Aging Characteristics of Polymer Modified Binders in Porous Asphalt Pavements

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ABSTRACT: This paper deals with the aging characteristics of SBS polymer modified binders in porous asphalt pavements. Field cores were taken from test roads of different ages and structurally analyzed using X-ray tomography. Binders were extracted from the upper and under layers of the field-aged cores and characterized using gel permeation chromatography, Fourier transform infrared spectroscopy, dynamic shear rheometer, as well as conventional penetration test. The chemical and mechanical characterization was also carried out on the binders recovered from loose mixes, and on the binders aged in laboratory by RTFOT and PAV. The study indicated that the structure of porous asphalts can be visualized using X-ray tomography. The distributions of air voids, mortars and stone materials were displayed over the pavement depth; however, for an accurate quantification of the mix components, the boundary conditions must be validated. It was found that at the earlier time of service (< 2 years), the polymer modified binders aged quite uniformly over the pavement depth. With increasing years of service, the modified binders in the upper layer were more aged than those in the under layer even though both layers consist of high air void contents. Degradation of the polymer was found to occur mainly during asphalt production and during earlier stage of pavement service. The polymer degradation may compensate for the oxidative hardening of bitumen components, thus mitigating overall age-hardening of the binders. Rheological evaluation showed that, after four years in the porous asphalt pavement, the modified binders continue to perform well. The study also implied that a proper selection of base bitumen is of importance in achieving long-term durability of a modified binder in porous asphalt. As regards laboratory aging tests, field aging prediction by PAV was found to vary substantially, depending on air void content in the asphalt and asphalt position (layer) in the pavement.

KEY WORDS: Porous asphalt pavement, polymer modified bitumen, aging characterisation, x-ray tomography.

1 INTRODUCTION

In recent years, increased demands for noise reduction and safety improvement have been strong driving forces for developing durable bituminous materials in porous asphalt pavements. Porous asphalt consists of high interconnected air voids (> 15%). In a wearing course, porous asphalt is permeable and water on road surface is minimized under wet conditions. Consequently, driving safety is improved significantly. Porous asphalt also has been proven effective with respect of noise reduction (Gibbs et al. 2005). Typically, the magnitude of noise reduction by using porous asphalt is around 3 dB(A) as compared with conventional dense asphalt concrete (EAPA 2007).

Because of a high amount of air voids in porous asphalt, aging resistance of the binder becomes crucial. Void content in asphalt mix determines the rate of aging by controlling oxygen access to the binder. Higher air voids would facilitate the oxidative aging of the binder even deeper in asphalt pavement. Aging makes bituminous materials harder and more brittle, thus increasing risk of pavement failure, such as ravelling and cracking (Hagos et al. 2007). To minimize the failures, polymer modified binders (PMBs) are often recommended for porous asphalts in Europe (Nielsen, 2006). Polymer modified binders may have better aging properties as compared to unmodified binders. They are also believed to increase the lifetime of porous asphalt by increasing cohesion and adhesion in the mixes and by increasing binder film thickness without risking segregation of the material.

To simulate bitumen aging in laboratory, various tests are currently used, including RTFOT (EN 12607-1) for simulation of the aging at high temperature during e.g. asphalt production, and PAV (Pressure Aging Vessel, EN 14769) for the aging at ambient temperature in the field. For unmodified bitumen, these tests normally give reasonable prediction. For polymer modified binders, particularly when applied in porous asphalt, relevance of the tests must be evaluated.

The main objective of this paper is to study the aging characteristics of styrene-butadiene-styrene (SBS) modified binders in porous asphalt during asphalt production and in asphalt pavement. A variety of test methods were chosen to characterize field mix samples and binders, physically or chemically. For evaluating prediction of the field aging in porous asphalt, laboratory tests of PAV at different conditions were performed.

2 MATERIALS, FIELD SAMPLIES AND TEST METHODS

Polymer modified binders containing 6% SBS were studied. Penetration and softening point of the modified binders are typically 95 dmm and 80°C. According to Superpav binder specification, the modified binders are classified as PG70-34.

The modified binders were used in various projects of porous asphalt pavement in Sweden, including test sections in Stockholm area on highways E4 and E18, and road Rv 260. The test sections were in-service under similar climatic conditions. Field cores were drilled from several sites of the test sections: Site 1 on Rv260, Site 2 and Site 3 on E4, and Site 4 and Site 5 on E18. The field cores consist of two layers (30 mm upper layer and 50 mm under layer), which was identified by a visual inspection, as well as by X-ray tomography (see details later). Two loose mixes produced for the upper and under layers on E4 were also sampled from the job site for binder recovery and analysis. As a reference, a stored sample of pure modified binder was characterized.

For the recovery of binders from asphalt mix samples, a standard procedure similar to EN 12697-3 was followed. The method uses dichloromethane as a solvent to extract binders from mixes and uses a rotary evaporator to remove the solvent at the end. In order to obtain sufficient amount of samples, for each site several cores were used for binder extraction. The binder samples obtained are summarized in Table 1 along with results of penetration measurements.

A variety of laboratory test methods were selected. X-ray tomography was used for the structural characterization of field cores, and gel permeation chromatography (GPC), Fourier transform infrared spectroscopy - Attenuated total reflection (FTIR-ATR) and dynamic shear rheometer (DSR) used for binder tests. To simulate aging of binders, RTFOT and PAV were chosen. PAV tests were conducted at 100°C, and also at a lower temperature of 75°C which is believed to be more relevant in terms of prediction of the field aging, particularly for polymer modified binders.

Table 1: The modified binder samples extracted from loose mixes and field cores

Sample codes	Description	Years in the field	Penetration, 1/mm
R-M1	Loose mix 1	0	77
R-M2	Loose mix 2	0	74
R1-UP	Upper layer on Site 1	<1	57
R1-UN	Under layer on Site 1	<1	61
R2-UP	Upper layer on Site 2	2	45
R2-UN	Under layer on Site 2	2	49
R3-UP	Upper layer on Site 3	2	32
R3-UN	Under layer on Site 3	2	53
R4-UP	Upper layer on Site 4	4	31
R4-UN	Under layer on Site 4	4	46
R5-UP	Upper layer on Site 5	4	25
R5-UN	Under layer on Site 5	4	40

3 THE X-RAY TOMOGRAPHY OF FIELD CORES

As a non-destructive test, X-ray tomography was used to visualize the internal structure of asphalt field cores. Tests were carried out using a medical computed tomography scanner (CT-scanner, Siemens SOMATOM Plus4 Volume Zoom). The CT-scanner consists of an X-ray source and a row of detectors which are rotating around the sample and measure the attenuation of the X-rays of the sample from different angles. The degree of attenuation depends on the density of the material and is measured in Hounsfield Units (HU). The CT-scanner used has a maximum horizontal resolution of 0.293x0.293 and a vertical resolution of 1.0 mm. In the tests, field cores of 100 mm in diameter were scanned with a slice thickness of 1 mm from the upper layer to the under layer.

The typical CT images of the upper layer and the under layer of an asphalt core are shown in Figure 1. The dark fractions represent air voids and the brightest fractions are stone materials. Due to the limited resolution mentioned above, it was hard to detect individual elements which are smaller than 2 mm. Therefore, divisions were made between air voids, mortar (mixture of bitumen and fine particles of less than 2 mm), and stones (> 2 mm). The boundary conditions used for such divisions are 250 Hounsfield Units (HU) between voids and mortars, and 1500 HU between mortars and stones. Selection of the boundaries was based on Delft’s experiences obtained on porous asphalts and stone mastic asphalts (Remijn 2005). The computed volume percentages of air voids, mortars and aggregates over the height of the sample are exemplified in Figure 1.

As shown in Figure 1, the field core consists of two layers of porous asphalt (or double-layer porous asphalt), and the upper layer and the under layer differ considerably in aggregate gradation. Obviously, the aggregates used in the upper layer are much finer as compared to those used in the under layer. In each layer, distribution of air voids over the height is quite uniform except for the interaction zone (probably interlayer) between the two layers. It is found that, for the under layer mix, the amount of air voids estimated by the X-ray tomography is fairly similar to that measured by the conventional standard method (FAS Metod 448). Examples of air void comparison are shown in Figure 2 for Site 2 and Site 3. For the upper layer samples, however, the air voids measured by the X-ray tomography are significantly lower than those obtained by the standard method. The difference is probably attributed to that, in the upper layer, fine particles had partly clogged air voids and the clogged air voids were erroneously measured as mortars by the X-ray tomography.

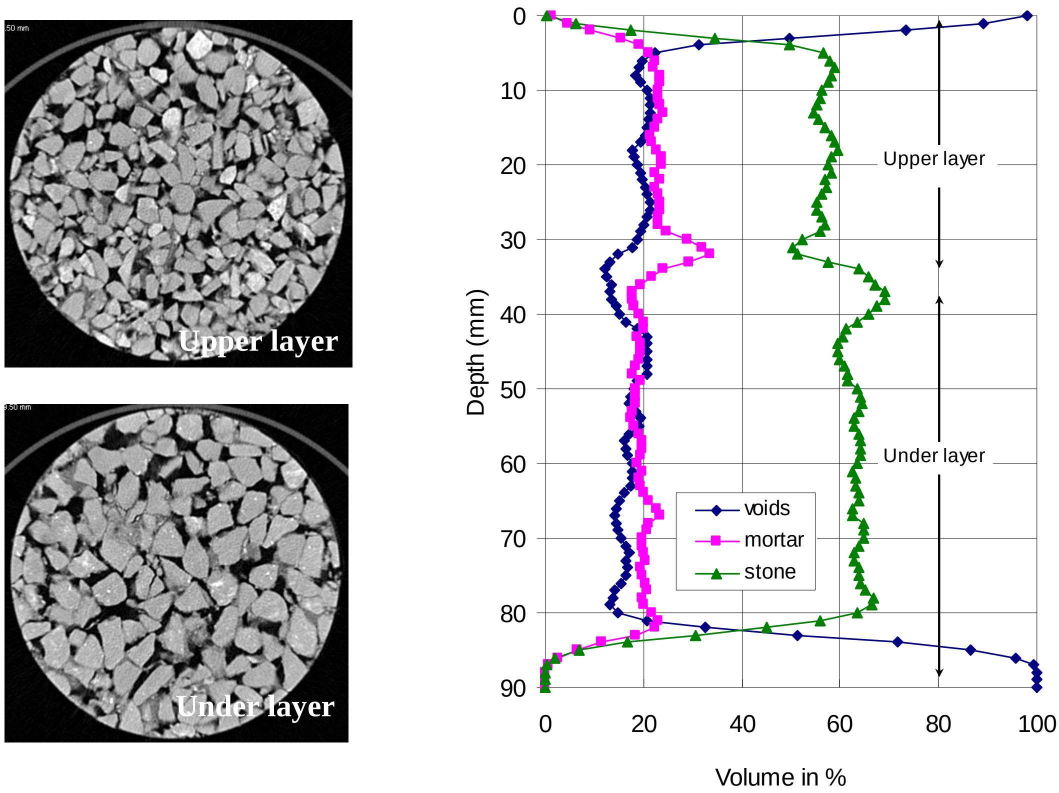


Figure 1: X-ray tomography analysis of a field core

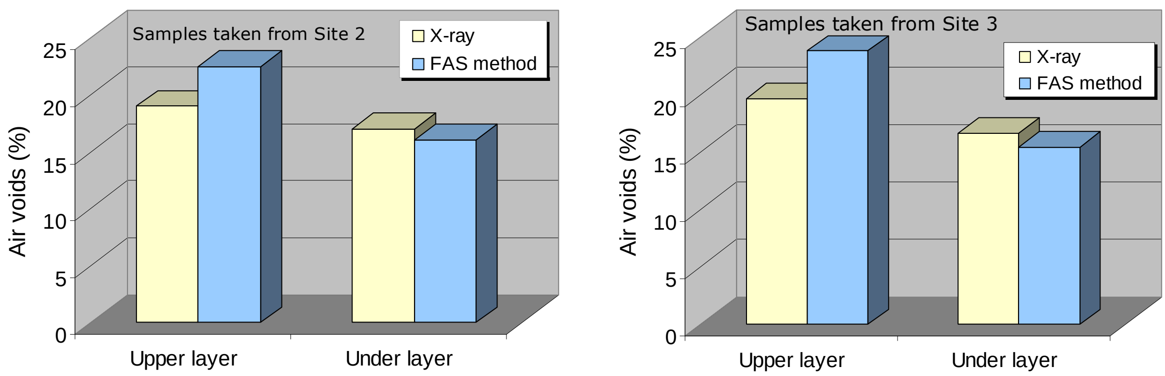


Figure 2: Comparison of air void contents measured by X-ray tomography and FAS method

The clogging of air voids in porous asphalt pavement can be a problem for retaining a sufficient noise reduction over time in the countries like Sweden, where the use of stud tyres is permitted in winter. For the field cores studied here, it is not sure if any clean measure was made before their sampling. Moreover, the boundary condition defined between air voids and mortars in the X-ray tomography is somewhat arbitrary, and need to be further verified. This is beyond the scope of the present study.

3 AGING OF POLYMER MODIFIED BINDERS IN THE FIELD

Various tests were applied to characterize field aged modified binders. In Table 1 presented earlier, results of penetration test are summarized. Accordingly, retained penetrations are calculated and compared in Figure 3. A lower value of retained penetration means a higher degree of age-hardening. As expected, all the recovered binders display age-hardening over the service time. The binders in the upper layer were more aged than those in the under layer even though both layers consist of high amount of air voids. It is also indicated that the age-hardening of the modified binders mostly occurs during the mixing in asphalt plant and asphalt transportation, as well as during earlier time of service on the road. The short period of time has decreased penetration of the binder by almost 40%, as illustrated by the samples recovered from the loose mixes and from Site 1. The big decrease in penetration is probably attributed to the use of relatively soft base bitumen in the modified binder.

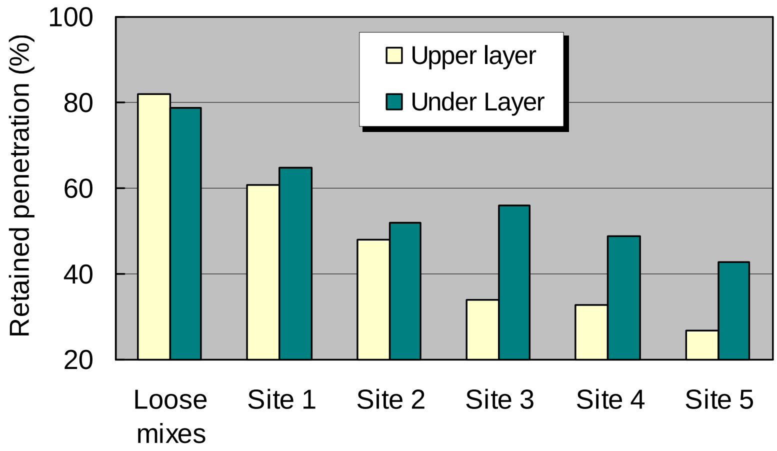


Figure 3: Retained penetrations of the recovered binder samples

To further characterize ageing-induced rheological changes, DSR measurements were carried out at 10 rad/s and from 10 to 100°C. The rheometer used was Physica MCR 501, Anton Paar. Examples of DSR tests are shown in Figure 4. Due to a combined effect of bitumen oxidation and polymer degradation, changes in complex modulus and phase angle do not appear to be "parallel" shift, which is well known for unmodified bitumen after aging. Apparently, degradation of the polymer may negatively influence elasticity of the modified binders as indicated by increased phase angles in a temperature range of 40 to 90°C for the recovered binders.

For a quantitative evaluation, rutting and fatigue parameters are determined according to the SHRP binder specification. As can be seen from Figure 5, the field aged binders satisfy rutting criterion ($G^*/\sin\delta \geq 2.2$ kPa) at 64°C and far exceed fatigue requirement ($G^* \sin\delta \leq 5000$ kPa) at 22°C. Even though low temperature measurements by a bending beam rheometer (BBR) or direct tension tester (DTT) have not been performed due to limited amount of samples, the field aged binders are classified at least as PG64-28 based on tests made on similar modified binders. A binder of PG64-28 is sufficient for application in the climatic conditions where the test sections were built. According to weather data reported in "www.weatherpage.se", in Stockholm and since 2000, the maximum five-day average temperature was 24°C (July 15-19, 2003), and the minimum five-day temperature was -12.4°C (January 1-5, 2003). Above observations suggest that the modified binders will continue to perform well in the porous asphalt pavements from the rheological point of view.

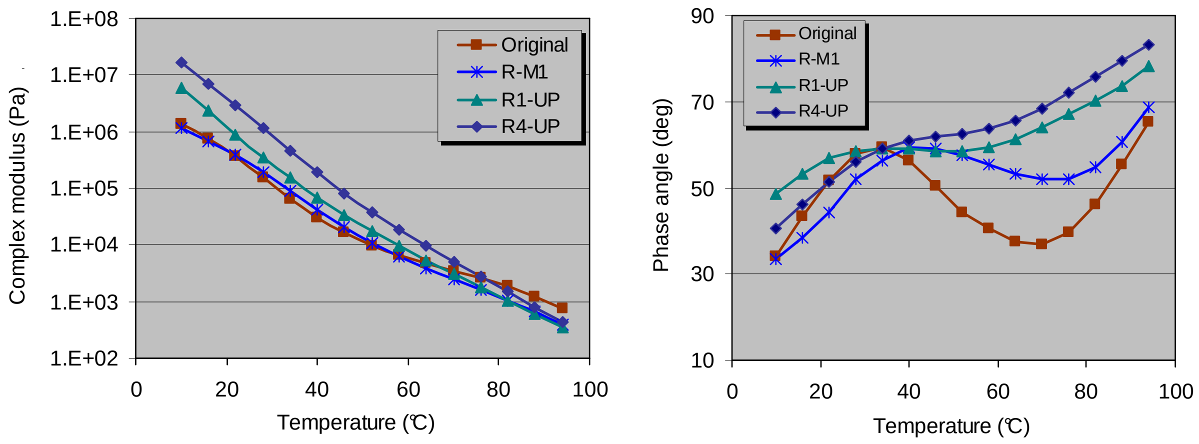


Figure 4: Examples of complex modulus and phase angle as a function of temperature

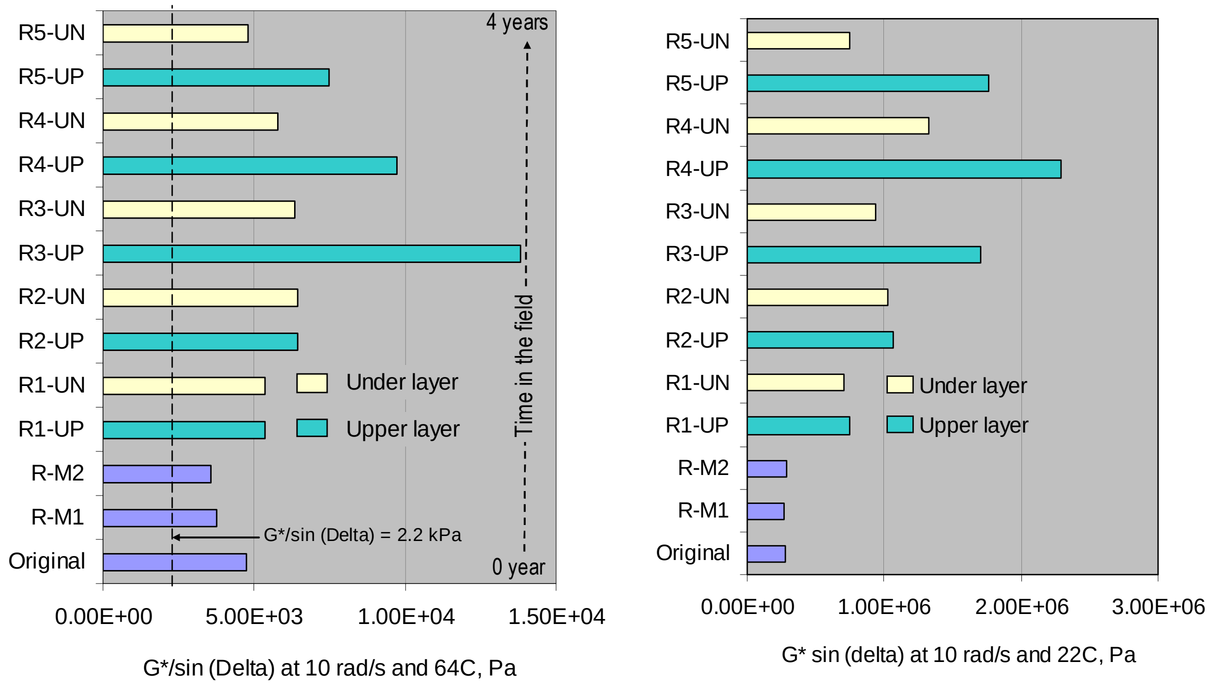
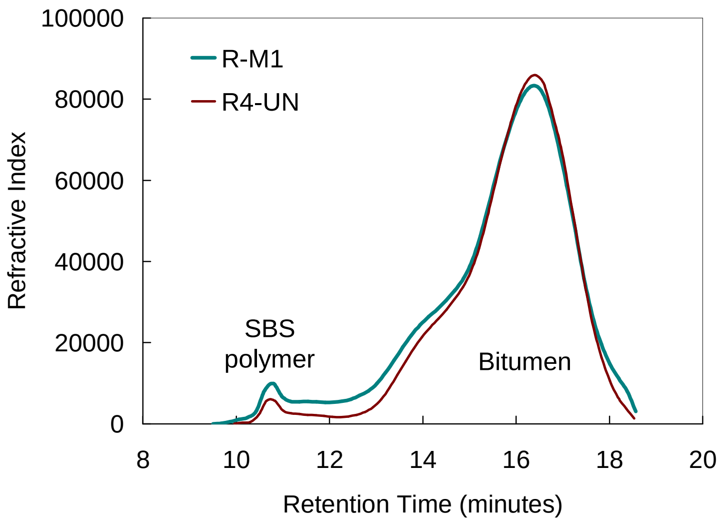


Figure 5: Rheological comparison according to the SHRP rutting and fatigue parameters

Figure 5 also shows that the overall stiffing effect is not proportional to the time of the binder in the field, suggesting simultaneous effects of various factors. On earlier service, there is almost no rheological difference between the upper layer and under layer samples. With increasing time in the field, the upper layer samples become stiffer than those from the under layer. Compared to binder samples extracted from the upper layers, the under layer samples show much less variations in the rheological properties. This appears to be consistent with the observations on air voids of the field samples. For example, the content of air voids in the upper layer mixes decreases by more than 10%-units as the service time is doubled from 2 years in Site 3 to 4 years in Site 5; however, for the under layer mixes, much less change is found in air voids (< 5%-units by FAS method, and < 1%-unit by X-ray tomography). As aging mechanism of SBS modified binders may be thermal oxidation and/or photo-oxidation, part of the differences observed between the upper layer and under layer samples could also be attributed to the variation of temperature over the pavement depth and the effect of ultraviolet light (Durrieu et al. 2007).

The degradation of SBS polymer is easily shown by GPC, which is a common technique to fractionate mixtures based on molecular size. In this study, an equipment Alliance 2690 Separation Module (Waters) was used. Sample solutions (0.4%) were prepared with tetrahydrofuran (THF). GPC chromatograms are exemplified in Figure 6 along with the weight average molecular weights (M_w) measured for the fractions of polymer and bitumen. By using an internal calibration procedure, polymer contents in the recovered binders were estimated. The estimation was made within a molecular weight range (100,000 – 300,000) similar to that of the polymer in the original unaged sample. This means that certain fragments of the degraded polymer are not measured although their molecular size can be larger as compared to bitumen components.



Samples	M_w – Bitumen	M_w – Polymer
Original	1630	178000
R-M1	2010	142000
R-M2	2190	146000
R1-UP	2290	155000
R1-UN	2050	154000
R2-UP	2120	161000
R2-UN	2300	160000
R3-UP	2100	156000
R3-UN	2290	162000
R4-UP	2210	157000
R4-UN	2060	155000
R5-UP	2350	158000
R5-UN	2070	167000

Figure 6: Examples of GPC chromatograms and the measured molecular weights

As indicated in Figure 7, degradation of the polymer mainly occurs during asphalt production and during the earlier service time of the pavement. Afterwards, while the aging of bitumen components continues with time, the polymer does not change very much in concentration. This has also been confirmed by FTIR-ATR, which showed that SBS concentrations estimated by using a ratio of butadiene signal at 966 cm^{-1} to bitumen signal at 1450 cm^{-1} only differed between 2.6% - 3.0% for the samples of two or four years in the field.

However, degradation of the polymer does not mean disappearance of the beneficial effect of adding the polymer. In fact, polymer degradation may compensate for the oxidative age-hardening of bitumen components, thus retaining desirable rheological properties for the modified binders over service time of the pavement. Such advantage has been demonstrated by the rutting and fatigue property measurements. It is believed that, for a long-life porous asphalt pavement, a proper selection of base bitumen is also of great importance to ensure durability of the modified binder.

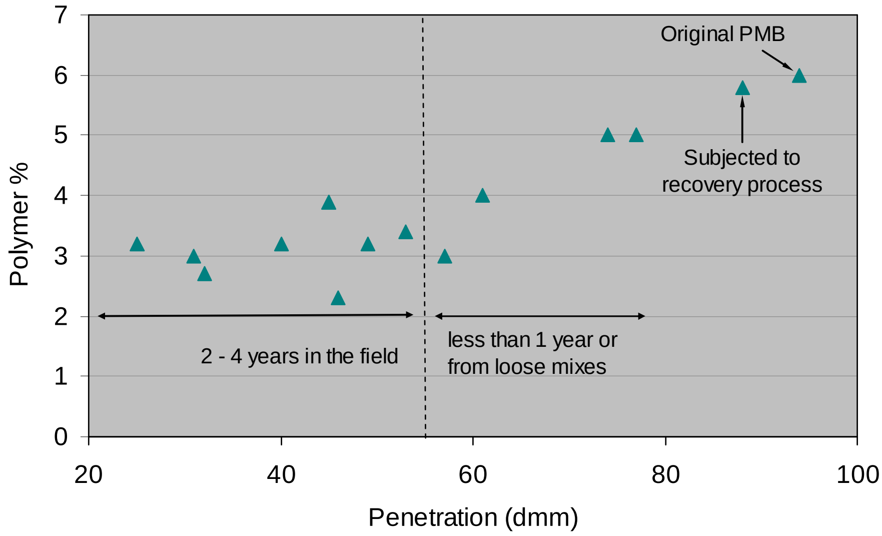


Figure 7: Polymer contents estimated by GPC *versus* penetration of the binders

4 AGING OF POLYMER MODIFIED BINDERS SIMULATED IN LABORATORY

The short-term aging of the same modified binder as in the test roads was simulated by RTFOT. It is compared with the actual aging occurred after the plant mixing, mixture storage and transportation. DSR test along with GPC measurements of polymer content and penetration test are shown in Figure 8. Besides the lab aged sample and those recovered from the loose mixes (R-M1 and R-M2) taken from the job site, the original modified binder and the one treated by the recovery process are shown as references. Although GPC does not show difference in polymer contents, the RTFOT sample appears to be more aged than those recovered from the loose mixes. The larger difference observed in the region of 10^3 – 10^4 Pa complex modulus is probably attributed to less polymer networks in the RTFOT sample.

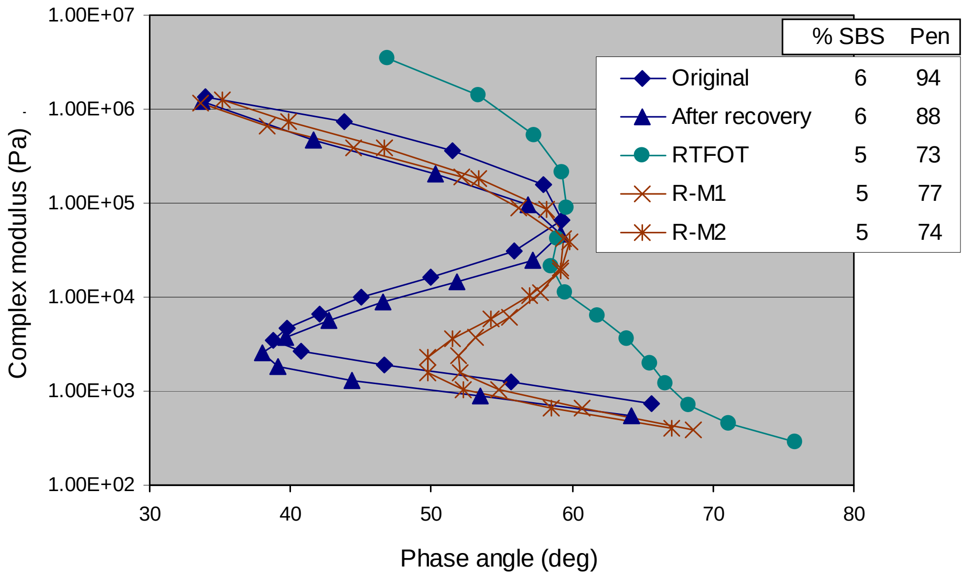


Figure 8: Complex modulus *versus* phase angle comparing the short term aging

In Figure 9, the long-term aging simulated by PAV on a similar modified binder at two different temperatures and the field aging on porous asphalt pavements are compared. The

PAV tests were conducted on the binders after RTFOT. In the figure, complex modulus (G*) is measured at 60°C and 10 rad/s, and the G* ratios are calculated by dividing G* of the PAV aged or recovered samples by G* of the original modified binder.

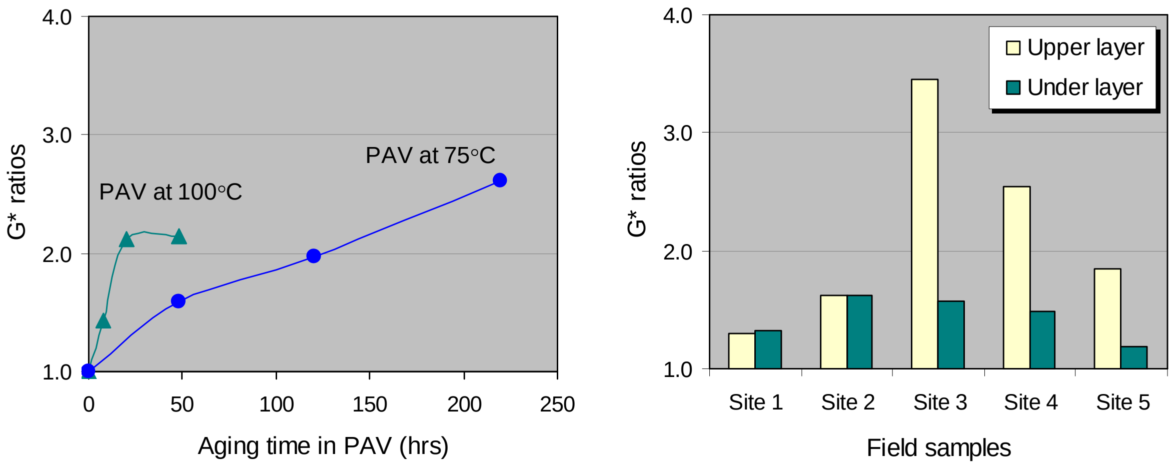


Figure 9: Comparison of the long-term ageing test PAV and the field aging

It can be seen that PAV can significantly under-predict field aging of the binders on the upper porous asphalt layer if PAV at 100°C and 20h or at 75°C for 140h is assumed for simulating 5 – 10 years aging in the field. On the other hand, it become more severe when used to predict the aging that occurs on the under layer.

To further study the relevance of PAV, the polymer modified binders used in other mix types are compared in Figure 10. The retained penetrations were calculated based on the data reported by VTI (Jacobson and Hornwall 1999, 2000). Evidently, prediction of the field aging by PAV is strongly dependent on air voids of the mixes. For the mix ABD16 of 15% starting air voids, the severe age-hardening of 10-year in the field is not predictable by PAV, neither at 100°C nor 75°C. On the other hand, in the mix ABS16 of 3% or 5% air voids, the extent of binder age-hardening is low, which can't be properly predicted by running PAV test.

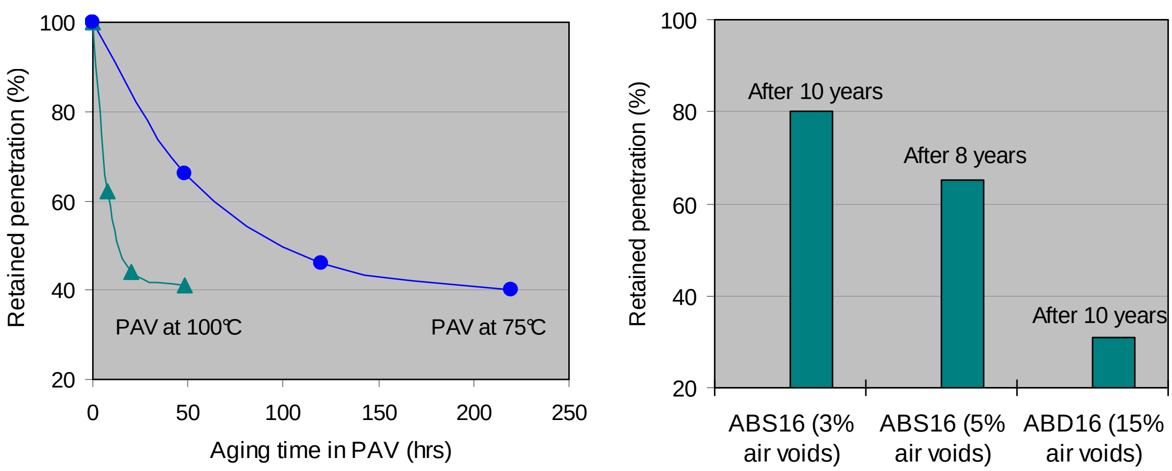


Figure 10: Comparison of PAV and the field aging of other mixes

5 CONCLUSIONS

The following conclusions can be drawn from the present study:

The structure of porous asphalts is visualized using X-ray tomography; the distributions of air voids, mortars and stone materials are displayed over the pavement depth. However, for an accurate quantification of mix components, the boundary conditions have to be validated.

At the earlier time of service (< 2 years), the polymer modified binders in porous asphalts aged uniformly over the pavement depth. With increasing years of service, the modified binders in the upper layer are more aged than those in the under layer even though both layers consist of high air void contents.

Degradation of SBS polymer mainly occurs during asphalt production and during earlier time of pavement service. The polymer degradation does not mean disappearance of beneficial effect of adding the polymer. The rheological evaluation shows that, after four years in the porous asphalt pavements, the modified binders will continue to perform well. The study also implies the importance of a proper selection of base bitumen to achieving a long-term durability of the modified binder in porous asphalt.

Aging of the modified binders in the field is not properly predicted by PAV. The test significantly under-predicts or overestimates the field aging, depending on air void content in the asphalt and asphalt position (layer) in the pavement.

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