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Ageing of hot mix asphalt

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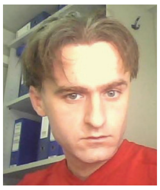

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Subject review

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Ageing of hot mix asphalt

An overview of current surveys related to the ageing of placed hot mix asphalt and asphalt pavement courses is presented in the paper. Asphalt ageing occurs as a result of external and internal factors. Internal factors include bitumen content in the mixture, thickness of bitumen layer enveloping aggregate grains, air void content, and properties of aggregates and binders. All factors affecting the ageing process of binders and asphalt mixtures are described in detail, including also some of the existing models that are presently used to predict ageing of produced and placed asphalt.

Key words:
binder ageing, ageing of asphalt mixture, volatilisation, oxidation, ageing prediction models

Pregledni rad

Ivica Androjić

Starenje vrućih asfaltnih mješavina

U radu je dan pregled dosadašnjih istraživanja vezano za starenje proizvedenih vrućih asfaltnih mješavina i izvedenog asfaltnog sloja. Starenje asfalta događa se uslijed djelovanja vanjskih i unutarnjih čimbenika. Unutarnji čimbenici podrazumijevaju udio bitumena u mješavini, debljinu sloja bitumena oko zrna agregata, volumni udio šupljina te svojstva agregata i veziva. Detaljno su opisani svi utjecajni čimbenici na proces starenja veziva i asfaltnih mješavina te dio prisutnih modela koji se u današnje vrijeme koriste za predviđanje starenja proizvedenog i ugrađenog asfalta.

Ključne riječi:
starenje veziva, starenje asfaltne mješavine, isparavanje, oksidacija, modeli za predviđanje starenja

Übersichtsarbeit

Ivica Androjić

Alterung heißer Asphaltmischungen

In dieser Arbeit wird ein Überblick bisheriger Untersuchungen bezüglich Alterung hergestellter heißer Asphaltmischungen und ausgeführter Asphaltschichten gegeben. Zur Alterung von Asphalt kommt es durch die Einwirkung externer und interner Faktoren. Interne Einflüsse beziehen sich auf den Bitumenanteil in der Mischung, die Stärke der Bitumenschicht um das Gesteinskorn, den Volumenanteil an Poren, sowie die Eigenschaften von Gesteinskörnung und Bindemittel. Daher werden alle Einflussfaktoren in Bezug auf Alterungsprozesse von Bindemitteln und Asphaltmischungen, sowie ein Teil vorhandener Modelle, die heutzutage zur Voraussage der Alterung hergestellten und verbauten Asphalts angewandt werden, detailliert beschrieben.

Schlüsselwörter:
Alterung von Bindemitteln, Alterung von Asphaltmischungen, Verdunstung, Oxidation, Modelle zur Voraussage der Alterung

1. Introduction

An asphalt mixture obtained by heating its constituent components contains stone dust, bitumen and possibly various additives. Today, the use of asphalt mixtures prevails in road construction. In the US, there are over 4,186 million km of paved roads, of which 93 % are coated with asphalt [1]. In Europe, asphalt mixtures are commonly built into final layers of the road structure [2]. An average production of asphalt mixtures for the period from 2007 to 2013 amounted to 307,1 million tons in the territory of Europe [3], whereas this amount is significantly greater in the US, where the production of asphalt mixtures amounts over 500 million tons per year [1]. This production leads to the significant energy demand of 85 kWh per tonne of HMA produced [4], resulting in an adverse impact on the environment due to discharge of large quantities of gases of CO_2, CH_4 and N_2O gases [5].
According to some authors, billion dollars are spent annually on the construction and maintenance of asphalt pavements. Notwithstanding the considerable cost involved, the durability of asphalt pavements may be up to 10 to 20 years, when their quality deteriorates due to fatigue damage, the appearance of temperature cracks and other cracks [6]. Asphalt pavements are mainly designed for the validity period of up to 20 years, with the appearance of certain performance problems in the first five years [7, 8]. An overview of the estimated durability of asphalt pavements, according to different sources, is given in Table 1.

Table 1. Durability of asphalt pavement layers

Authors	**Estimated durability of road pavement structures** [years]
Michigan Concrete Association [9]	15,5
Babić [10]	10 – 20
EAPA [11]	20
CEDR (2014) [12]	20 - 30

The exposure of asphalt pavements to various influencing factors during production and service life results in short-term and long-term ageing problems. The short-term ageing occurs in a mixer or drum, as a result of contact of heated aggregate with a very thin layer of bitumen. In addition to the mixer, the short-term ageing also occurs at other phases of production and placing. The long-term ageing of asphalt pavements occurs due to their exposure to external weather conditions. The ageing is the result of work of both internal and external variables. The internal variables refer to the properties of materials, asphalt mixture, binders, air voids content, and thickness of binder around aggregate. The external variables refer to the temperature of the mixture, external weather conditions, and the long-term exposure of asphalt surfaces to weather conditions.

The mechanism of asphalt ageing involves volatilisation, oxidation and steric hardening. The volatilisation and oxidation result from the change in molecular structure, whereas steric hardening is a result of a molecular rearrangement. Volatilisation occurs due to the rise in temperature during production, storage, transport and placing of hot mix asphalt. Produced asphalt temperatures in excess of 150 °C lead to the start of volatilisation of some bitumen fractions, and each additional 10-12 °C heating increment can double the volatilisation rate [13]. This leads to an increase in the bitumen viscosity from 150 to 400 % [14]. The oxidation of asphalt is due to reaction of complex organic compounds in the bitumen, and is also caused by atmospheric oxygen and UV radiation. The oxidation leads to an increased fragility and appearance of cracks in the asphalt layer [15]. Steric hardening of bitumen takes place over time at room temperature resulting in the molecular rearrangement in bitumen [16]. Physical hardening results in an increase in viscosity and appearance of volume contractions.
Depending on the type of mixture and its purpose, asphalt mixtures are predominantly designed with air voids content of 4 % (3 to 5 %). The properly constructed asphalt pavement will contain 6-8 % of air voids following its placing and 3-5 % of air voids content after a certain period of pavement use [17]. The ageing of asphalt layers results in physical and mechanical changes in bitumen. The physical changes refer to the reduced ductility, penetration, higher softening point and ignition, which is the result of a change in molecular size in the binder. The mechanical changes of bitumen properties lead to the growth of the mechanical and dynamic moduli from 100 to 400 %, according to some authors [18].
The laboratory simulation of asphalt ageing is focused on accelerating bitumen ageing as related to temperature, oxidation and Figure degradation. At that, the thickness of bituminous film around aggregate grains, and air voids content in asphalt, are taken into account. Tests are carried out at various time intervals ranging from days (short-term ageing) to months (long-term ageing). The most common tests used in the simulation of long-term ageing are the pressure ageing vessel for an accelerated ageing (PAV) and the rolling thin-film oven (RTFO) for the short-term ageing. The rolling thin film oven tester (RTFOT) is a test procedure in which a thin bitumen layer inside the bottle is exposed to a temperature of 163 °C for 85 minutes. The PAV treatment is based on the use of a vessel heated to 100 °C and pressurized to 2.07 MPa for 20 hours.
The paper provides an overview of the present surveys related to the ageing of bitumen and asphalt mixtures during production and exploitation. The aim is to explore the factors potentially influencing the ageing process, as well as the ways in which it is possible to increase durability of asphalt pavements during exploitation.

2. Impact of ageing on asphalt properties

Bitumen is a mixture of organic compounds resulting from petroleum distillation. Bitumen consists of a very large number of chemically different molecules that have not as yet been fully explored and determined by chemists [19]. Thus bitumen contains hydrocarbon and oxygen compounds with an admixture of nitrogen and sulphur compounds. Petroleum bitumen may contain about 80-90 % atoms of carbon, up to 11 % atoms of hydrogen, up to 1.1 % of nitrogen, up to roughly 5.5 % of sulphur, and other substances for various bitumen types [20]. According to the form of carbon skeleton, hydrocarbons can be acyclic (saturated, unsaturated) and cyclic (carbocyclic, heterocyclic). Carbon atoms in bitumen can be mutually linked in straight or branched chains (paraffinic types), in simple or complex saturated rings (naphthenic types), or in an unsaturated ring structure with six carbon atoms. Molecular interaction in bitumen depends on molecular size, its composition, polarity, and other. It is estimated that the worldwide use of bitumen amounts about 102 million tons per year of which 85 % is used in road construction, 10 % for roof covering, and 5 % for achieving water resistance of various construction materials [21]. The mass content of bitumen in the hot mix asphalt of continuous composition generally ranges from 3 to 8 %, and amounts to at least 6 % in mastic asphalt [22].

Bitumen ageing involves the change in physical properties of asphalt due to the change in its chemical composition. These changes result from interaction of internal and external variables defined as the short-term and long-term ageing. The ageing mechanism consists of volatilisation, oxidation, and steric hardening.

2.1. Volatilisation

The temperature rise in asphalt during its production, transport and placing, leads to the evaporation of lighter constituents of asphalt binder, which is defined as the short-term ageing. Light asphalt volatilisation is linked to the external weather conditions and UV radiation, which constitute an important influencing factor [23]. Volatilisation leads to the degradation of asphalt pavement, thus resulting in the increase in its stiffness and in negative impact on the functional and structural performance of road pavements. In his paper, Lolly (2013) examines the impact of bitumen exposure to high temperatures over time [24]. The corresponding results indicate that the highest growth in bitumen viscosity occurs as the result of increase in exposure time. Significant changes in dynamic modules develop during the first few hours of exposure to elevated temperatures, leading to a rise in the short-term ageing of the binder. In their survey, Cui et al. (2014) analyse properties of two bitumens during their volatilisation at high temperatures [25]. The results indicate that the mass content of bituminous binder decreases as a result of its exposure to high temperatures. A shorter volatilisation is followed by the growth in its softening point and fall in its penetration value. In their study, Kuszewski and others seek to determine the value of volatilisation for 20 bitumen samples [26]. The initial testing was performed on test samples each involving 50 g of bitumen (approx. 2.5 cm in thickness), which were heated to 163°C for 5 hours. Losses in the mass content of test samples due to volatilisation ranged from 0.01 to 0.14 %. The authors carried out further analysis of 20 samples of bitumen, each 12 g in weight and 6 mm in thickness. Losses in mass content due to volatilisation in the second series ranged from 0.6 to 0.64 %. This test leads to the conclusion that the reduction in mass content of test sample from 50 g to 12 g leads to a 4 times higher loss in mass content of the test sample due to volatilisation. In their paper, Cui et al. (2014) examine volatilisation of light organic constituents and behaviour of bitumen samples [27]. The results are compared to those obtained by testing samples containing the SBS and active carbon filler. The results obtained show that the combined addition of 4 % of SBS and 4 % of active carbon filler reduces volatilisation of bituminous test samples, which ultimately leads to their increased resistance to deformation at higher temperatures.

2.2. Oxidation

Complex organic constituents of bitumen react with atmospheric oxygen and UV radiation. This results in surface hardening, cracks and penetration of oxygen into cracks, which ultimately leads to oxidation. In his paper, Sung (2006) examines oxidation of 15 different types of binder [28]. He concludes that the oxidation does not occur on the surface of the pavement only, but it also adversely affects the entire depth of the structure. The binder on the road surface becomes harder and more brittle at a depth section of up to 15 cm [28]. In their paper, Hagos et al. (2009) state that it is difficult to accurately simulate in laboratory conditions the ageing that will occur during exploitation, mainly because of interaction of a large number of effects on ageing [29]. Furthermore, they also state that the existing laboratory ageing tests describe relatively well the ageing of asphalt of standard densities, which is not the case with porous asphalts. In their study on porous asphalt, Van Vilet et al. (2012) conclude that the increase in ageing leads to the growth of a complex modulus, and to the fall in the phase angle [30]. The sensitivity of bitumen to hardening due to oxidation is significantly affected by the origin of bitumen because its durability significantly depends on chemical composition [31]. The field demonstration of this dependence was performed at the test section in the US in 1950s [32]. The obtained results indicate that the durability of pavement is significantly affected by the origin of bitumen and its composition.

2.3. Steric hardening

Steric hardening refers to the hardening of bitumen at room temperature, due to molecular rearrangement in the binder [33]. In their paper, Masson et al. [33] note that the time period between the heating, pouring and testing must be at least 24 hours, so that a good bitumen testing repeatability can be achieved. In their study, Guern et al. (2010) examine physical and mechanical properties of five different types of bitumen prior to and after ageing [34]. Bitumens are chosen on the basis of their origin, processing methods, and modification characteristics. The aim has been to determine the influence of ageing on chemical composition of the binder. The obtained results indicate that the asphaltene content during ageing expectedly increases in all samples and that its agglomeration significantly depends on the type of bitumen, which ultimately indicates that the presence of crystallised fractions has a significant impact on the entire process.

Figure 1 shows the asphalt course whose properties have reached the limit exploitation values.

Figure 1. Asphalt surface deteriorated by use

3. Testing, models and techniques against ageing

Many laboratory methods for testing bitumen ageing are aimed at achieving the highest possible correlation with results obtained by field testing. They are divided into tests performed in oven and tests based on pressure oxidation. The short-term ageing of binder is linked to oxidation and the loss of volatile constituents. For the purpose of testing, the most commonly used tests include the thin-film oven test (TFOT), the rolling thin film oven test (RTFOT), and the rotating flask test ((RFT-12607-3: 2008 [35, 36]). The method of hardening in a thin film test with horizontal rotation is performed by placing a 50 g bituminous sample of predefined dimensions (HRN EN 12607-2 [37]) on the shelf located inside the ventilated oven in which a 163 °C temperature is maintained. The shelf rotates at a speed of 5-6 rpm for 5 hours. Such bitumen must meet requirements regarding minimum percentage of retained penetration or maximum viscosity. On the other side, the method of hardening in the thin film test with vertical rotation is performed by exposing the defined sample (HRN EN 12607 -1 [38]), contained in 8 bottles of bitumen each weighing 35 g, to a temperature of 163 °C for 85 minutes. This method simulates binder ageing during the mixing, transport and asphalt paving. The goal of the testing is to determine properties of bitumen prior to and after testing, and to define changes in the mass content of the sample [39].

The most important methods for testing long-term ageing involve the use of the pressurized ageing vessel (PAV-EN 14769 [40]) and the rotating cylinder ageing test (RCAT – HRN EN 15323:2008 [41]). The pressurized ageing vessel (PAV) involves bitumen pre-hardening (RTFOT or TFOT), which is followed by exposure of a bituminous sample 50 g in mass, 140 mm in diameter, and 3,2 mm in thickness, to the controlled conditions. The sample is exposed to the air pressurised at 2,1 MPa at temperatures of 85 °C (65 hours), 90 °C (20 hours), 100 °C (20 hours) or 110 °C (20 hours). The method with a rotating cylinder (RCAT) has been developed to simulate the short-term and long-term ageing process. Prior to the exposure of test sample to long-term ageing, it must be exposed to conditions appropriate for achieving the short-term ageing. This can be realised by applying the RTFOT method, the TFOT method or, directly, by the RCAT method, which can greatly facilitate the testing process.

3.1. Ageing models

Many studies consider empirical or semi-empirical models that take into account behaviour of the material in terms of its chemical, physical, and mathematical features. According to Vallerga et al. (1957), the time hardening of bitumen is affected by oxidation, volatilisation, polymerisation, thixotropy, syneresis, and separation [42]. One of older models is the ageing index (AI), which involves examination of bitumen viscosity [43] prior to and after its exposure to a temperature of 107° C for 2 hours:

$$AI = \frac{\eta_A}{\eta_B} \tag{1}$$

where is:

η_A - the viscosity after treatment

η_B - prior to the treatment.

An overview of viscosity value changes for specific test samples, according to Kandal et al. (1973), is shown in Figure 2 [44].

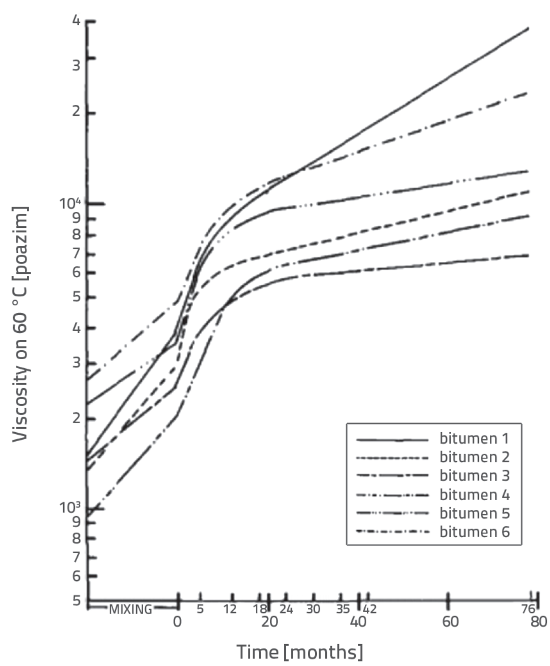

Figure 2. Viscosity of test samples [44]

Figure 2 shows that the bitumen viscosity value increases significantly in the phase of mixing of asphalt mixture components, as well as in the initial period due to installation. In his paper, Lee (1973) examines changes in the rheological and chemical properties of eight different bitumens during 48 months of use [45]. A parallel testing is performed in laboratory on bitumen samples of identical composition. The results of the study point to the following hyperbolic dependence of their physical properties:

$$\log(T_1) = \frac{T_f}{a + bT_f} \tag{2}$$

where is

T_l - he time required to achieve a certain property in laboratory

T_f - the time in the field needed to achieve a certain property..

In his paper, Benson (1976) examines factors affecting bitumen hardening [46]. As a result, he presents a model for predicting binder hardening as a function of time for viscosity (*V*) and penetration (*P*):

$$V = at^b \tag{3}$$

$$P = a + b\ln(t) = a \tag{4}$$

Many other authors [47-49] also present their ageing models for predicting properties of asphalt binders and mixtures. According to Roberts et al. (1996) [20], the value of time hardening, along with the ageing index, can also be expressed via the percentage of retained penetration (PRP):

$$P_{zp} = \frac{Penet_{old}bitum}{Penet_{new}bitum} \times 100 \tag{5}$$

The penetration value decreases due to ageing. According to the models shown, it can be concluded that the penetration value of less than 20 can lead to the occurrence of serious cracks. Bitumen is considered to be resistant to the occurrence of cracks if its penetration value is higher than 30 [44].

3.2. Techniques against ageing

Binder oxidation leads to the hardening of bitumen during bitumen production and placing, and during use of the road. Antioxidants of variable composition are used to minimise hardening and increase bitumen durability in the mixture. Adding polyethylene waste and gum into the asphalt mixture allows absorption of light fractions of bitumen, which provides for greater binder resistance to ageing [50]. The use of warm mix asphalt also increases durability and resistance to ageing due to reduction in oxidation and volatilisation as a result of lower production temperatures. Warm mix asphalts are produced at an approximate temperature of up to 135 °C, [51], which is significantly lower than the production temperature for hot mix asphalt (> 160 °C).

In their paper, Iwanski et al. (2013) state that during a road reconstruction project (Kielce, Poland), 30 % of lime was added to the SMA asphalt mixture instead of a part of stone dust [52]. After a few years, the tests conducted on binders indicated that the addition of hydrated lime increases binder resistance to ageing, which enabled realisation of higher resistance of bituminous mixture to the presence of water and freezing.

In their study, Zafari et al. (2004) explore the use of nano-silicon as an additive for enhancing resistance of asphalt binder to ageing [53]. The selected bitumen is exposed to the short-term radiation using the RTFO method. The obtained results indicate that the use of nano-silicon as a supplement to bitumen increases the values of complex modules and binder viscosity, and that its use improves the binder resistance to ageing, and other properties.

Lesueur et al. (2013) suggest that the hydrated lime acts as an active filler in the asphalt mixture and that it reduces the chemical ageing of bitumen, and stiffens it more than the normal stone dust above room temperature [54].

In their study, Edwards et al. (2005) analyse the influence of adding wax to road construction bitumen marked 160/220, and its impact on binder ageing [55]. The changes in rheological properties of wax-modified bitumen depend on the type of bitumen used, and the type and amount of wax added. The authors note that the addition of wax has no positive effect on bitumen resistance to ageing.

A lot of attention is nowadays paid to the use of the greatest possible content of recycled asphalt in the production of new asphalt mixtures. One of the problems when using a large proportion of recycled asphalt is an increase in stiffness of the total binder in the mixture, which may lead to reduction in the overall durability of the asphalt layer. In order to minimise this problem, softer binders are used in order to achieve required characteristics of the total binder. Air voids content is reduced to increase the binder content in the layer, and rejuvenators are used to recover its basic physical and chemical properties [56]. Rejuvenators often contain lubricating oils with a high content of maltene, which is lost by basic bitumen during service life. The effect of rejuvenators depends on the uniformity of addition to the recycled mixture and the diffusion of rejuvenators around the aggregate in an aged binder. In their paper, Zaumanis et al. (2013) explore the possibility of applying 6 different types of rejuvenators, which are added to the extracted binder sample [57]. The use of rejuvenators indicates that they cause softening of the extracted binder and the decrease in an optimum compaction temperature for 15 to 25 °C in relation to the extracted binder.

4. Conclusion

A short-term ageing of bitumen occurs during production, transport and placing of asphalt mixture. A long-term binder ageing occurs during the use of asphalt pavement when various external influences affect bitumen. Bitumen ageing is the result of the interaction of various external and internal variables. Internal variables include properties of materials, mixture and binders, air voids content, etc. External variables refer to the temperature of the mixture, external weather conditions and long-term exposure of asphalt surfaces to the weather conditions. The ageing mechanism of bituminous mixtures consists of the volatilisation, oxidation and steric hardening. Volatilisation occurs when heating bitumen during production, storage, transport and placing of hot mix asphalt. Bitumen oxidation occurs as a reaction of its complex constituent components to atmospheric oxygen and UV radiation. Steric hardening is the result of molecular rearrangements in binder during the bitumen hardening process at room temperature.

Ageing test methods are divided into tests performed in the oven, and test involving pressure oxidation. For testing the short-term ageing, the most common tests include the method of thin film hardening with horizontal rotation, the method of thin film hardening with vertical rotation and with the rotating flask. The most significant methods for testing the long-term ageing involve the use of a pressurized ageing vessel, and the method with a rotating cylinder (RCAT). The goal of these laboratory ageing tests is to achieve the highest possible correlations of the results obtained as related to field samples.

In order to reduce the negative ageing effect on properties of asphalt mixtures, various antioxidants are applied, warm mix asphalts are widely produced, and various additives are used to decrease the loss of volatile constituents from the produced and placed asphalt. Asphalt mixtures requiring lower temperatures are used during production, which results in a much lower consumption of thermal energy, reduced bitumen ageing, and extended durability of road pavements.

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