalt mixtures. More recent work from Sutharsan (2010) [5] fully confirms these findings. They were all applying intermittent loading. Kim found little influence from SBS modification on healing [6] (so no negative effect) and Little even a negative effect [7], but they had used the less relevant storage method.

Hence vs. the four positive and one neutral observation, there was one that reported a negative effect of polymer modification, but that one used the fatigue healing with storage method.

Qiu himself then executed tests of which the test results varied. In a ductility specimen test a slower, but more complete strength recovery was found for the SBS-modified binder. In mastic testing (also strength recovery after complete breaking), the SBS-modified binder was the same at 10 °C, slower at 20 °C and faster at 40 °C. The explanation for the 20 °C result was found in the higher viscosity of the PMB hampering the wetting and diffusion, but no explanation was given for the better performance at 40 °C, while the viscosity at that temperature would also be higher.

In the loading-healing-reloading test carried out in a displacement controlled tension test, the PMB recovered the strength as quickly as the unmodified binder, but also a more complete recovery up to displacement levels of 2 mm. At 3 mm the PMB performed worse, but this may in asphalt terms already be considered as a macro-crack.

When looking into the effect of other parameters that are affecting the healing rate there appears to be consensus on the following: harder bitumen, lower temperatures and ageing have a negative effect. They all point into the same and plausible direction: viscosity dominates the healing process. However, already in 1999 Phillips executed some model experiments using a DSR in which he found strong indications that viscosity does not affect the rate at which healing takes place. He used wax to gel the bitumen and SBS to increase the viscosity and found no effect and a positive effect on healing rate respectively [8].

3. EXPERIMENTAL

3.1 Zero Shear Viscosity

In the next section reference is made to Zero Shear Viscosity (ZSV) data. The ZSV is determined using a method proposed by Phillips and Robertus [9]. In brief it is a repetition of low stress creep and relaxation cycles at the temperature of choice. The method provides valuable information on the viscosity, but is not useful outside the scope of research as one result takes about 3 to 4 days to be generated.

3.2 Binder morphology by microscopy

There are various ways to observe the morphology of PMBs, but the method that was used was chosen to allow demonstration of the mechanism. Hence we allowed the binder to grow to a coarse two-phase dispersion at a suitable temperature between two glass plates, after which a photo was made. Then the sample was cooled at 1 °C per minute and then a second picture was taken at 120 °C. We are aware of the imperfection of this method, due to the more or less two-dimensional limitation, but freeze fracturing would bring other limitations, which would hamper studying the basic mechanism.

4. MORPHOLOGY ASPECTS ON BITUMEN/SBS BLENDS

Morphology aspects related to SBS modified bitumen are fairly well understood and documented [10, 11]. After complete dissolution of the SBS into the bitumen there is one of either morphology at temperatures around 160 - 180 °C: a single phase system or a two-phase system with one phase rich in asphaltenes and one rich in SBS. The single phase system will turn into a two-phase structure upon cooling. As early as 1989 Vonk and Bull presented a paper on this matter based on roofing compounds, in which they also characterized the separate phases after hot centrifugation [12].

To bring this somewhat closer to paving binders, we have examined binders based on two different bitumens, one leading to a single phase at 160 $^{\circ}$ C (bitumen B) and the other to a two phase system (bitumen A; no compatibilization reactions done). The latter was centrifuged at 160 $^{\circ}$ C and in order to get a two-phase structure those with the highly compatible bitumen were done at 100 $^{\circ}$ C. Centrifugation was continued until full separation had been achieved. The binders and the separate phases were then subjected to Zero Shear Viscosity testing at 40 $^{\circ}$ C. The results for the base bitumen, the blends and separate phases are shown in Figure 1.



Figure 1: Zero Shear viscosity at 40 °C for base bitumen, blends and their separate phases

The weight fractions for the separated phases were 15/85 and 25/75 for the 3% and 5% blends from bitumen A and 17/83 and 26/74 for the 3% and 5% blends from bitumen B resp. It should be kept in mind that the centrifugation temperatures were 160 and 100 °C resp. and the similarity was thus pretty accidental. The two important observations from these tests are:

1. Not only is the blend viscosity higher than that of the base bitumen, but also those of the separate phases.

2. The blend viscosity of the blend with 5% SBS in bitumen B appears to be closer to the viscosity of the Polymer Rich Phase (PRP) than with the other bitumen, indicating blend based on bitumen B to be closer to Polymer Rich Phase continuity than the blend based on bitumen A.

These observations are not surprising as the dissolving process is generally as follows: the polymer absorbs oily components from the bitumen and the asphaltenes tend to hold on to the remainder of the maltenes. So the PRP is highly concentrated in oily components (like an oil extended polymer) and so soft and elastic, while the concentration of the asphaltenes in the remainder is higher than in the base bitumen and thus higher in viscosity. One needs to take care with the PRP viscosities as in the ZSV test there is never a linear deformation against time, but it shows an estimate of how high the viscosity is or -in other words- how limited the flow will be.

With the base bitumens and the 5% SBS-modified binders we produced dense asphaltic concrete and subjected asphalt beams to fatigue tests. The tests were carried out under constant stress at 10 °C in three point bending. The latter is not optimum, but all beams were tested in the same way and thus comparable. The test frequency was fairly high (40 Hz), but that was chosen to speed up the test. The intermittent loading was done with one load cycle and 5 rest cycles. In the usual way the fatigue curves were determined at various stress levels and the comparison of the healing factor was determined for each of the systems at the stress level at which 10^6 cycles were achieved.



Figure 2: Healing factors generated on dense asphalt mix beams in 3-point bending fatigue

From the data presented in Figure 2 it is quite clear that the observed healing factors are not in-line with the normally observed trends of better healing at lower viscosities/higher temperatures; Figure 1 shows the comparison of Zero Shear Viscosities at 40 °C and although there are no data available of ZSVs at 10 °C there is no reason to believe that the relative data would be significantly different.

Now there are different lessons to learn; first of all the cracking or rather crack propagation. Any bitumen / polymer blend consists at service temperatures of a two-phase structure with a relatively brittle phase rich in asphaltenes and a soft and elastic phase consisting of the polymer and maltenes. In reference [12] it has been shown that there is hardly any asphaltenes in the PRP and no polymer in the Asphaltenes Rich Phase. At lower SBS concentrations, at which the PRP is definitely not the continuous phase, a morphology is present that is best compared with so-called rubber toughened crystalline polymers, such as in polystyrene or in systems such as epoxies. There are numerous publications that have demonstrated not only the positive effect of a rubber dispersed phase in such systems on things such as impact resistance

and other forms of cracking, but also showed the importance of particle size and thus the morphology. This then also explains at least that there are differences in the effectiveness of SBS modification on fatigue resistance in bituminous binders. However, the effect is always there: cracks tend to propagate through the brittle phase and through various mechanisms they are retarded by the dispersed soft elastic phase.

A dramatic step in crack resistance is achieved when the PRP does become the continuous phase. A good example of the step change in fatigue resistance is the recent development of highly modified asphalt [13]. These morphology effects explain the improved fatigue resistance of SBS modified asphalt, but at the same time, taking into account the above consideration, do not explain any positive effect on the healing that has been observed in so many investigations. A plausible explanation is described below.

5. THE POSSIBLE MECHANISM TO EXPLAIN IMPROVED HEALING FOR SBS-MODIFIED ASPHALT

Vonk and Bull [12] not only separated the phases with the centrifuge, but also examined the effect of the temperature at which the blend was centrifuged. At all temperatures the time of centrifugation was chosen such that the separation was complete. Figure 3 shows that the mass fraction of PRP decreases with decreasing temperature. This is no surprise as it is in-line with theory.



Figure 3: The effect of centrifugation temperature on the mass fraction of PRP [12]

As a result of the decreasing PRP mass fraction, the ARP gets less concentrated and should thus become less brittle. This is illustrated in Figure 4 where the penetration values of the separated ARP after centrifugation at different temperatures are shown.



Figure 4: The effect of centrifugation temperature on the pen value of the ARP [12]

However, another observation was made at the same time: upon cooling of a two-phase structure, new darker colored islands appear in the PRP, next to some increase of the ARP islands already present. As it had been shown that nearly the entire asphaltenes fraction has been concentrated in the ARP, the material that is released by the PRP are oily or maltenes components that are lower in viscosity than the original binder, as this is free of asphaltenes matter (Figure 5). During the centrifugation process this third phase is forced into the original ARP, which thus increases in mass fraction, but at the same time also increases in pen value.



160 °C

120 °C

Figure 5: Mechanism for increased phase separation upon cooling of bitumen/SBS binder [12]

The third phase islands of lower viscosity material may thus form a naturally arising healing agent like it has been developed by White et al. [14] (Figure 6). They introduced microcapsules with a brittle shell, which upon cracking released a healing agent that reacted with another agent to form a polymeric healing material. In the case of SBS-modified bitumen no reaction is required to take care of the healing, while the highly viscous PRP assures a natural encapsulation. Upon crack development in the PRP it is likely to rupture the third phase islands and heal the crack.



Figure 6: Autonomic self-healing polymer concept by White et al. [14]

6. CONCLUDING REMARKS

The resistance to fatigue cracking of a pavement structure is one of the most important parameters in pavement thickness design. There are still a lot of investigations going on whether asphalt mix fatigue can be derived from a (fairly simple) binder test. Asphalt fatigue testing is laborious and time consuming. In order to reduce testing time to acceptable levels, fatigue is tested in a continuous mode, although one knows that this is not a realistic simulation of actual practice, while it is also recognized that rest periods lead to a significant increase in fatigue life.

In numerous publications healing has been dealt with and was not seldom related to viscosity parameters. It was thus counter-intuitive to expect that, although SBS modification of the bitumen leads to improvement of the fatigue resistance, it would also add to the healing: as a result of modification the viscosity increases which should lead to less healing. However, any polymer modified bitumen tends to consist of a two phase structure at service temperatures, but the mass fraction ratio of the separate phases is dependent on the temperature. As an asphalt mix is produced and applied at much higher temperatures than the normal service temperatures it is more than likely that oily material is released from the Polymer Rich Phase upon cooling. This released oily materials will partly be absorbed by the existing second phase rich in asphaltenes, but partly exist as an independent third phase of low viscosity material, If that third phase is affected by a crack, the oily material is released into crack which can thus heal the crack easily. This is a proposed mechanism based on morphology changes and associated (separate) phase properties that have been observed outside of the asphalt mix, but there is little reason to believe that the basic processes will be different inside the asphalt mix. It may be a challenge for future fundamental studies to prove whether this supposed mechanism is valid or not.

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