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THE EVALUATION OF AGEING PROCESSES OF THE POLYMER MODIFIED BITUMEN BY IMAGE ANALYSIS

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In this paper the influence of short and long term ageing processes on polymer dispersion in the polymer modified bitumen has been analyzed. The bitumen was modified by various amounts of SBS elastomers. Microstructural changes of polymer dispersion were tested using fluorescent microscopy. It has been shown that the phase separation of SBS modified binders is influenced by the base bitumen and the characteristics and content of the polymer. Correlations were sought between viscoelastic properties and microstructure after ageing. The results showed that it is essential to describe the microstructure of the polymer modified bitumen, especially the description phases, shapes and proportion of dispersed parts.

Key words: The polymer modified bitumen, ageing, microstructure, fluorescence microscopy

1. INTRODUCTION

The influence of vehicle traffic on road pavements causes natural aging processes and gradual loss of viscoelastic properties of bitumen binders. However, these phenomena have not been thoroughly investigated yet [2,3,5]. This is why the methods of simulating short-term and long-term ageing processes play an important part in the studies of the polymer modified bitumen.

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The results of these tests enable not only to determine the properties of the polymer modified bitumen, but also forecast the durability of composites with these binders during the use of road pavement [7].

In order to determine the changes of properties of the binders in the course of time, one must allow for the influence of external factors such as: exposure to high temperature, UV radiation and air. These factors result in the oxidation and evaporation of bitumen components and this, in turn, results in hardening of the binders [1,9]. However, when assessing the ageing processes of the polymer modified bitumen one must additionally take into consideration the behavior of bitumen, polymer and the changes taking place in their dispersed polymer-bitumen system [5]. Visual assessment methods of the microstructure of the polymer modified bitumen using fluorescence microscopy and image analysis tools allow to find relations between the microstructure and rheological properties.

In practice, this allows to determine more accurately the level of compatibility of bitumen and polymer, which enables efficient shaping of the polymer modified bitumen properties in relation to the initial bitumen and influences its durability [8].

2. MATERIALS AND TESTING METHOD

The bitumen was modified by various amounts of SBS elastomers, being subject to short-term ageing (TFOT) and long term ageing (PAV).

The ageing of modified polymer bitumen was performed by two methods. The thin film oven test (TFOT, PN-EN 12607-2) simulates changes in the properties of asphalt during the plant hot mixing and the lay-down process. The second method, the pressure ageing vessel (PAV, AASHTO PP-1-98) uses the residue from the TFOT test and is representative of long term ageing due to in situ field ageing for the first 5-10 years of road use.

The standardized binder tests used to characterize the base and the polymer modified bitumen, both original and after ageing processes, included the softening point (R&B, PN-EN 1427), penetration (at 5, 15 and 25˚C, PN-EN 1426), elastic recovery test (PN-EN 13398) and dynamic viscosity test (rotational viscometer, ASTM D 4402-02).

The observation of morphology of the polymer modified bitumen was performed with a fluorescence microscope (NIKON ECLIPSE E-200F PLUS). Photomicrographs with x200 magnification were taken at room temperature. The recorded image of tested samples was subject to quantitative image analysis using a computer program.

2.1. Tested materials

Bitumen from the Płock Refinery of 50/70 Pen grade, was modified by the addition of SBS 1101 elastomers with a linear molecular structure and SBS 1184 elastomers with a branched molecular structure whose properties are reported in Table 1.

Table 1. Characteristics of polymer SBS polymer

Polymer	Type of molecule	Characteristics
1101	line	Sometimes yields no stable product during hot storage. It is most often an SBS polymer used to modify road bitumen.
1184	branch	It considerably increases the softening point (R&B) and it is often used in combination with SBS 1101.

2.2. Sample preparation

Six the polymer modified bitumen samples were prepared by mixing a linear SBS elastomer (1101) and a branch SBS elastomer 1184 with base bitumen at three polymer contents in the quantities shown in Table 2. Also non-modified 50/70-type bitumen was tested for comparison purposes. All the modified bitumen was prepared with a low shear laboratory mixer at a temperature of 190°C, mixed for 4 h.

Table 2 Composition of binders used for testing

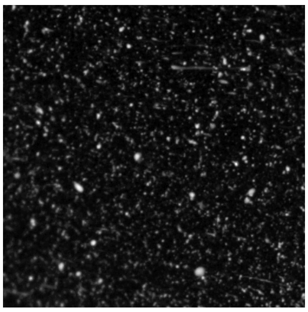
Composition of binders	SBS modification, % m/m	Content of polymer in binder, % m/m	
		SBS 1101	SBS 1184
0) 50/70	0	-	-
1) 50/70 3% SBS 1101	3	3	-
2) 50/70 6% SBS 1101	6	6	-
3) 50/70 9% SBS 1101	9	9	-
4) 50/70 + 2% SBS 1101 + 1% SBS 1184	3	2	1
5) 50/70 + 4% SBS 1101 + 2% SBS 1184	6	4	2
6) 50/70 + 6% SBS 1101 + 3% SBS 1184	9	6	3

3. VISUAL ASSESSMENT OF POLYMER MODIFIED BINDERS MICROSTRUCTURE

A visual assessment of polymer dispersion in bitumen binder can be made with the use of a microscope and through illuminating the samples with ultraviolet light. The observation was made at x 200 magnification with a system of optical filters. This method involves the phenomenon of different fluorescent induction (illumination) of materials illuminated with ultraviolet light.

When illuminating a sample of modified bitumen with UV light, there is a visible difference in the luminescence of the polymer phase and the bitumen phase. Using a filter we can observe the polymer phase in the light yellow or gold color and the bitumen phase can be observed in the black or dark brown color [10].

The samples were prepared in accordance with the European Standards EN 13632:2003 „Visualization of polymer dispersion in the polymer modified bitumen". This document standardizes the visualization method of polymer distribution in the polymer modified bitumen, involving fluorescent microscopy and qualitative description. This Standard specifies ten model pictures involving the information about the continuous phase, description of the phase, description of the size and description of the shape. Example of a qualitative description of bitumen modified by 3% SBS 1101 after PAV ageing is shown in Figure 1.



continuous phase:	B
description of the phase:	H
description of the size:	S
description of the shape:	r

B: continuous bitumen phase
H: homogeneous
S: small (< 10 µm)
r: roundish

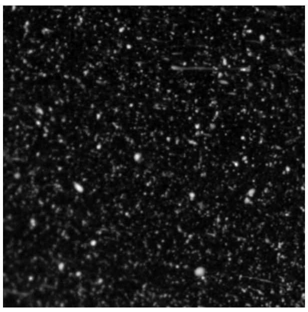
Fig. 1. Qualitative description of bitumen modified by 3% SBS 1101 polymer after PAV ageing

The description of the sample shown in Figure 1 in accordance with EN 13632:2003 allows us to asses the morphology of polymer-bitumen dispersion based on model pictures. It is connected with a very subjective evaluation of the microstructure and does not allow to join the microstructure with the properties of the analyzed material directly.

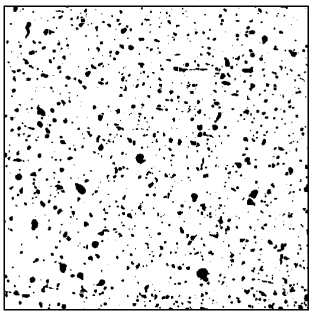
The study tries to describe the microstructure of the polymer modified bitumen quantitatively and connect it with the rheological properties. This is possible thanks to a limited number of strictly defined values describing the spatial structure of a solid, the estimators of which can be relatively easily defined on plane sections. This estimation does not require assumptions regarding the shape of the solid but only assumes its convexity [11].

4. CHANGES IN MICROSTRUCTURE ANALYSIS VERSUS VISCOELASTIC PROPERTIES

In order to characterize quantitatively the microstructure of the polymer modified bitumen, microscope images were recorded and transformed into a binary image. After the transformation, the image was filtered in order to remove unnecessary details and isolate the elements to be analyzed. Particle separation was used, selected from a number of image transformation methods involving computer image analysis program. Figure 2 shows the image of a microstructure transformed into a binary image after particle separation. Microstructure pictures were taken for all the polymer modified bitumen analyzed in the study; the pictures were subject to transformation into a binary image and quantitative analysis.



a)



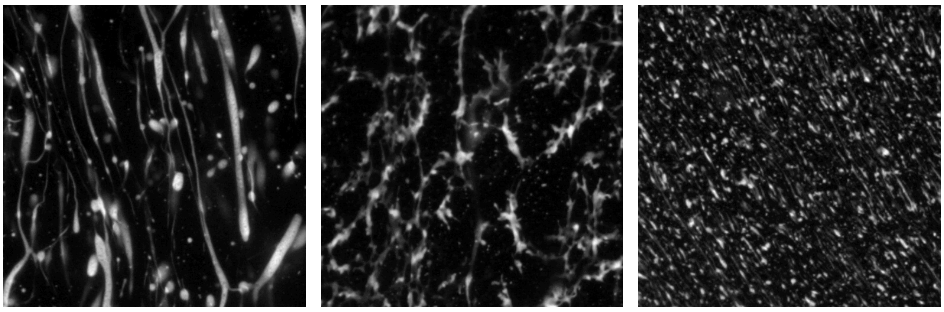
b)

Fig. 2. Transformation examples of the image of the microstructure of bitumen modified with 3% SBS 1101 after PAV ageing: a) microscope image recorded with a digital camera, b) image after transformation into a binary image and particle separation

A processed, separated phase of the material was subject to quantitative analysis that mathematically described the area of a polymer contained in the bitumen, the aspect ratio and the variation of particle shape in relation to the total population of the sample.

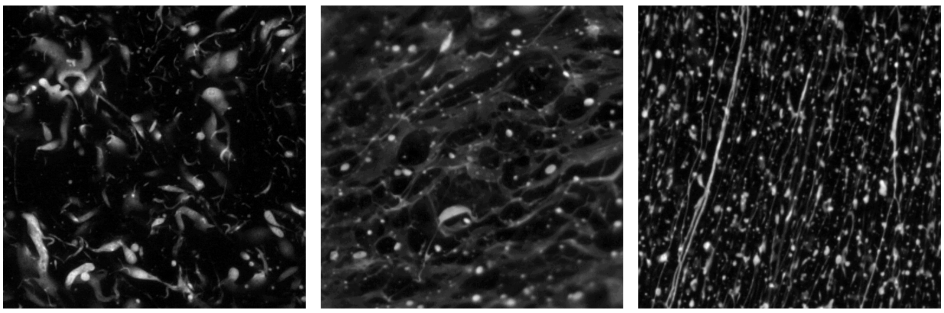
The initial analysis of the microstructure testing results has shown that the shape of grains or the layout of a polymer phase network cannot be comparatively analyzed at diversified modification level. As the tests have shown, it is justified to perform the quantitative analysis of the polymer area in the binder prior to ageing, after TFOT PAV ageing.

The initial attempts of the assessment concerning the influence of the microstructure change of the polymer modified bitumen on the viscoelastic properties (based on the microscope images) were made by analyzing the changes of the temperature sensitivity expressed by the penetration index (PI). The calculation of the penetration index was based on penetration tests at 5, 15 and 25 °C.



a) b) c)

Fig. 3. Computer photomicrographs of bitumen microstructure modified with a 6% addition of SBS 1101 and SBS 1184 polymer: a) prior to ageing, b) after TFOT ageing, c) after PAV ageing



a) b) c)

Fig. 4. Computer photomicrographs of bitumen microstructure modified with a 6% additive of SBS 1101 polymer: a) prior to ageing, b) after TFOT ageing, c) after PAV ageing

Figure 3 shows computer photomicrographs of bitumen modified with a 6% additive of a mixture of SBS 1101 and SBS 1184 polymer and Figure 4 shows computer photomicrographs of bitumen modified with a 6% additive of SBS 1101 polymer, prior to ageing, after TFOT and PAV ageing.

The analysis of the measurement results of the polymer content in the binders modified with a mixture of SBS 1101 and SBS 1184 polymers with variable quantities shows that the proportions between the polymer and the bitumen phases do not change (Figure 5). The short-term (TFOT) and long-term (PAV) ageing processes result in a slight increase in the polymer phase content (the area of a swollen polymer) irrespective of the modification level. A comparative analysis has shown that there is a correlation between the penetration index and the area of swollen polymer phases for the binders modified with a mixture of SBS 1101 and SBS 1184 polymers (R^2=0,8848). During all the stages of ageing, a small scatter of result points, marked with a dashed line, of one part of the modification as shown in Figure 5, indicates constant viscoelastic properties. This can also be confirmed qualitatively e.g. by the observation sample with an average polymer content (6% SBS 1101+1184). One can notice a similar structure character (Figure 3 a,b,c) with a tendency to diminish polymer particles after ageing, which indicates higher resistance to the degradation of a micro-structural system and ageing.

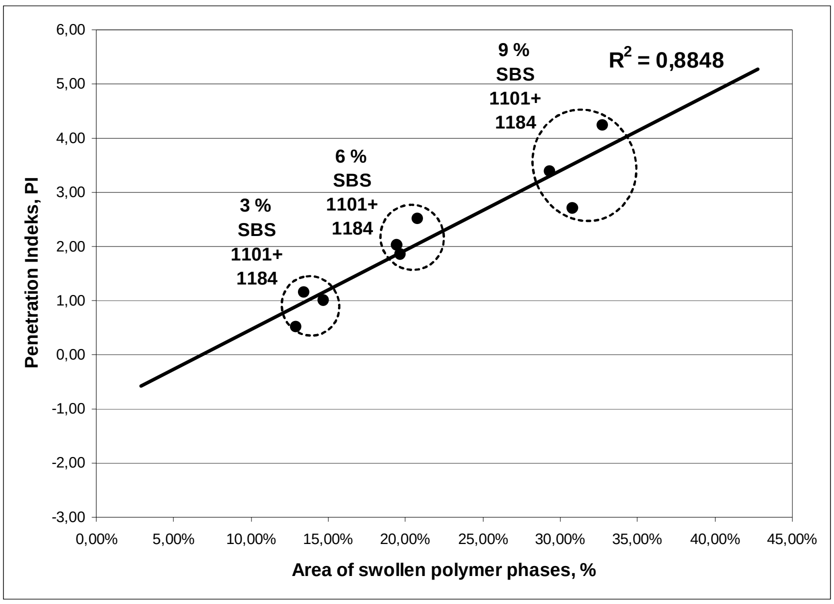


Fig. 5. Correlation between the penetration index and the area of swollen polymer phases of binders modified with a mixture of SBS 1101 and SBS 1184 polymers

The analysis of the measurement results of polymer content in the binders modified with various quantities of SBS 1101 polymer shows that the proportions between the polymer phase and the bitumen phase change (Figure 6). As the comparative analysis has shown, there is no correlation between the penetration index and the area of swollen polymer phases for binders modified with SBS 1101 polymers (R^2=0,0382).

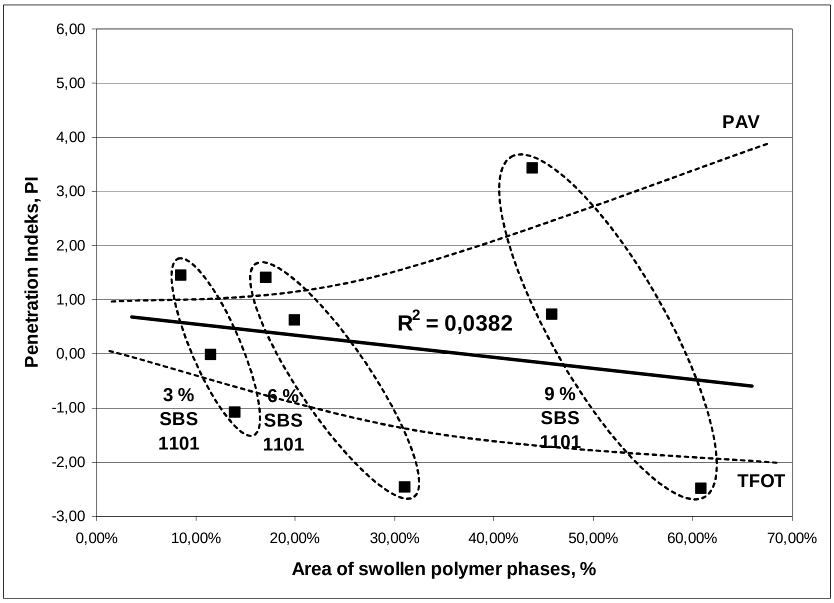


Fig. 6. Correlation between the penetration index and the area of swollen polymer phases of binders modified with SBS 1101 polymers

A short-term ageing process results in increasing the percentage area of the polymer phase and long-term ageing results in re-stabilization of the system. Wide microstructural changes account for no correlation. This phenomenon can be observed for all bitumen modified with various quantities of SBS 1101 polymer along with the increase of the modifier content. This increase can also be assessed qualitatively (Figure 4) and is the highest for the highest modified bitumen. In this case, as the modification level increases one can observe a sudden drop of penetration indexes resulting from TFOT short-term ageing. Negative PI values indicate high temperature sensitivity [6], which is reflected in the behavior of a polymer-bitumen system (Figure 4b). After TFOT+PAV long-term ageing the binders again accept high PI values, and the microstructure observed in the microscope becomes uniform (Figure 4c).

The results of microstructural observation have shown, with increased polymer content, a decreased stability of microstructure. It can be assumed, with the polymer and the bitumen compatibility. On a micro scale, bitumen and polymer cause the two phase system with a polymer reach phase. Polymer absorbs maltenes (oils) from the bitumen and the asphalten fraction is the dominant residue. The elastomeric phase of the SBS copolymer can swell up to nine times its initial volume but its capacity of maltenes absorption decreases due to the polymer content [1,4].

In the case of binders modified with a mixture of SBS 1101 and SBS 1184 polymers there is a tendency of increasing penetration index values due to the increase of the modification level. The lack of significant changes in the penetration indexes resulting from the ageing processes for bitumen modified with a mixture of elastomers is related to the unchanging content and character of the polymer phase in these binders.

5. CONCLUSIONS

The nature of a polymer swelled network and its influence on the polymer modification is a function of the base bitumen properties, content and characteristic of the polymer and the bitumen–polymer compatibility. Quantitative analysis of the polymer modified bitumen microstructure, based on the microscope images, is a good method for identifying the changes in viscoelastic properties taking place in modified binders during the modification and ageing processes. It can be used to make deductions regarding the homogeneity of polymer-bitumen systems in time and the properties of the polymer modified bitumen.

Viscoelastic properties of road bitumen are improved by means of SBS polymer modification. It is advisable to replace a part of the elastomer with a linear particle for modifying road bitumen with a branched particle elastomer. Binders with a branched particle display increased resistance to the ageing processes. They have lower temperature sensitivity and lose the properties slower in comparison to bitumen modified with a linear particle polymer only.

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