LONG LASTING ASPHALT PAVEMENTS AND BITUMEN AGEING

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ABSTRACT

Long lasting asphalt pavements are particularly desirable to ensure the efficiency of road transport. To find out key factors that determine the lifetime of an asphalt pavement, an extensive study was carried out on a number of long lasting road sections with known construction data and performance track records. Asphalt cores were drilled from the selected road sections and investigated by means of mechanical tests and non-destructive X-ray computed tomography. The field samples were further examined with respect to mixture composition, and binder chemistry and rheology. The study showed that long lasting pavements consist of thick asphalt layers with higher binder contents and low air voids. In general, the lower asphalt layer was stiffer than the upper layer. Although small local cracks were seen inside some cores, there was no propagated cracking. In all the studied road sections, bitumen ageing was found to be slow, as assessed either by a stiffening effect or by chemical compositional changes. The slow rate of bitumen ageing had kept asphalt layers flexible enough to resist cracking, which has been confirmed by the analyses using HMA fracture mechanics, as well as field performance observation. It was also shown that a tight or dense upper layer may prevent bitumen ageing in the underlying pavement. The present study highlights the importance of binder durability (resistance to ageing) in achieving a long lifetime asphalt road.

Keywords: Asphalt pavement, durability, mix characteristics, non-destructive testing, bitumen ageing, chemical analysis, rheology

1. INTRODUCTION

In developing a sustainable road transportation system, long-term durability of the pavement is of great importance. This has been achieved to some extent in well designed and constructed thick flexible pavements, where classic bottom-up fatigue cracking and structural deformation did not appear as the main cause of failure, in these cases, it was ageing-related surface deterioration (such as cracking and raveling, etc.) that determined the lifetime of the roads. In principle, a strong pavement structure of long-lasting performance (> 50 years) may be obtained by proper materials selection [1, 2], optimal mix and pavement design [3], and fundamental performance testing and monitoring, while the surface of the pavement is replaced periodically, approximately every 20 years. Such a concept of long-life or perpetual pavements has been practiced for many years in Europe, as well as in the United States [3-6]. It has a number of benefits [7] including eliminating costly reconstruction at the end of the structural life, lower user delay costs for rehabilitation activities, lower consumption of non-renewable resources, reduced energy cost over the life of the pavement, and lower life-cycle cost of the pavement system.

Undoubtedly the lifetime durability of asphalt pavements is affected by many factors. In order to fully understand key contributors, road pavements with known construction data and performance track records are needed. In Denmark, as well as in Sweden, road authorities and contractors have over a long period of time constructed test roads to study the performance of different types of asphalt. A catalogue of those test roads was prepared. The catalogue contains about 170 Danish test sections, which were built in the period of 1980 – 2004 and with the intention to examine various bituminous materials and paving technologies. The majority (about 70%) of the recorded test sections were built in the 1980's. From the old test sections, more than 40 well-documented sections were selected for a desktop study. While many of these test sections were overlaid due to severe fretting, raveling, surface cracking, or extensive patching, several sections were still surviving with satisfactory performance after a long time in-service (> 20 years). The purpose of this paper is to characterize the bituminous materials used in the long lasting road sections, particularly with respect to field ageing and its impact.

2. TEST SECTIONS AND FIELD SAMPLING

Four test sections (coded 2a, 4d, 4e, and 4f) located on Danish primary roads were studied. The sections were constructed in the early 1980's on the existing old roads [8, 9], one on Hldv 119 (primary highway 119) and three on Hldv 411 (primary highway 411). Pavement structures are illustrated in Figure 1. Field sampling was done in July 2006. For each test section, 21 asphalt cores (100 mm in diameter) were drilled from different positions (see Figure 1), seven from the wheel path or under track, seven between the tracks, and seven near the road shoulder line. Heights of the obtained cores varied with the test sections, from the surface down to at least the whole thickness of the old wearing course AC d12 (see Figure 1).

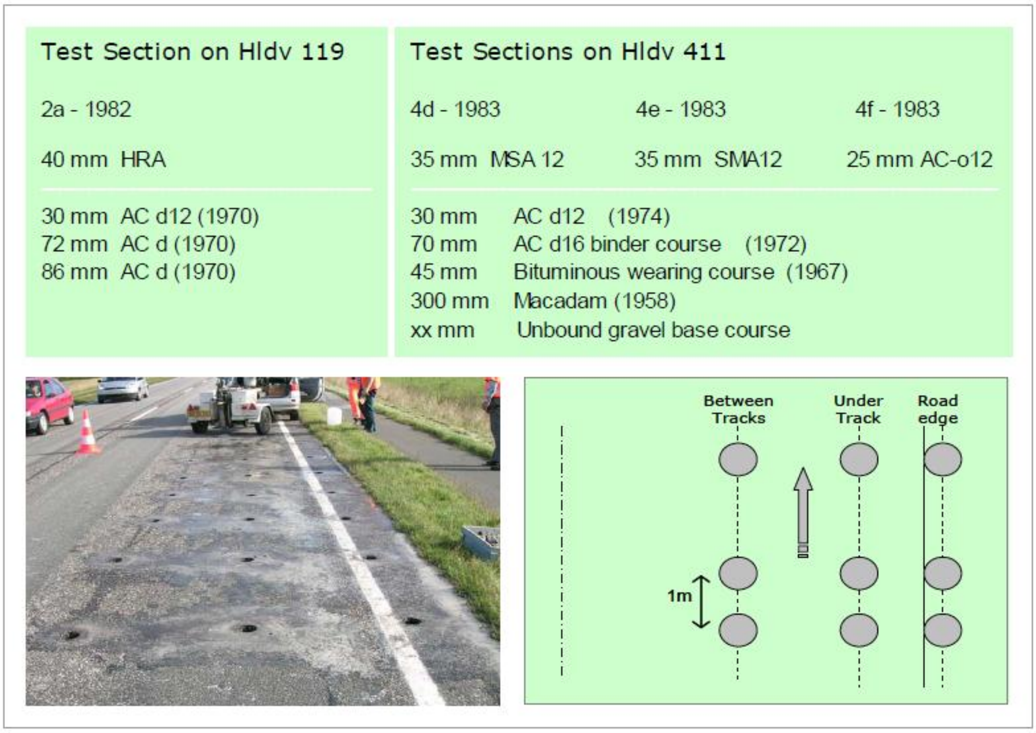

Figure 1: Pavement structures of the selected test sections and illustration of field sampling

Hldv 119 is a four lane highway with ADT of 4429 and ESAL of 609 reported in 2004. The purpose of the test sections on Hldv 119 (Skovvejen) was to investigate the potential of other types of asphalt wearing courses as compared to dense asphalt concrete (AC d). AC d was a dominant type of surfacing used on major Danish highways at that time; however, it had shown problems with low durability. In the beginning, the trial consisted of open graded asphalt concrete (AC o, 2 sections), AC-o modified with Chemcrete modifier (1 section), soft asphalt (1 section), stone mastic asphalt (SMA, 1 section), hot rolled asphalt (HRA, 1 section) and sand asphalt modified with Chemcrete (1 section). The test sections were constructed in 1982, and visual inspections were carried out regularly. Damages were also recorded in BELMAN, a pavement management system normally used to support decisions concerning re-surfacing of bituminous pavements on primary and secondary roads in Denmark. At the time of field sampling (July 2006), only the HRA section built with plain B60 bitumen was surviving. Damages reported in 2005 for this section are shown in Table 1. During the field sampling in 2006, it was observed that the section had significant stone loss, and also started to crack on its shoulders.

The purpose of the test sections on Hldv 411 was to investigate the performance of various asphalt surfaces which were expected to be durable under heavy traffics. Hldv 411 is a two lane highway, and ADT/ESAL in 1988, 1995, and 2004 were 8000/1000, 7700/1400, and 8659/1786, respectively. The trial consisted of AC d with EVA modified binders (2 sections), MSA – a special designed low void AC d (2 sections), AC o (1 section), HRA (1 section), and SMA (1 section). The test sections were constructed in 1983. The surviving sections in 2006 were: 4d – MSA 12 (this is a special designed low void AC-d12) with plain B60 bitumen, 4e – SMA 12 with plain B60 bitumen, and 4f – AC o12 with plain B100 bitumen. Damages reported in 2004 for these sections are shown in Table 1. It was reported that in spring 2005 test section 4d showed limited cracking, raveling and fretting, while sections 4e and 4f showed limited cracking but significant raveling and fretting. All the three sections showed rutting, however, were repaired by leveling (micro asphalt surfacing) in 1999. At the time of field sampling in 2006, a lot of patching (including rut filling) was seen on section 4d, but test sections 4e and 4f looked good even though rut repair was also noticed.

Table 1: BELMAN damage report and field inspection on the test sections

Test sections	2a - HRA	4d - MSA12	4e - SMA12	4f - ACo12
Bitumen type	B60	B60	B60	B100
Damages report 2004/2005				
Raveling, %	25	7	5	5
Cracking, m	--	300	150	150
Allig. Cracking, %	--	1	0	1
Patching, %	17	84	51	46
Inspection 2006	Stone loss	A lot of patching	Rut repair	Rut repair

3. TESTS ON ASPHALT CORES

Field cores were first visually inspected, and possible surface damages, such as loss of stones and rut filling, were recorded. By a visual inspection, identification of different asphalt layers was also made. The field cores were then investigated by means of X-ray tomography and indirect tensile test (IDT).

3.1 X-ray tomography analysis

The computed tomography scanner (CT-scanner) used in this study was a Siemens SOMATOM Plus 4 medical scanner of horizontal and vertical resolutions of 0.293x0.293 and 1.0 mm, respectively. The CT-scanner consists of an X-ray source and a row of detectors. In asphalt testing, X-ray source and detectors are rotating around the sample and attenuation of X-ray is measured from different angles. The computer then calculates the attenuation of a certain volume element (Voxel) inside the asphalt sample, and results are recorded in 2D images. The degree of attenuation depends on the density of the material and is measured in Hounsfield Units (HU). For compacted asphalts, distributions of air voids, mortar and stone materials may be estimated using certain boundary conditions between these different components. Based on the experiences obtained on porous asphalts and stone mastic asphalts, the boundary conditions were chosen as: HU < 250 for air voids, 250 < HU < 1550 for mortar, and HU > 1550 for stones. Due to the maximum spatial 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 < 2 mm), and stones (> 2 mm). A typical slice from an open graded asphalt is shown in Figure 2. The dark fractions represent air voids, the brightest fractions are aggregates, followed by the fraction between representing mortar. Using the selected boundaries, the volume percentages of air voids, mortar and aggregates are calculated.

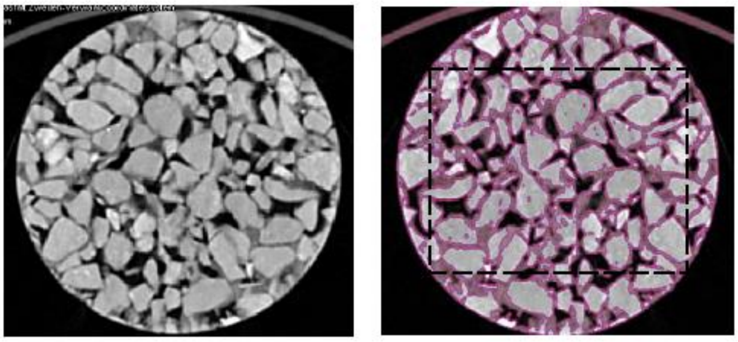

Figure 2: X-ray tomography image analysis for an open graded asphalt core. The square area is used for volume calculations of air voids, mortar and aggregates

Typical results of the X-ray tomography analysis are shown in Figure 3 for a core drilled from the test section 4d. It was found that all the asphalt cores taken from the four long lasting test sections show low air void contents, even for those from section 4f which was intended to test open graded asphalt concrete (Figure 4). For a given test section, samples taken from different positions differ in air void contents, and generally in the order of Under Track < Between Tracks < Road Edge. For some cores, the measured air void contents were also enlarged by crack damages inside the sample. This is the case for the sample in Figure 3, which reveals slightly high air void contents in the top of the leveling course. However, those cracks were very local, and did not propagate through the asphalt layer.

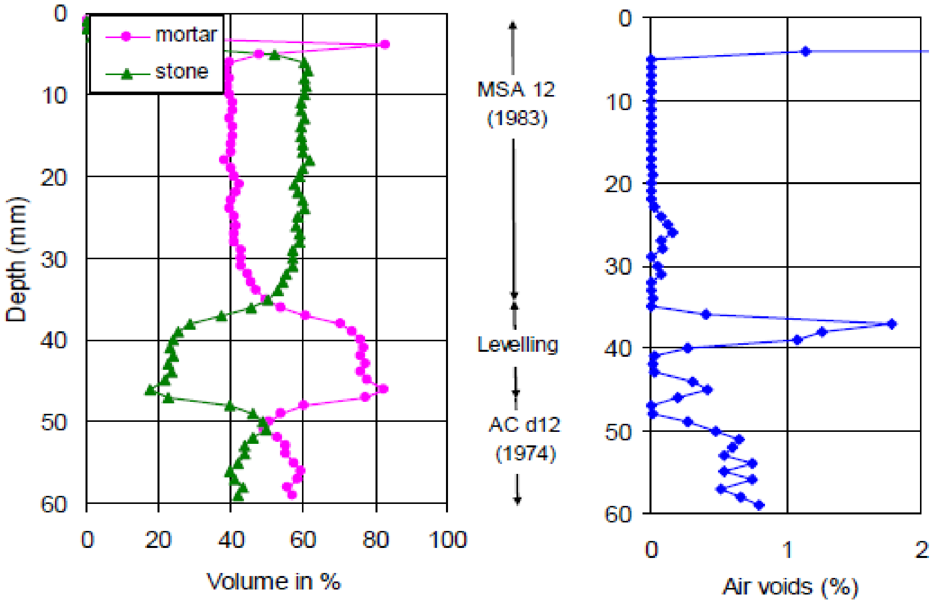

Figure 3: Distributions of air voids, mortar and aggregate in a field core taken from test section 4d

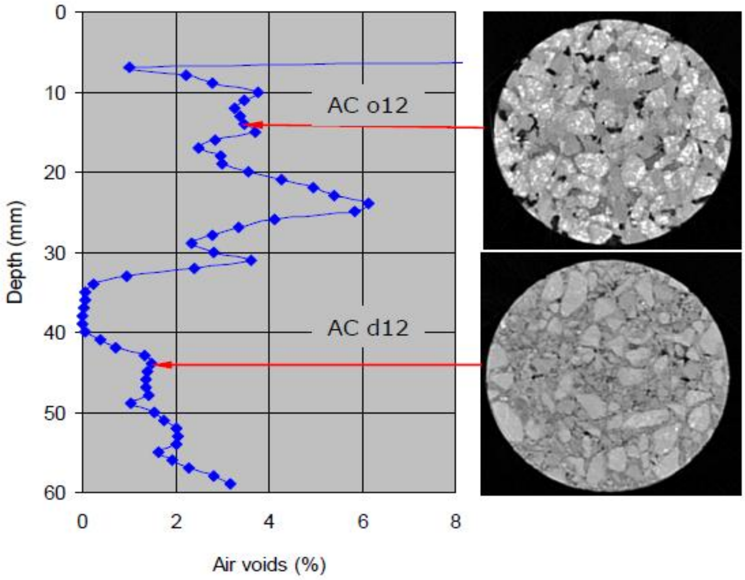

Figure 4: Air voids distribution and CT images for an asphalt core taken from test section 4f

By X-ray tomography, the different asphalt layers of field cores may be identified. As illustrated in Figure 3, the sample taken from the test section 4d consists of the wearing course MSA12, which was constructed in 1983, and the old wearing course AC d12 built in 1974. Between the two asphalt courses, a thin layer of significantly higher volume of mortar can be seen. This is probably a leveling or bond coating made in conjunction with the constructing of the test section. This type of thin layer was observed for all the studied test sections. In addition, for the wearing courses tested on sections 4d, 4e and 4f, the thickness estimated by X-ray tomography was found to be close to the designed one. However, the thickness of HRA on test section 2a had been diminished considerably (15 – 28 mm for the field cores as compared to the designed thickness of 40 mm), not only for the cores taken from the wheel path, also for those from other positions.

3.2 Mechanical properties

In IDT, the asphalt stiffness modulus was measured at different temperatures (10, 20 and 30°C) according to the procedure described in EN 12697-26 annex C. Prior to the stiffness test, asphalt cores were treated by cutting off the top and bottom parts. The trimmed cores were further sawed into individual layers. Stiffness measurements were carried out for the four wearing courses (HRA, MSA12, SMA12, and AC o12), as well as for the old wearing course AC d12. A series of tentative tests on laboratory compacted asphalts indicated that thickness of the specimens should be at least 28 mm to ensure precision of the measurements (The EN standard test method specifies a minimum thickness of 30 mm). Thus, no tests were made on the HRA taken from test section 2a, as all samples had a thickness of less than 20 mm.

Figure 5 shows that for test section 4e with SMA12, the under-track samples are significantly stiffer than those taken from the road edge and between-tracks, while for test sections 4d (4f as well), the stiffness of the wearing courses seems not to vary with the sampling positions. Variations in asphalt stiffness may be attributed to the air void contents or to extent of ageing. Air void contents may change differently due to varying compaction under traffics, and differences in air voids will also result in different ageing rates (see data and discussion in next section).

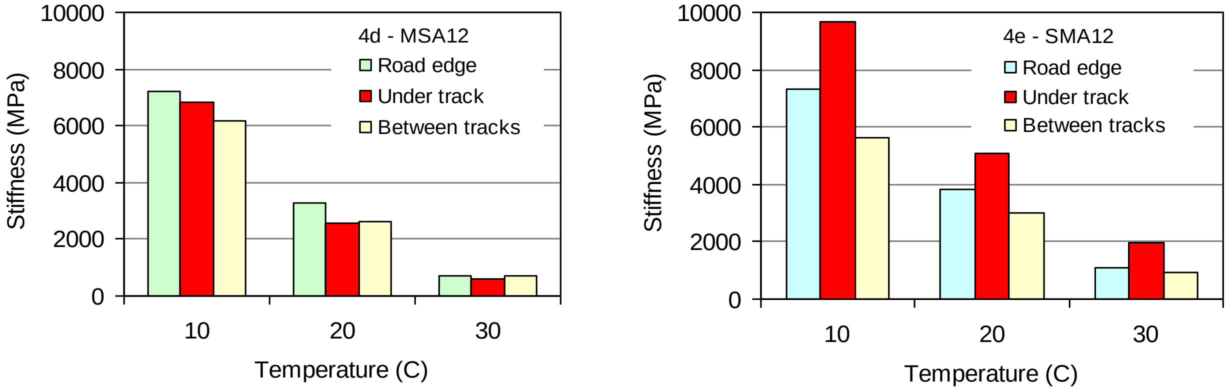

Figure 5: Stiffness of asphalt wearing courses from different sampling positions

As for the old asphalt layers built in the 1970's, large differences in stiffness are observed. For example, for the same type of asphalt AC d12 and on the same highway (Hldv 411) shown in Figure 6, the samples covered by the open graded asphalt AC-o are much stiffer than those under the dense asphalt MSA. This is again due to the binder which has aged differently under the two different surfaces (slower ageing in 4d as compared to in 4f). The observation also suggests that a dense surface layer may prevent ageing of the binder in the under layer by reducing the availability of oxygen to the binder.

Moreover, the old asphalt under-layers in general are found to be stiffer than the wearing courses tested. For example, on test section 4d, the old underlying asphalt AC d12 is about 30% stiffer than MSA12 when tested at 20˚C. This has provided good support to the surface layer of the pavement, thus improving the resistance to surface cracking. At the same time, the old stiff asphalts, which acted as binder courses, may further prevent permanent deformation in the pavement.

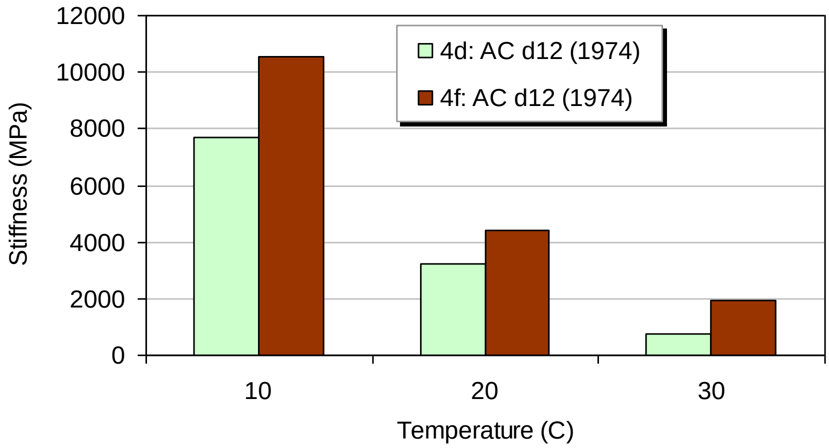

Figure 6: Stiffness measurements of the old wearing course AC d12 from the two different test sections

The mechanical properties of the field cores were further characterized using SuperPave IDT at 10˚C. The measured parameters include resilient modulus (instantaneous MRT, and total MRT), creep compliance parameters (D0, D1, and m), failure strain and tensile strength (St) [10]. As shown in Table 2, the studied wearing courses in 2a, 4d and 4e exhibit relatively high creep rate, indicating low ageing of the materials. Based on HMA (Hot Mix Asphalt) fracture mechanics framework, the fracture resistance of the asphalts was evaluated. In Table 2, the dissipated creep strain energy to failure ($DCSE_f$) is a measurement of how much damage a mixture can tolerate before micro-crack initiates, while the energy ratio (ER) measures a relative proportion between the damage thresholds of the mixtures and the rate of damage accumulation due to loading. Generally higher ER indicates better fracture resistance, with ER = 1 being a threshold between good and poor field performance. As shown in the table, all the wearing courses tested except for 2a-HRA have ER higher than 1, suggesting good field performance in terms of cracking resistance. The predictions correlate quite well with field observations.

Table 2: Mechanical parameters obtained from IDT and HMA fracture mechanics

Test sections	2a – HMA	4d – MSA12	4e – SMA12	4f – AC o12
M_{RI}, GPa	10.7	13.7	10.4	6.5
M_{RT}, GPa	8.0	10.4	8.7	5.5
D_0	0.09	0.07	0.10	0.15
D_1	0.16	0.20	0.08	0.15
m	0.51	0.58	0.45	0.28
Failure strain	691	2879	1989	767
St, MPa	0.86	2.28	2.09	1.34
FE, KJ/m^3	0.63	4.11	3.17	0.67
$DCSE_f$	0.52	3.86	2.90	0.50
ER	0.26	1.02	3.66	1.58

4. BINDER RECOVERY AND CHARACTERIZATION

For binder recovery, a procedure as described in EN 12697-3 was followed. The method uses dichloromethane as a solvent to extract binders from asphalts and uses a rotary evaporator to remove the solvent at the end. The recovered binders were weighted and binder contents in the mixtures were determined (EN 12697-1). In addition, the air void contents of the field samples were determined in accordance with EN 12697-8, and particle size distribution analyzed according to EN 12692-2. The recovered binders were tested by penetration and dynamic shear rheometer (DSR, Physica MCR 501, Anton Paar), and the results obtained for the wearing courses are shown in Table 3.

As can be seen, all the tested wearing courses contain relatively high binder contents except for AC o12 which has a binder content slightly below 6%. The relatively lower binder contents in AC o12 is probably due to segregation of the binder during transport and laying as the open asphalt concrete did not use stabilizing fibres in that time (1983). For the old asphalt layer AC d12 under different test sections, binder contents were found between 5 to 6%. Table 3 also shows that most of the field cores consist of lower air voids (< 5%), even for some samples taken from the section 4f which was originally designed as open graded asphalt concrete. Comparing the samples from different positions, a general trend in air voids is (as expected): Under Track < Between Tracks < Road Edge. As will be discussed later, the lower air void of the asphalt mixtures to some extent has retarded bitumen ageing in the field.

In Figure 7, the black diagrams (complex modulus *versus* phase angle) obtained by DSR tests are shown for the binders recovered from the different wearing courses. As a reference, unaged bitumen of 50/70 type is also shown. It can be seen that the black diagrams are quite smooth and all the field aged samples follow a similar tendency in the complex modulus – phase angle function. At the same stiffness level, the field aged samples are, as expected, more elastic (lower phase angle) as compared with the unaged bitumen.

It is known that, for unaged conventional bitumens, there is a good relationship between penetration and complex modulus measured at 10 rad/s and 25˚C [11]. Such relationship also exists for the field aged bitumens; for all the recovered binders (21 in total), correlation coefficient (R^2) was found to be 0.96. Knowing such correlation is helpful, as the amount of recovered binders is not always sufficient to make penetration test, and at the same time, this kind of empirical data is often needed for tracking and evaluating the binders in the old pavements.

Table 3: Analyses of asphalt cores and recovered binders

Test sections	Sample position	Asphalt Cores		Recovered Binders	
		Binder, %	Air voids, %	Pen, dmm	Vis. @ 60C, Pa s
2a – HRA	Under track	6.3	0.4	29	1270
	Between tracks	6.6	0.6	29	1030
4d – MSA12	Under track	6.3	0.4	43	838
	Between tracks	6.4	0.3	35	905
	Road edge	5.4	0.6	35	1010
4e – SMA12	Under track	6.9	3.6	17	3590
	Between tracks	6.6	2.4	26	1310
	Road edge	6.6	3.8	21	2520
4f – AC o12	Under track	5.8	5.4	8	12800
	Between tracks	5.4	4.9	15	2900
	Road edge	5.0	9.7	5	19400

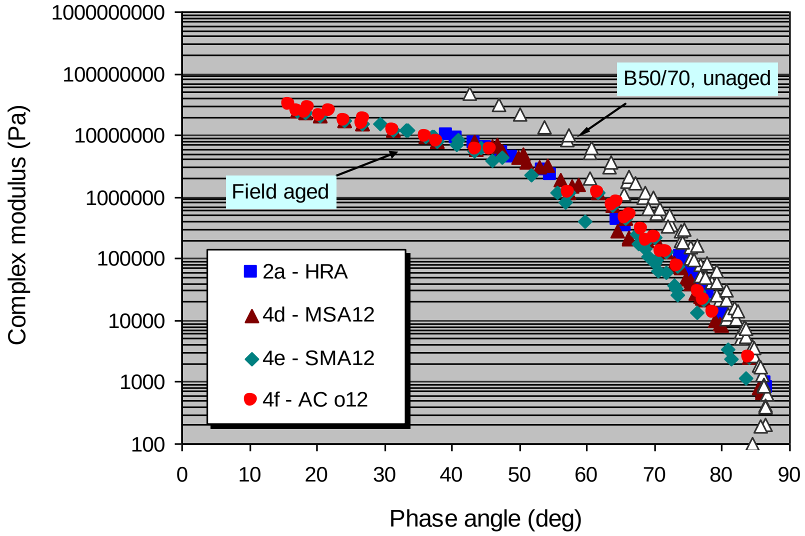

Figure 7: Complex modulus versus phase angle for the binders recovered from the different wearing courses

In chemical characterization, Fourier transform infrared spectroscopy with attenuated total reflectance (FTIR-ATR) and gel permeation chromatography (GPC) were used. Typical examples of IR spectra with absorbance assignments are shown in Figure 8. Apparently, the recovered binders differ in the range of 1200 – 900 cm^{-1}, where the absorbance at around 1030 cm^{-1} is assigned to sulfoxides (S=O). Differences are also seen for carbonyl compounds (C=O) at about 1700 cm^{-1}, as well as for aromatic carbons at about 1600 cm^{-1}, as illustrated in Figure 9. The compositional changes in terms of carbonyl compounds and sulfoxides are commonly used to assess the oxidation sensitivity of bitumen.

As for GPC, the recovered binders show similar chromatographic pattern (Figure 10), except for some samples from test section 4e, which display two distinct peaks, indicating that the bitumen probably was blended.

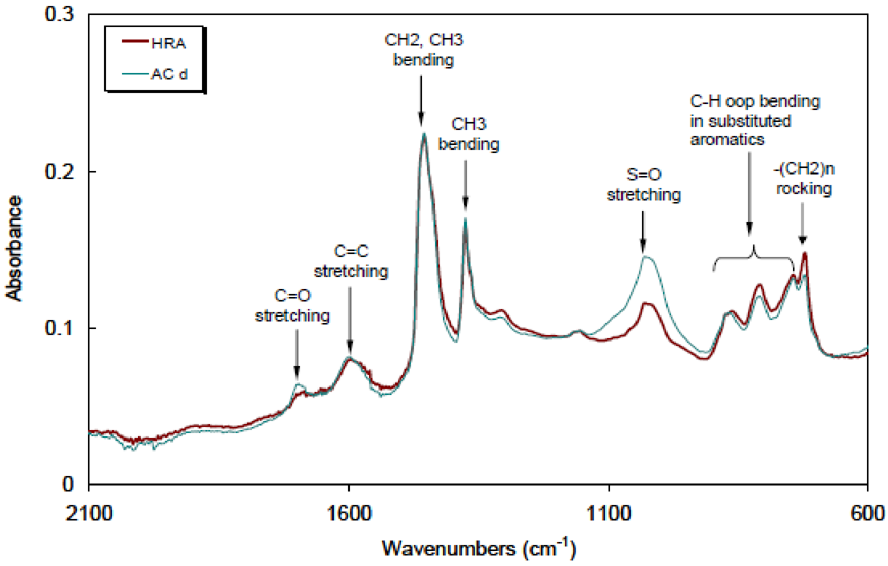

Figure 8: Examples of IR spectra for the binders recovered from test section 2a

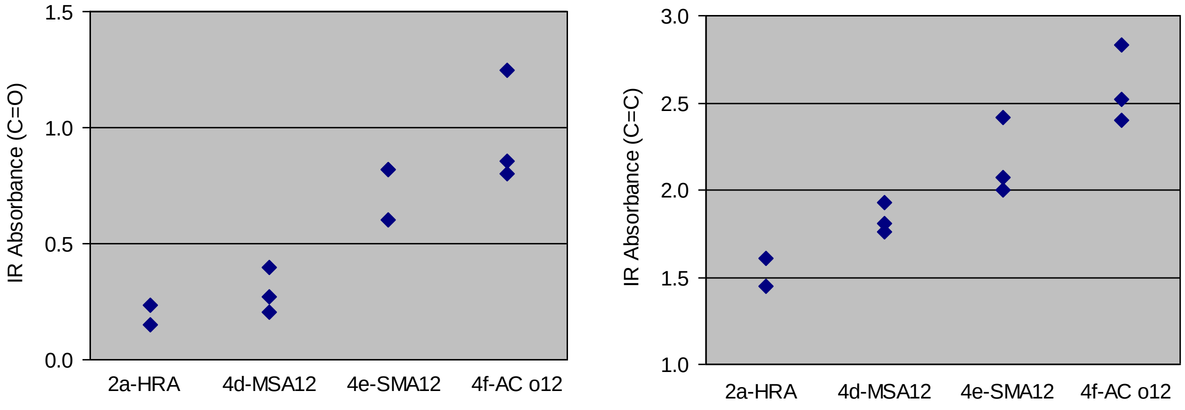

Figure 9: IR absorbance of carbonyl compounds and aromatic carbons for the binders recovered from the different wearing courses and at different sampling positions

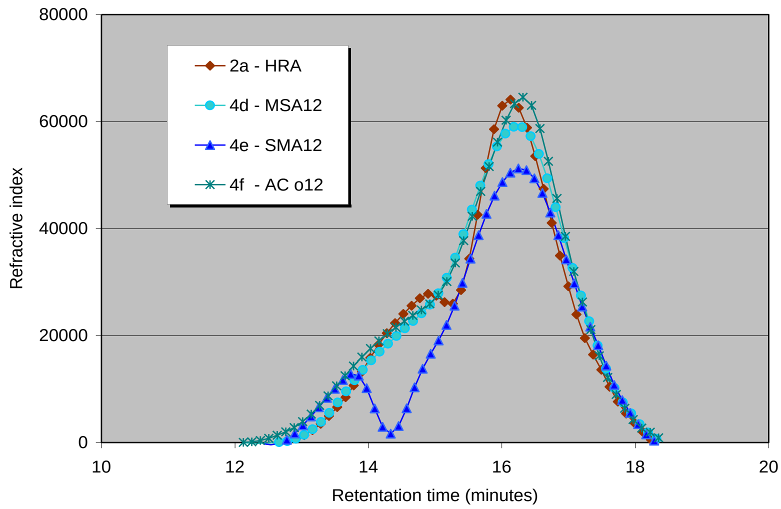

Figure 10: GPC chromatograms of the binders recovered from the different wearing courses

In general, the rheological property changes of a binder in asphalt pavements are attributed to the oxidation-induced compositional changes [12]. It was found that complex viscosity (logarithm) tends to increase with molecular weight and carbonyl compounds with correlation coefficients R^2 of 0.68 and 0.47, respectively. However, such tendency is very weak or non-existent when sulfoxides are considered ($R^2 < 0.1$). This is not surprising, since relationships between the rheological and chemical parameters can vary with the type and origin of the bitumen, and the ageing-induced viscosity increase could also be attributed to increased aromaticity or aromatization that enhances molecular interactions between the molecules.

5. AGEING OF BITUMEN IN THE FIELD

As already shown, the asphalt mixtures used in the test sections consist of low air voids. The low air void contents had retarded the extent of bitumen ageing in the field. For the bitumen used in HRA on test section 2a, penetration of the bitumen decreased by only 14 dmm after almost 25 years on the road (0.6 dmm per year). In the old dense asphalt layers AC d and AC d12, the corresponding change in penetration is as small as 0.3 dmm per year during 1981 and 2006 (Figure 11).

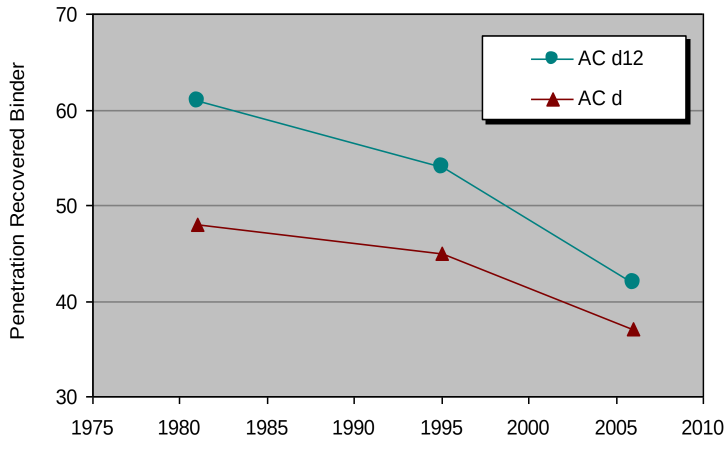

Figure 11: Penetration of the binders recovered from the underlaying old mixtures on test section 2a

The effect of air voids on the ageing of bitumen is further shown in Figure 12. In Figure 12, the degree of bitumen ageing is expressed as a retained penetration, which is the penetration of the recovered bitumen divided by the penetration of the original unaged bitumen. A higher value of this parameter means slower ageing of the bitumen. For each type of mixture, the three points shown in the figure represent the samples taken from different positions. Evidently, ageing of the bitumen in the field is extremely slow when air void content is below about 3%. This is in agreement with observation reported in the literature [13].

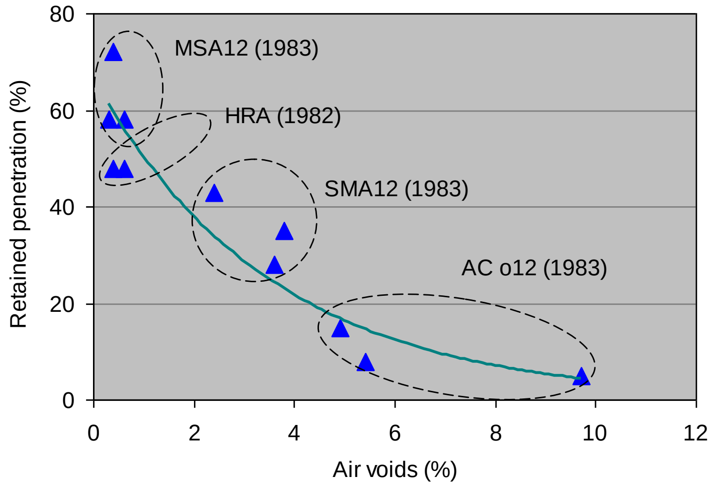

Figure 12: Ageing of bitumen in the field versus air void content of the asphalt mixtures

The facilitating effect of air voids on bitumen ageing is also reflected by the compositional changes. In Figure 13, the relative amounts of carbonyl compounds as measured by IR absorbance are shown along with complex viscosity for the binders extracted from the asphalt cores having different air void contents. As implied, for given bitumen, its oxidation in the field may be quantified by the formation of carbonyl compounds.

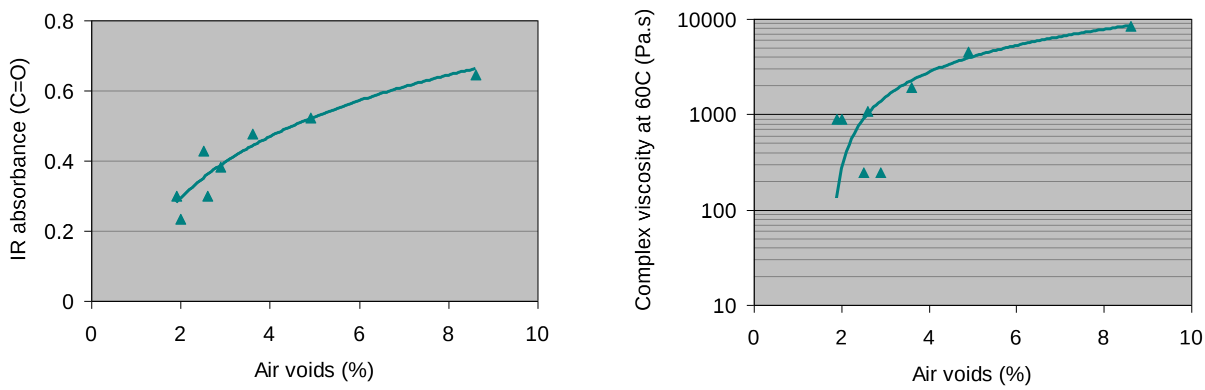

Figure 13: Bitumen composition and viscosity versus air voids for the underlaying old mixture AC d12 on Hldv 411

The field ageing of bitumen is also known to depend on the depth below the pavement surface. Such dependence is very much affected by the air void content or the permeability of the mixtures. When a dense upper-layer is applied, aging of the bitumen in the under-laying mixture can be reduced considerably. This is clearly shown in Figure 14 by the chemical and viscosity tests on the binders recovered from the same mixture AC d12 but under two different wearing courses (4e - SMA12 and 4f - AC o 12) placed next to each other on Hldv 411. On the other hand, for a mixture of high air voids, ageing of the bitumen could be quite uniform through the pavement [14].

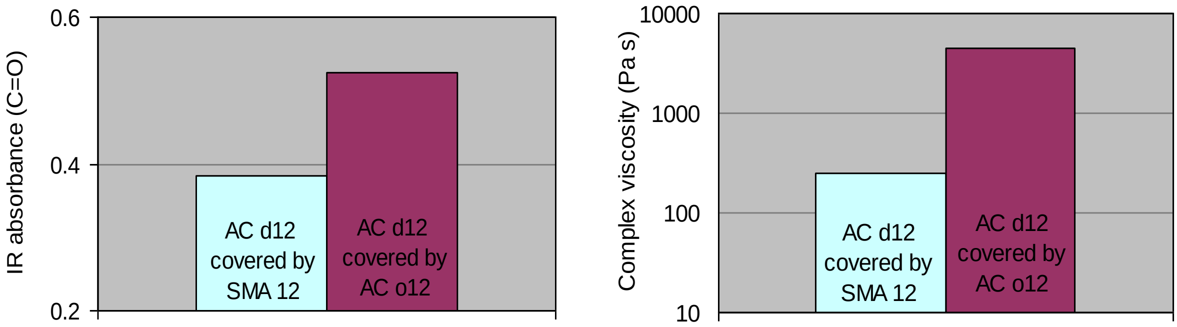

Figure 14: IR absorbance and complex viscosity at 60˚C for the binders recovered from AC d12 under the two different wearing courses

6. DISCUSSION AND CONCLUSIONS

In general, on the primary road network in Denmark, the structural strength of pavements is high and the pavements do not deteriorate structurally [15]. Therefore, overlaying of a road mostly was due to extensive surface damages, such as surface cracks, fretting and raveling, rutting, and patching. The surface damages are believed to be initiated and/or accelerated by high air voids in the asphalt mixture and severe ageing of the bituminous materials. As evidenced in this study, three of the selected road sections had an extremely slow ageing of the binder, which may be attributed to low air void contents of the asphalt material or low permeability of the pavement. The slow ageing of the binder had kept asphalt layers flexible enough to resist cracking. This has been confirmed by SuperPave IDT and the analyses using HMA fracture mechanics.

The use of higher binder content in asphalts is also believed to ensure longer durability for the pavement. This seems to be supported by the present study. In addition, the old asphalt layers constructed in 1970 and 1974 still consisted of relatively soft binders, indicating that the age-hardening of the binder had practically stopped when the layers were overlaid by a tight (dense) asphalt mixture. This is also true for test section 4f, even though the wearing course tested was noted as open graded. In fact, the cores taken from this section are rather dense with an aggregate gradation quite similar to SMA, except for those from road edges having 9.7% air voids. Unlike other test sections, section 4f has a binder content of less than 6% and shows rather severe age-hardening of the binder. However, this section was constructed with softer bitumen B100 compared to B60 used in other sections. The lower amount of bitumen ageing was also found on Swedish long-life roads [16]. After more than 25 years in the field, the retained penetration of the bitumen used was still higher than 40%.

It has been shown that all the surviving sections had relatively little surface cracking, and bottom-up fatigue cracking was not present. One speculation is that if a softer asphalt layer covers a harder asphalt layer, the risk of fatigue cracking is decreased. This is the case for the studied test sections, where the old wearing course AC d12 is stiffer than the surface layers tested. The traditional bottom-up fatigue cracking may be further prevented if thicker asphalt layers are applied [13]. As the test sections were constructed on the existing old asphalt roads, the total asphalt pavement structure become rather thick (> 200 mm, see Figure 1). Meanwhile, the asphalt mixtures of low air voids and high binder contents had probably minimized moisture effect, thus keeping a high strength for the pavement structure. All those suggest deteriorations be confined to the surface of the pavements and the risk of structural damages be eliminated.

In the Danish environment and on the roads with sufficient structural strength, the typical deterioration mechanisms include surface cracking, fretting and raveling, longitudinal unevenness, and rutting originated in the bituminous layers. Extensive raveling and fretting had caused many roads to be overlaid. The four test sections studied were also suffered from these damages. According to other investigations [17], stone loss is more aggregate property related rather than binder property related. However, based on this study, there is no evidence supporting that the four long lasting sections had better adhesion as compared to those failed sections.

In conclusion:

- A short lifetime of test roads in Denmark was mainly due to various surface damages, such as surface cracking, raveling, fretting, and patching. The surface deterioration is affected by the ageing of the asphalt materials used.
- The long lasting test sections studied in this paper were constructed on the existing old asphalt pavements. Thus the total pavement structure is rather thick. In addition, the existing old asphalt layers are stiffer than the test wearing courses, which provides a strong base in the road sections.
- The long lasting test sections consist of low air voids and high binder contents. These had minimised bitumen ageing (as expected), consequently contributed to the long-term durability of the pavements. By visual inspection and X-ray tomography on field cores, no propagated cracks are observed. The cracking resistance is also confirmed by the analyses using HMA fracture mechanics.
- Bitumen ageing versus the depth in the pavement is affected by the structure of the asphalt pavement. There is indication that a tight (dense) asphalt top layer may prevent ageing of the binder in the under-layer.
- The binders aged in the field show similar tendency in rheological changes and at a given stiffness level are more elastic as compared to unaged bitumen. The stiffening effect tends to correlate with bitumen carbonyl compounds and molecular weight, but not with sulfoxides, suggesting other chemical changes involved in the process of ageing.

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