Efficiency of using shungite filler for modifying organic binder

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Abstract. Modification of bitumen with various additives, including polymer ones, is one of the most effective ways to regulate the performance properties of asphalt concrete. Due to the high cost of polymers, a promising area of research is to reduce the polymer content, for example, by introducing mineral filler into its composition. A wide range of properties and unique structure of fullerene-containing natural shungite allow it to be used in various branches of science, industry and technology to create a variety of materials and determine the search for new areas of its application. One of the ideas that are currently under development is the use of shungite in conjunction with an organic binder used for the production of asphalt concrete. The work is devoted to the study of the influence of fine shungite on the characteristics of polymer-bituminous binder (PBB) and asphalt concrete. It is shown that the introduction of shungite into the PBB increases its viscosity, adhesive and cohesive properties, and the homogeneity of the mixture. This is due to the unique composition, structure and properties of shungite. The positive influence of the use of polymer shungite binder on the quality of asphalt concrete was established. With the introduction of 5% shungite in the PBB composition, the strength at 20 and 50°C, the strength of water-saturated samples, and the shear stability of polymer asphalt concrete significantly increased. This made it possible to reduce the polymer content of PBB by 10%.

1. Introduction

One of the most effective ways to regulate the physical, mechanical and operational properties of asphalt concrete is the modification of bitumen with various additives. Currently, the most common method of improving the quality of bituminous binders is the introduction of polymers in them. The use of polymer modifiers reduces the sensitivity of bitumen to temperature changes, extends the temperature range of its operation, and increases the cohesive strength and elasticity. However, this technology is quite expensive [1-3].

One of the promising ways to reduce the cost of polymer-bituminous binders (PBB) is to reduce the polymer content, for example, by introducing mineral filler. The use of calcium carbonate, clays, metal oxides, and carbon nanotubes is known as polymer matrix filler [4-5]. The use of natural shungite, which has a unique structure and properties: high adsorption activity, bactericidal, catalytic, and reducing properties is of significant interest in this direction [6-8].
The positive effect of shungite when used in bituminous binders and asphalt concrete is known from the literature [9-11].

The purpose of this work is to obtain a polymer-bituminous binder with a reduced amount of polymer while maintaining or improving the quality of PBB and asphalt concrete with its use.

2. Methods and materials
This task was solved by introducing a polymer-bituminous binder heated to a temperature of 160°C, a fine dispersed filler – shungite. BND 90/130 bitumen was used as the initial binder, polymer-styrene-butadiene thermoplastic layer of Korea Kumho Petrochemical Co., plasticizer – Unioplast produced by Selena LLC (Shebekino, Belgorod region). The filler was fine dispersed shungite (Karelite), provided by the Karelian investment company “RBK”.

PBB and asphalt concrete characteristics were determined using standard methods. Additionally, the bitumen cohesion according to ASTM D3910-98 and the PBB microstructure were studied using the AXIO SCOPE A1 (Carl Zeiss) laboratory research microscope in transmitted light.

3. Results and discussions
In order to find out the rational amount of shungite that allows replacing an expensive polymer in the composition of PBB, experiments were conducted on the selection of PBB compositions with different amounts of polymer and shungite. The amount of SBS was from 3.5 to 2.97 %, shungite from 3 to 10 % relative to the mass of BND 90/130 bitumen.

From the data obtained, it follows that when shungite is introduced into the polymer-bituminous binder, its softening temperature remains at the level of the initial PBB. The depth of penetration of the needle, which determines the plasticity of bitumen at 25 and 0°C, decreases with the introduction of shungite. This trend is observed for all compositions with different polymer content (3.5; 3.15 and 2.97 %), which indicates a significant structuring effect of the filler. For the same reason, which is quite logical, there is a decrease in extensibility at 25 and 0 °C after the introduction of shungite. This is especially expressed when the content of shungite in the amount of 10 % of the mass of PBB. The elasticity of the PBB remains at the level of elasticity of the original composition. At the same time, due to the introduction of shungite, complete homogeneity of the modified binder is achieved (in comparison with the binder without an additive), which was estimated from micrographs (Fig. 1).

![Figure 1](image)

Figure 1. Micrographs of polymer-bituminous binder a) without the introduction of shungite; b) with the introduction of shungite.

It also follows from the results that increasing the amount of shungite by more than 5% in the PBB is impractical. Although the introduction of 10 % shungite allows getting a binder that meets the regulatory requirements, however, the depth of penetration of the needle does not correspond to the PBB 60 brand. When using shungite in the amount of 3%, this indicator is at the lower limit of the requirements of GOST R 52056-2003, so reducing the amount of filler is also impractical.

From the results obtained, it follows that the compositions containing 3.15% SBS and 3 and 5% shungite are rational. This allows for a 10% reduction in the concentration of the polymer compared to the control compositions. All indicators of these compositions meet the requirements of GOST R
52056-2003. Thus, it is shown that it is possible to reduce the amount of polymer by 10 % in the PBB due to the introduction of shungite.

Research was conducted on the effect of shungite on the cohesive strength properties of PBB. The definition of cohesion is based on measuring the shear strength of a thin layer of bitumen (10 mc) at a constant rate of load increase. The binder compositions are shown in table 1.

| Type of additive | № of compound | The amount of SBS from the mass of the initial bitumen, % | Shungite content, % |
|------------------|---------------|-----------------------------------------------------|-------------------|
| BND 90/130, without additive | 1 | 0 | 0 |
| SBS 3.5 % | 2 | 3.5 | 0 |
| | 3 | 3.5 | 5 |
| SBS 3.15 % | 4 | 3.15 | 0 |
| | 5 | 3.15 | 5 |

Test results are presented on Fig. 2.

![Graph](image)

**Figure 2. Cohesive strength properties of the binder.**

Cohesion is similar in nature to viscosity and is determined by intermolecular bonding forces and the structure of bitumen. The results show that shungite has a significant structuring effect. The cohesion of composition 5 with 5 % shungite and with lower polymer content (3.15%) is equal to this indicator for composition 2, which contains a high concentration of SBS (3.5%). Accordingly, it can be assumed that the obtained PBB compositions filled with shungite will have higher strength characteristics than the original ones.

The adhesion of bitumen-polymer-shungite binder to the surface of crushed stone was also evaluated.

The quality of adhesion was assessed visually by the degree of preservation of the film of bituminous binder on crushed stone grains, prepared beforehand in accordance with GOST 12801-98, after boiling it in distilled water for 30 minutes. The bonding quality of the binder was determined for the same compositions as the cohesion.

Test results are given on Fig. 3.
Figure 3. Adhesion of test samples to granite crushed stone.

The results indicate that PBB using shungite has better adhesion compared to the control composition, which is due to the positive effect of the filler on the properties of bitumen: increased viscosity and cohesive strength. In addition, shungite contained in bitumen gives the binder increased activity due to the high concentration of paramagnetic centers. This should have a positive impact on the properties of the road surface.

The influence of PBB filled with shungite on the properties of asphalt concrete cover was studied using the example of polymer asphalt concrete. Samples of polymer asphalt concrete were formed on PBB with a concentration of SBS polymer of 3.5 and 3.15 % and on PBB with the same SBS concentrations, but additionally containing 5 % shungite. The compositions are shown in table 1.

Fig. 4 shows the results of changing the strength of samples of polymer asphalt concrete prepared on modified bitumen compared to samples on the original BND 90/130 bitumen.

Figure 4. Changes in strength characteristics of polymer-asphalt concrete samples.

The analysis of the obtained data shows that there was an increase in the main strength characteristics of the polymer-asphalt concrete mixture for all compositions using the studied additives.

A significant increase in the strength at 20 and 50˚C, as well as the strength of water-saturated samples, was found. Thus, the compressive strength at 20˚C when using SBS thermoelastolayer as an additive increases by 22.66 and 24.44 % (compounds 2 and 4), and when introducing shungite in the amount of 5 % (compounds 3 and 5) – by 30.44 and 26.22%, respectively. The introduction of SBS,
and especially shungite, had a much greater impact on the strength at 50°C. The compressive strength at 50°C when used as an additive of SBS thermoplastic layer increases by 53.46% and 48.51% (compounds 2 and 4), and when introducing shungite in the amount of 5% (compounds 3 and 5) – by 76.23% and 63.36%, respectively. This is due to an increase in the viscosity, softening temperature, cohesion and adhesion of the binder when using the studied additives and should have a positive effect on the stability of the asphalt concrete coverage to rutting.

As expected, the introduction of shungite had a significant impact on the adhesion at shifting (Fig. 5). This indicator increased with the introduction of SBS in bitumen by 53.3% and 56.6%, and with the use of shungite by 80%.

![Figure 5](image_url)

**Figure 5.** Adhesion at shifting of asphalt concrete on various compounds of the binder.

Thus, the introduction of shungite in the PBB containing 3.5% and 3.15% SBS leads to an increase in the strength of polymer asphalt concrete at 20 and 50°C, water resistance, and shear stability.

**4. Summary**

It is established that the use of shungite as a part of polymer-bituminous binder leads to an increase in its viscosity, cohesive and adhesive characteristics, and the uniformity of PBB. This is due to the high adsorption capacity of shungite, which is associated with the presence of natural fullerenes, surface structure, and the presence of active adsorption centers. The introduction of 5% shungite with a reduction in the amount of SBS from 3.5 to 3.15% allows getting asphalt concrete, the characteristics of which are not inferior to those for a compound containing 3.5% SBS, but without shungite.

Thus, when using shungite, it is possible to reduce the amount of SBS in the mixture by 10% without losing the quality of asphalt concrete.

**5. References**

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