Estimation of impacts of cellulosic admixtures on SMA features

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Abstract. It is known that the supporting capacity of asphalt concrete depends on the weakest component of the system. Currently, there are two main ways to increase strength and shear resistance of asphalt concrete: modification of bitumen binder or regulation of granulometric composition, in the trend of increasing the rock stone content. However, in the first case, the structure-forming effect of multi-crushed stone asphalt concrete is not fully used, in the second – due to the high bitumen consumption necessary to fill intergranular bugholes, the mixture segregates. The elimination of this disadvantage was provided by the introduction of fine-fibred filler into mixture. Stone mastic asphalt concrete (SMA) was created, which fundamentally combines the advantages of impacted low-crushed stone concrete and maximum structural asphalt-concrete, which are widespread in some countries for roads with high traffic stream. In this paper, we studied the effect of various types of stabilizing additives on the performance of stone mastic asphalt concrete.

Key words: asphalt concrete, bitumen, strength, shear resistance, fine-fibred filler, IR-spectroscopy method.

1 Introduction

The supporting capacity of asphalt concrete largely depends on the weakest system component, that is, on the binder (bitumen). Currently, there are two main ways to increase the strength and shear resistance of asphalt concrete: changing of binder characteristics by modifying it, increasing the role of the crushed stone carcass, that is, increasing the structure-forming effect of crushed stone carcass both by adjusting the granulometric composition and by adding slag [1, 2].

In the recent past, these two trends were used separately. Moreover, the viscosity increase was limited: the transition to bitumen grades BND 40/60 was inevitably accompanied by an increase in the embrittlement of asphalt concrete and intensive cracks formation on the coating. This problem was solved by the bitumen modification by polymers. However, in this case, the structure-forming effect of multi-crushed stone asphalt concrete was not fully used. The increase of rock stone content in the asphalt concrete mix composition to 45-55 % led to an increase the shear resistance of asphalt concrete by 1.3-1.4 times in comparison to low-crushed stone asphalt concrete. At the same time, high-crushed stone asphalt concrete turned out to be unstable under the water influence and repeated freezing-thawing.

There was the fundamental problem of obtaining shear-resistant, water and freeze resistant asphalt concrete, while having a rugged surface. To increase shear resistance and nonskid quality, the rock stone content was up to 70-78 % and the poriness between granular components was filled up by large number of mineral components that increased the bitumen content in the mixture. On the one hand, the specific bitumen consumption increased due to formation of thicker bitumen film (almost up to volumetric state) due to increase of large grains. On the other hand, an increase of bitumen consumption was caused by an increase of mineral powder content. Thus, the mixture satisfies the
conditions of wettability and mixing, acquired a new negative property - it was separated during transportation. The elimination of this drawback was ensured by the introduction of fine-fiber filler with large specific surface into the mixture [3]. The stone-mastic asphalt concrete (SMA) was created, which fundamentally combines the advantages of impacted low-crushed stone concrete and maximum structural asphalt-concrete. Stone-mastic asphalt concrete is mainly used for roads with large traffic stream in some countries [4].

The stone mastic asphalt combines the advantages of structure-forming (up to 80 % rock stone) asphalt in relation to increased strength characteristics, as well as mastic asphalt (increased binder content) with respect to temperature crack and corrosion resistance, especially in climatic conditions of Russia. Many researchers pay attention to optimizing the mixture selection, combining the choice of mineral aggregates, type of binder material and stabilizing additives. It is interesting that road builders have not finally decided on the choice of the most important composition SMA component – stabilizing additives. Stabilizing additive is a substance that has the stabilizing effect on SMA and ensures its separation resistance. A stabilizing additive is the special component in the SMA composition and requires a more detailed consideration, based on its significant role [5].

The term "stabilizing additive", applied to SMA, primarily means the substance that holds the cementitious material to ensure the mixture homogeneity during transportation and paving. Among the many stabilizing additives, the most commonly used are artificial additives, most often cellulose, as well as some synthetic fibers [6]. Varieties of cellulose fibers can be presented in free form, as well as in the form of granules, both from neat cellulose and mixed with various hydrocarbon products: bitumen coating (the granular type of additive was taken in this work as the main one due to its popularity and availability on market) [7, 8]. Granular fibers have several advantages from the point of view of their lower hydroscopicity compared to free fibers, they do not clump and are better distributed in a mixer [9].

Currently, work is being done to improve the operational properties of stone mastic asphalt concrete in various trends, such as: introducing rubber crumb into the composition [15, 16], introducing mineral and organic fibers [17], and introducing various modifications for granular polymers [18].

These measures, in turn, incur large economic costs in the first stages of commercialization, in contrast to an elementary and material selection of a stabilizing additive [19, 20].

2 Materials and methods

2.1 Experimental

For research, we used 4 types of stabilizing additives of the most famous manufacturers in Russia. In this work, they are represented under the numbers № 1, № 2, № 3, № 4. The material composition of stabilizing additives was evaluated using spectral analysis of the samples. The test was carried out on the Fourier transform infrared spectrophotometer Perkin-Elmer, model Spectrum 65, using a Miracle ATR ATR attachment (ZnSe crystal) in the region of 4000-600 cm⁻¹, usually with 20 scans. To register the IR spectrums, the samples of the treated cellulose in the form of black granules were pressed to the ATR crystal with a special clamp included in the set-top box. After registration, an ATR correction and spectrum preservation were automatically carried out. The spectrums were processed using the supplied software (Perkin-Elmer Softwear V.10.1.1).

The influence of the stabilizing additive type on the physicomechanical properties of SMA was carried out on standard samples, for which mixtures with separate fractionation of mineral grains and the stabilizing additives № 1, № 2, № 3, № 4 were made in laboratory conditions. For carrying out the work, the particle size distribution of SMA 20 was selected, containing: 76 % of the rock stone, 11 % of the ragging siftings and 13 % of the mineral powder (not dispersed). The bitumen amount in the mixtures composition was 6.1% over the mineral part and 0.4 % of the stabilizing additive by weight of the mixture, the granulometric composition, the optimal binder amount, and the stabilizing additive amount were taken based on works [10, 11].
The bitumen was used as a binder for the SMA preparation, and was characterized by: penetration at 25 °C - 89 × 0.1 mm, softening point - 48 °C, tensibility at 25 °C > 100 cm, which allows it to be classified according to DSTU 4044-2001 to bitumen grade BND 90/130, as well as it makes notice of the improvement of bitumen properties when stabilizing additive from cellulose were added to the mixture [12, 13].

3 Results and discussion

IR-spectrums of the studied samples are shown in figure 1.

![Figure 1. IR-spectrums of cellulose samples 1, 2, 3 and 4.](image)

All four samples are characterized by the presence of characteristic bands at 2920 and 2851 cm⁻¹, corresponding to stretching vibrations of CH and CH₂ groups, as well as a peak at 1029 cm⁻¹, which is caused by vibrations of C-O bonds, which are part of the pyranose cellulose cycles. The first thing to pay attention to is the presence of intense bands of 1409, 872, and 712 cm⁻¹ in the spectrum of sample 2, which are practically absent in the spectra of samples 1, 3, and 4. Analysis of these peaks allows us to conclude that they are due to the presence in the spectrum sample 2 of the large amount of calcium carbonate CaCO₃, or simply limestone. Indeed, a comparison of the spectra of sample 2 and neat limestone shows that peaks of 1409, 872, and 712 cm⁻¹ are present in both spectrums, i.e. in the spectrum of sample 2, its respond to calcium carbonate (figure 2).

![Figure 2. IR-spectrums of calcium carbonate (1) and sample 2 (2).](image)
The next step in the work was to establish differences in the properties of samples 1 and 3. The IR-spectrums of these samples, as well as the spectrum of neat bitumen, are shown in figure 3.

![Figure 3](image)

**Figure 3.** IR-spectrums of samples 1(1), 3(2) and neat bitumen grade BND 70-100 (3).

The comparison of the spectrums 1 and 3 shows that the content and structure of cellulose are approximately the same, because the 1029 cm\(^{-1}\) band corresponding to cellulose coincides in configuration and intensity. Analysis by the peaks of stretching vibrations of the \(\text{CH}\) and \(\text{CH}_2\) groups in this case is not possible, because in this area, there is a strong crossover of vibrations of these groups from both cellulose and bitumen. However, it is possible to trace differences in the region of bending vibrations of the \(\text{CH}_2\) groups at the frequency of 1460 cm\(^{-1}\) and the \(\text{CH}_3\) at the frequency of 1376 cm\(^{-1}\). As it is seen from figure 3-3, in the bitumen there are intense peaks of both types of oscillations. Moreover, if we compare the spectrums of the samples 1 and 3, then we can see that in the second spectrum the peak intensity of 1460 cm\(^{-1}\) corresponding to the \(\text{CH}_2\) groups of bitumen is somewhat higher than in the first. From this we can conclude that the bitumen content in sample 3 is slightly higher than in the sample 1.

![Figure 4](image)

**Figure 4.** Compression strength at 20° C, MPa, of 4 compositions.
The estimated indicators in comparative tests were: strength at 20°C and 50°C, shear resistance by internal friction coefficient, shear resistance to adhesion during shear at temperature of 50°C. The results obtained generally corresponded to the requirements of domestic regulatory documents.

Nevertheless, despite the same composition, as well as the mineral components and bitumen used, there are still some differences in the results. In the composition with stabilizing additive № 2, the compressive strength at 20°C is slightly higher than that of similar compositions with additive № 1 and 4, and on 8% higher relative to the composition with additive № 3 (figure 4).

The highest compressive strength at 50°C also has the composition with the stabilizing additive № 2 (figure 5).

![Figure 5. Compression strength at 50°C, MPa, of 4 compositions.](image)

Figure 5. Compression strength at 50°C, MPa, of 4 compositions.

Figure 6 shows the shear resistance test results of compositions. It can be seen that the composition with stabilizing additive № 2 also has a higher performance.

![Figure 6. Shear resistance of 4 compositions.](image)

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The fracture toughness index by the tensile strength at split at 0°C increases by 7-9 % (figure 7).

![Figure 7. Fracture toughness index of 4 compositions.](image)

The draindown index decreases by 1-3 % (figure 8).

![Figure 8. Segregation resistance of 4 compositions.](image)

An obligatory technological stage of SMA preparing is to determine the optimal bitumen content using the draindown method. The method of draindown evaluation is to determine the binder remaining percentage on glass walls after keeping the mixture in it at a temperature of 170°C for one hour. According to the results of determining the draindown index, the optimum bitumen amount was established as applied to the type of stabilizing additive.

We compared the obtained numerical values of designed compositions characteristics with the standards requirements. The compositions satisfy these requirements.

During the tests, the regularity of changes in physical and mechanical properties was revealed depending on used stabilizing additive.

Possibly, higher strength characteristics of this composition are associated with higher microparticles CaCO$_3$ content in a stabilizing additive composition, which form stronger chemisorption bonds with bitumen, with the formation of calcium soaps at the “bitumen-filler” phase boundary [21]:

$$\text{CaCO}_3 + 2\text{R COOH} = \text{Ca(R COO)}_2 + \text{H}_2\text{O} + \text{CO}_2.$$  \hspace{1cm} (1)

Indicators of average density and water saturation, in general, have approximately equal values. It indicates the uniformity of the macrostructure of considered compositions.
Conclusion
Thus, the number of cellulose samples treated by bitumen was studied in the work. It was shown that in the sample №2 there is the high content of calcium carbonate CaCO$_3$, which is the most likely limestone ragging up to micron-sized particles. In the samples 1, 3 and 4, only trace amounts of this mineral are observed (not more than 5% by weight). At the same time, in the sample 2, the CaCO$_3$ content is not less than 50%.

The differences in properties of samples 1 and 3 are probably caused by the difference in bitumen content in these samples. The sample 3 gives the more intense band of 1460 cm$^{-1}$, corresponding to deformation vibrations of the CH$_2$ groups of bitumen than the sample 1, with approximately equal cellulose content. The samples 1, 3 and 4 practically do not contain calcium carbonate.

The regularity of increasing the physico-mechanical properties of asphalt concrete mix in the composition containing stabilizing additive №2 was revealed. This is due to the high content of calcium carbonate CaCO$_3$ in the stabilizing additive №2, which forms stronger chemisorption bonds with bitumen at the phase boundary.

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