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USE OF SULFUR BINDER IN ASPHALT AND CEMENT CONCRETE MIXTURES IN THE CONSTRUCTION OF AIRFIELDS

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Abstract. Currently, work on the creation and production of new composite materials capable of long-term operation in industrial, climatic and other types of aggressive environments is one of the main tasks of the airfield industry. When organizing the production of new building materials in the context of rising prices for raw materials and energy resources, it is necessary to study the possibility of replacing the traditional raw material component with cheaper, more common raw materials, while simultaneously increasing the durability of airfield construction. One of these materials is technical sulfur. The organization of the production of sulfur-containing composites – sulfur asphalt concrete and sulfur concrete will ensure the production of new materials characterized by reduced energy consumption and ensuring reliable operation in conditions of variable temperatures and aggressive environments without increasing their cost compared to traditionally used materials. A significant reduction in excess sulfur will have a positive impact on the environmental situation in the areas of its production, a significant reduction in greenhouse gas emissions in the production of sulfur composites compared to the production of existing analogues based on traditional materials.

Keywords: sulfur binder, asphalt concrete, cement concrete, construction, airfields.

Introduction

One of the materials that can be used for the preparation of composite road building materials (sulfur concrete and sulfur asphalt concrete) is technical sulfur.

Sulfur concretes have a number of positive properties, which primarily include: rapid strength development associated with the cooling period of the mixture, relatively high compressive strength (up to 60 MPa) and bending tensile strength, wear resistance and water resistance, resistance to aggressive media, low water absorption and therefore high frost resistance (Fomin & Khozin, 2009).

In addition, the properties of sulfur and its melt make it possible to replace strong aggregates with weak stone materials and man-made industrial waste (ash, slag).

Also, a well–known direction for the use of sulfur in construction is the modification of oil road bitumen with it. In this case, sulfur can play the role of a modifier of the properties of individual fractions of bitumen (asphaltenes and resins), as well as its filler, which has a reinforcing effect. In the first case, the introduction of sulfur into bitumen in small quantities (up to 20 wt. %) is envisaged.

In the second case – a larger amount of sulfur to obtain a colloidal system "sulfur-bitumen", stable during storage and remelting. At the same time, the excess part of sulfur, not dissolved in bitumen, in the process of crystallization manifests itself as a kind of filler. Serobitumen or sulfur-bitumen binder is used in the technology of asphalt concrete produced by hot technology, and the final product is a material – sulfur asphalt concrete. Its effectiveness is due to the possibility of replacing up to 40% of bitumen in asphalt concrete mixtures with sulfur (Vasilyev, 2014).

The addition of sulfur to the asphalt concrete mixture improves its workability, increases strength up to 30%, increases the durability of asphalt concrete, increases its thermal and dynamic stability, water resistance and reduces the amount of shear deformations of pavement (Al-Hadidy, 2022a).

Studies conducted by the Al-Hadidy (2022b), McBee et al. (1985) show that when mixing the constituent components of asphalt concrete with sulfur, the mobility of the mixture improves. That is, sulfur can also be used for cast asphalt mixtures.

In general, the possibility of using sulfur in both solid and molten state to obtain sulfur concrete and sulfur asphalt mixtures that meet the requirements of current regulatory documents has been proven.

Considering the cheapness of this material and its presence in Ukraine in sufficient quantities, sulfur

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deserves considerable attention as an integral component of asphalt concrete and cement concrete.

1. Properties of technical sulfur

Technical sulfur is one of the most widespread non–metals in the nature around us, capable of combining with almost all chemical elements.

From the point of view of physical characteristics, technical sulfur is a solid crystalline substance, stable in the form of two modifications: α – rhombic (density 2.07 g/cm^3) and β – monoclinic (density 1.97 g/cm^3).

Depending on the temperature, sulfur can take three states: solid, liquid, gaseous. The reactivity, as well as the structure of the molecule, largely depends on temperature, so at a temperature of 110–119 °C sulfur begins to melt. When heated above 120 °C, cyclic molecules are converted into polymer chains; at 160 °C, this process begins to intensify (Galdina, 2011).

Sulfur has the ability to form a large number of allotropic modifications, this is due to the fact that when combined with each other, sulfur atoms form ring or chain molecules.

So, for cyclooctosulfur ($S8$) there are more than 20 different allotropic forms. Many allotropic forms exist in various isomeric states. According to the research of Markov (2015) it is possible to conditionally divide allotropic forms of sulfur into three groups: allotropic forms of cyclooctosulfur, polymeric sulfur and intermediate ones. Each form of sulfur has its own specific physical and chemical properties associated with the parent form (Uryev & Ivanski, 1989).

Sulfur reacts with many organic compounds, but does not dissolve in water, conducts electricity poorly, is resistant to aggressive media: solutions of acids and alkalis, resistant to temperature extremes, when modified sulfur is introduced into various binders, it gives them specific properties.

The main properties of sulfur at various temperatures are given in Table. 1.

Table 1. Properties of elemental sulfur

Index	Temperature	
	20 °C	150 °C
Density, g/cm^3	2.1	1.8
Compressive strength, MPa	12–22	–
Melting temperature	119	–
Viscosity, Pa*c	0.001	0.1
Heat capacity, kJ/kg	0.7	1.84
Color	yellow	yellow

The use of technical sulfur in its pure form is impossible due to its significant brittleness and the possibility of oxidation to sulfur oxide (IV) or sulfuric acid. To obtain a stable quality product, it is necessary to first obtain a modified sulfur. Sulfur is modified by introducing various additives into it (McBee & Sullivan, 1977).

One way to obtain modified sulfur is to mix molten sulfur with organic additives. For example, mixing organic polysulfides and sulfur, which are copolymers. Intensive mixing is carried out for 20–50 minutes in a hermetically sealed vessel until the product stabilizes. Changing the time and intensity of mixing depends on the amount of added sulfur.

For the production of sulfur asphalt concrete and sulfur concrete mixtures, various allotropic states of sulfur are used, for example, sulfur-bitumen binders and mixtures based on them, prepared using modified sulfur. Such mixtures have high strength characteristics, increased wear resistance, corrosion and chemical resistance, low water permeability, resistance to sudden temperature changes compared to traditional concrete and asphalt concrete mixtures.

2. Sulfur asphalt concrete

Sulfur asphalt mixes should be considered as a kind of asphalt mixes, in which the bituminous binder is partially replaced by sulfur, including modified one.

According to the requirements of the Ukrainian standard for asphalt concrete mixtures (Національний стандарт України ДСТУ Б В.2.7-119:2011), sulfur asphalt concrete mixtures do not differ significantly from traditional asphalt concrete mixtures made on the basis of petroleum road bitumen. The increase is observed only in terms of compressive strength at elevated temperatures, resistance to plastic rutting at high temperatures and resistance to wear rutting (Al-Hadidy, 2022a).

Abroad, sulfur asphalt concrete under the brand name Tiopave is considered one of the varieties of warm asphalts, which are characterized by lower temperatures for preparing, transporting, laying and compacting the mixture (AMC_S260. Specification Amendment).

Such mixtures can reduce energy costs in production by 10–20%, deliver mixtures over longer distances and extend the construction season.

When laying compacted sulfur-asphalt concrete mixtures, the use of modified sulfur creates the necessary temperature range, within which the effective compaction of the mixture is ensured.

According to a number of researchers (Vasilyev et al., 2014), the effect of sulfur on the properties of petroleum bitumen is determined by the following factors:

- interaction temperature;
- amount of added sulfur;
- structural type of bitumen;
- Interaction time.

The using of sulfur as an additive in asphalt concrete mixtures makes it possible to reduce the consumption of bitumen (the content of the sulfur component, as a rule, is 25–30 wt.%), reduce the temperature of preparation of asphalt concrete mixtures, and increase the wear resistance of road surfaces.

At 120 °C, sulfur melts and turns into a light-yellow liquid with a low viscosity (0.017 Pa s), which ensures high workability of sulfur asphalt mixtures.

At a temperature below the crystallization temperature of sulfur, a rather rigid framework is formed in the sulfur asphalt concrete mixture, which leads to the formation of a conglomerate structure of sulfur asphalt concrete (Le et al., 2022).

The optimal temperature regime for compaction of traditional asphalt concrete mixtures lies in the range of 110–120 °C.

Due to the fact that 119 °C is the critical temperature for the onset of sulfur crystallization, effective compaction of asphalt concrete mixtures with sulfur additives cannot actually be implemented according to the generally accepted scheme due to its features.

For the correct calculation of the composition of asphalt concrete with the addition of sulfur, assessment and prediction of the behavior of the material during operation, it is necessary to know the physicochemical processes that occur when sulfur is mixed with bitumen and its effect on the properties of the organic binder-bitumen (Vasilovskaya & Nazirov, 2011).

In the laboratory of the Department of Computer Technologies for the Construction and Reconstruction of Airports of the National Aviation University, studies of sulfur asphalt concrete were carried out for compliance with the requirements of State Standard of Ukraine (Національний стандарт України ДСТУ Б В.2.7-119:2011).

BND 90/130 grade oil road bitumen, as the most used for road construction, granulated sulfur of the Cherkassy plant, was chosen as the objects of study. The research results are shown in Table 2.

Studies of the effect of technical sulfur on the physical and mechanical properties of BND 90/130 bitumen according to State Standard of Ukraine (Державний Стандарт України ДСТУ 4044:2019).

With different percentages of technical sulfur.

Binder compositions, %:

1. Bitumen BND 90/130 – 75%

Modified sulfur ДСТУ 2181:1993 Сірка технічна модифікована. Технічні умови [Modified technical sulfur. Specifications] – 25%

2. Bitumen BND 90/130 – 70%

Modified sulfur ДСТУ 2181 – 30%

3. Bitumen BND 90/130 – 65%

Modified sulfur ДСТУ 2181 – 35%

As can be seen from Table 2, the addition of sulfur leads to a change in the basic physical and mechanical properties of petroleum bitumen. This indicates a change in the bitumen structure.

The nature of the change in the penetration index of oil bitumen from the sulfur content is extreme. Within concentrations up to 10%, the value of penetration increases from 81 to 84. The increase in penetration values is apparently due to the ability of sulfur at certain concentrations to plasticize the bitumen structure. This is due to the influence of the processes of interaction of sulfur with bitumen.

Table 2. Bitumen test results

Indicators	Binder compositions			
	Control (without sulfur)	1	2	3
Needle penetration depth 0.1 mm: at 25 °C at 0 °C	 81 23	 82 24	 83 25	 84 24
Extensibility, cm: at 25 °C at 0 °C	 58 4.2	 60 4.9	 62 5.1	 63 5.2
Softening temperature, °C	49	55	53	55
Brittleness temperature, °C	–17	–19	–21	–20

The dependence of the low-temperature properties of oil bitumen on the sulfur content is also extreme. Thus, the brittleness temperature of bitumen with the addition of sulfur up to 10% decreases from –17 °C to –20 °C, which is a positive effect.

Thus, the formation of organosulfur compounds as a result of chemical interaction contributes to an increase in viscosity and an improvement in a number of important physical and mechanical properties of sulfur – bitumen binders, which is an important factor in improving the quality indicators of bitumen, such as penetration, brittleness temperature. Along with this, the modification of bitumen with sulfur will ensure the qualified use of a by – product of oil refineries and involve additional resources in the production of binders in an amount of up to 35% of the mass of bitumen in the form of elemental sulfur (Doshlov & Kalapov, 2015).

So, with a sulfur content of up to 30%, the resulting crystalline aggregates are separated by thick layers of organic binder and are active fillers in the asphalt concrete mixture; with a sulfur content of up to 35%, bulk crystalline bonds are formed that can withstand significant loads, but are not restored upon destruction.

In this regard, in asphalt concrete containing sulfur, a concrete structure is formed, where sulfur acts as an active filler that actively (positively) affects the properties of this material and at the same time binds mineral particles and large grains. Bitumen, added to the mixture together with sulfur, envelops mineral parts and large grains, plays the role of a viscoelastic binder that gives asphalt concrete strength, flexibility, and increases fatigue strength (Gorbik et al., 2004).

Studies carried out using a scanning microscope show that in the structure of asphalt concrete, large and small grains of sand have a bitumen shell, and when bitumen cools, sulfur is located in the voids between mineral particles covered with bitumen. Sulfur penetrates into the pores and crystallizes there in accordance with their configuration. Thus, the sulfur acts as a void filler and replaces the amount of free bitumen normally found in the voids of the mineral portion.

The addition of molten sulfur to the mineral portion of an asphalt mixture or to bitumen increases the fluidity of that mixture so that such an asphalt mixture can be formed. At the same time, it is almost not necessary to compact. The hot mixture containing sulfur is very easy to handle.

Another feature of sulfur as a filler is the clogging effect. In contrast to conventional filler in the form of mineral powder, the introduction of sulfur, other things being equal, makes it possible to increase the density of asphalt concrete by 3–4 times. This is due to the fact that sulfur and organic binder at the time of enveloping the particles of stone material are in the same phase (liquid) and the organic binder is consumed only to cover the stone particles, and liquid sulfur, which is released from the emulsion, fills the pores and voids, firmly connecting them crystallization bond walls. Conventional mineral powder, which has a high specific surface area, takes up most of the organic binder to coat and bond its particles (Kumar & Choudhary, 2023).

The third feature of sulfur as a filler is that it acts as a reinforcing filler with astringent properties.

The temperature of mixing sulfur with bitumen should not exceed 130 °C. It should be taken into account that with an increase in temperature, a chemical reaction of sulfur and bitumen occurs – polymerization and a significant increase in the content of asphaltenes. At the same time, extensibility and penetration depth are sharply reduced, the softening temperature of the sulfur–bitumen binder rises, and it becomes brittle.

Aggregates of crystalline sulfur are able to form a spatial frame, which consists of interconnected smaller aggregates into a permanent spatial structure. These crystalline formations, interacting through highly structured layers of an organic binder, lead to a sharp increase in the rigidity of the intergranular space. In addition, with an increased sulfur content (more than 30% by weight of the binder), direct crystallization bonds between stone particles can also occur (Bai et al., 2022).

Studies of sulfur asphalt concrete for compliance with the requirements of National Standard of Ukraine (Національний стандарт України ДСТУ Б В.2.7-119:2011) were carried out using a sulfur bitumen binder.

As an object of study, asphalt concrete type "B" was chosen according to National Standard of Ukraine (Національний стандарт України ДСТУ Б В.2.7-119:2011) as the most used in road construction in Ukraine.

The composition of asphalt concrete type "B" is as follows:

– Granite crushed stone of fraction 5–20 – 48.5 %;
– Granite screening fraction 0–5 – 44.0 %;
– limestone mineral powder – 7.5 %;
– Binder 6.5 % (Bitumen BND 60/90 – 65%, modified sulfur – 35%).

The samples were formed according to the requirements of State Standard of Ukraine (Державний Стандарт України ДСТУ Б В 2.7-89-99).

The research results are shown in Table 3.

Table data 3 indicate that the introduction of technical sulfur into bitumen in an amount of 35% improves the physical and mechanical properties of asphalt concrete compared to the traditional composition without technical sulfur. This is especially true for strength indicators at 20 °C (R_{20}), at 50 °C (R_{50}) and at 20 °C in a water–saturated state.

The introduction of sulfur into the asphalt concrete mixture does not complicate the technological process of preparing the mixture and can be done in the following ways (Nosko, 2022):

I – sulfur is introduced into the organic binder at the stage of its heating to the operating temperature.

Table 3. The results of studies of the physical and mechanical properties of asphalt concrete type "B" with the addition of sulfur

Type of asphalt concrete, warehouse numbers	Medium density, g/cm^3	Porosity of the mineral skeleton, % by mass	Residual porosity, % by mass	Water saturation, % by volume	Swelling, % by volume	Compressive strength limit, MPa, at temperature °C			
						20 °C in a dry state R_{20}	20 °C in water saturation condition R_{20}	50 R_{50}	0. R_0
Fine-grained type "B"									
Control composition	2.36	17.5	3.4	1.8	0.35	3.8	3.3	1.35	12.5
I	2.35	13.6	5.0	1.2	0.90	2.5	2.06	1.1	12.3
II	2.22	18.5	4.2	0.5	0.75	3.47	3.25	1.11	13.0
III	2.37	16.4	3.6	1.05	0.80	3.9	3.7	1.40	12.2
Requirements (Національний стандарт України ДСТУ Б В.2.7-119:2011)		15–19	2–5	1.0–3.5	0.85–1.0	No less 2.3–2.4	–	No less 1.1–1.2	No more 12–13

II – sulfur is introduced into the mineral material at the stage of its drying and heating to the operating temperature.

III – sulfur is introduced directly into the mixture at the last stage of mixing.

3. Sulfur concrete

Rigid road construction with cement concrete pavement is one of the most durable. The estimated service life is 30–40 years. But in most cases, the service life is much less.

The problem of ensuring the durability of cement concrete pavements is complex and depends on the properties of the components that make up the concrete mixture, structural properties, technological factors, operating conditions and climatic influences (Gnateyko, 1990).

From the standpoint of physicochemical mechanics of dispersed structures, concrete can be characterized as a coagulation-crystallization capillary-porous conglomerate (composite) structure that hardens over time. The processes of cement hydration take years and affect its constructive and destructive changes under the influence of fluctuations in temperature, humidity and natural and climatic factors. This is accompanied by a change in the direction of increasing porosity, water absorption, deterioration of the physical, mechanical and rheological properties of the concrete mixture (Prikhodko & Vasilyev, 2014).

To slow down the processes of corrosion (Attiogbe & Ruzkalla, 1995), carbonization, and reduce the strength of concrete, various polymeric and other additives are introduced into its composition to obtain the so – called modified concretes (organosilicon, polymer-polyester, polymethyl methacrylates).

In this work, technical sulfur was introduced into the composition of concrete according to ТУ У 23.9–39089479–001:2017 Технічна сірка. Сіркобетон [Technical sulfur. Sulfur concrete] in the molten state as a thermoplastic binder to obtain sulfur concrete.

The composite material, which includes sulfur binder, inert fillers and aggregates, is sulfur concrete (Fontana et al., 1998).

Unlike the dense rocks that are used as building materials, concrete is less durable. By its nature, it has a highly developed inner surface, which is formed by pores, capillaries and microcracks. Their presence is due to the use of excess water in the manufacture of concrete mix. A water/cement ratio (W/C) of 0.13 is sufficient for cement hydration, but modern equipment for making and placing concrete requires a water/cement ratio of 0.38–0.5. Excess water forms a capillary-porous structure of cement stone, which contributes to the migration of water and chemically active substances from the environment into the concrete body during operation, which causes its corrosion (Baikon & Davis, 1985).

The presence of microdefects in the cement stone increases its porosity and reduces its strength, which causes various deformations during operation – peeling, chipped edges, cracks (Okumura, 1975).

The introduction of sulfur helps to reduce the capillary porosity of the cement stone, the water demand of concrete and increases the degree of cement hydration, thereby forming a new microstructure of the cement stone both in the main volume and in the microvolumes bordering the aggregate surface (Khamydullin & Gaynullin, 2012).

When sulfur is added, neoplasms appear on the cement stone, which grow together with the surfaces of the aggregate grains, due to the forces of physicochemical and chemical interaction, and at the interface with the aggregate from dense, durable rocks, so – called contact layers appear in the cement stone, which differ much more strong structure (Bratchun et al., 2008).

Sulfur concretes are sufficiently strong and corrosion-resistant materials that are widely used in countries where there are natural reserves of sulfur and sulfur ores. They have developed technologies for obtaining sulfur in the course of oil and gas refining. The method of impregnating products from ordinary concrete with a sulfur solution has not become widespread due to the fact that it is not technologically advanced and is associated with significant energy costs, which is associated with the need to heat sulfur to 120 °C and hold the concrete products. The exposure time of samples in hot sulfuric solution reaches 24 hours (Bratchun et al., 2007).

Analyzing the above data, in laboratory studies on the production of sulfur concrete, sulfur was introduced into the composition of concrete as a thermoplastic binder.

Experimental and laboratory studies of sulfur concrete included the determination of physical and mechanical properties.

Studies of sulfur concrete were carried out for compliance with the requirements (Державний Стандарт України ДСТУ Б В. 2.7–176:2008). Two concrete compositions were tested: one – sulfur concrete; the other – cement concrete grade B 40. Samples were formed according to the requirements of Concrete mixes: Test methods (Державний Стандарт України ДСТУ Б В 2.7-114-2002 (ГОСТ 10181-2000)).

The research results are shown in Table 4.

Table 4. Physical and mechanical properties of sulfur concrete

Name of indicators	Concrete composition	
	sulfur concrete	cement concrete
Compressive strength, MPa, aged, days 28	56.2	36.5
Frost resistance after 100 cycles	F 200	F 200
Density, kg/m^3	2400	2400
Bending tensile strength, MPa, aged, days 28	5.3	4.7
Water resistance	W10	W8

Table 4 data indicate that the introduction of technical sulfur improves the physical and mechanical properties of concrete compared to traditional cement concrete.

Conclusions

Conducted laboratory studies have established that technical sulfur has a number of properties that have a positive effect on concrete and asphalt mixtures. So, at 120 °C, sulfur melts and turns into a light-yellow liquid with a low viscosity (0.017 Pa*s), which ensures high workability of sulfur asphalt concrete mixtures.

At a temperature below the crystallization temperature of sulfur, a sufficiently rigid frame is formed in the sulfur asphalt concrete mixture, which leads to the formation of a conglomerate structure of sulfur asphalt concrete.

The absence in the composition of sulfur concrete of the usual components – cement and water ensures the formation of a dense material characterized by high frost resistance parameters that are not achievable for traditional concrete based on Portland cement. Sulfur concrete is actually a kind of polymer concrete, in which sulfur plays the role of a binder.

Sulfur concretes have a number of positive properties, which primarily include: rapid strength development associated with the cooling period of the mixture, relatively high compressive strength (up to 60 MPa) and bending tensile strength, wear resistance and water resistance, resistance to aggressive media, low water absorption.

The addition of sulfur as a modifier to the binder (the optimal amount is 35% by weight of bitumen) for asphalt concrete mixture improves its workability, increases strength up to 30%, increases the durability of asphalt concrete, increases its thermal and dynamic stability, water resistance and reduces the amount of shear deformations of pavement.

The low cost of technical sulfur and its sufficient quantity on the territory of Ukraine will ensure the cost-effectiveness of the preparation of concrete and asphalt concrete with its addition.

The development of an energy- and resource-saving technological process for sulfur modification and the involvement of the resulting product in the production of building materials and road surfaces will make it possible to obtain a significant economic and environmental effect.

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