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Mineral Additives Based on Industrial Waste for Modifications of Bitumen Polymers

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Mineral Additives Based on Industrial Waste for Modifications of Bitumen Polymers

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Abstract



Journal of Sustainable Architecture and Civil Engineering Vol. 1 / No. 32 / 2023 pp. 196-204 DOI 10.5755/j01.sace.32.1.32505 prompted scientists to use alternative raw materials as components of the construction industry. This study investigated the suitability of waste products from the coke industry as modifying agents for polymer bitumen. The by-product of the coking industry reaches a high level of silicon and is called silicon-containing additive (SCA), the composition of which is similar to the mineral solutions for bitumen. In this article, the results of using SCA as standard additives for modified polyethylene terephthalate (PET) with bitumen waste is presented. Using the Simdist boiling point distribution method, simulated high temperature distillation by gas chromatography, differential thermal (DTA) analyzes, and other methods of physic- chemical properties of bitumen after adding of SCA were studied. It was revealed that the connection and consumption of additives occurs because of the content of aromatic components, as well as an increase in the consumption of bitumen characteristics. Thus, the presented method for obtaining road surfaces based on PET waste and coke production waste meet the requirements of sustainable development, which implements both a significant use of industrial waste as secondary products and their processing into a new product using an energy-saving technology that reduces the consumption of raw materials and resources.

The increase in cost of carbonaceous materials, as well as significant costs of industrial waste, have

Keywords: bitumen, chemical industry waste, mineral additives, modifiers, road materials, silicon-containing additive.

It is known (Zhang et al. 2022) that bitumen is one of the most important components of asphalt concrete, characterizing the strength and service life of road surfaces. As a result of numerous studies of various characteristics of bitumen, it has been found that bitumen modification can minimize the problems associated with damage to road materials, an increase in the frequency of their laying, as well as maintenance and repair costs (Yaro et al., 2021).

Various polymeric materials are used as modifiers for bituminous binders, such as thermoplastic polymers (polyethylene, polypropylene, polyvinyl chloride, etc.), thermoplastic elastomers (styrene butadiene styrene (SBS), styrene isoprene styrene (SIS), etc.). However, the use of polymers according to Azahroosh et al. (2020) is limited due to the following disadvantages: their disproportionate effect on asphalt viscosity, incompatibility with the bituminous matrix and, most importantly, high cost.

In this regard, the use of waste that has no residual value and causes a serious environmental problem plays a leading role as a green investment movement (Santagata et al., 2007; Abdul-Rauf et al., 2010).

Waste rubber (Nongnard et al., 2004) and used car tires in the form of crumbs and rubber powder are of great interest for the modification of bituminous binders. Research by Adhikari et al., (2000); De Almeida Junior et al. (2012); Jamal et al. (2020) showed that waste rubber tires could be incorporated into an asphalt mix to improve asphalt pavement performance. Therefore, Mashaan et al. (2013) describes the improvement in the stability and adhesion of asphalt mixtures after the introduction of crumb rubber. However, the authors (Porto et al., 2019) concluded that a large number of factors could influence the characteristics and mechanical properties of the rubber/ bitumen system, such as rubber size and content, chemical structure, particle surface properties (granulation in ambient conditions, environment / cryogenic grinding of tire rubber), method and conditions for obtaining a mixture). Scientists believe that the rubber content of about 4% does not have a significant effect on the operational and mechanical properties of bitumen, and increasing the content to 20% is unsuitable.

Another waste used to modify asphalt mixes for non-rigid pavement construction is crushed concrete. The use of these wastes has a number of benefits, including increased pavement productivity, since it involves the production of new road materials by crushing old roads and using these materials as the base layer for a new asphalt pavement (Jung et al., 2020; Atisan et al., 2009; Milad et al., 2018). However, the use of recycled concrete crushed stone as a modifier requires the removal, destruction and transformation of concrete into a material of a certain guality and size.

The paper (Remišová et al., 2017) concluded that additives in the form of various fabrics, fibers, zeolites, etc. do not affect the structure of bitumen, therefore, modification does not occur, and however, a change in the properties of bitumen is observed.

There is considerable interest in plastic waste, such as polyethylene terephthalate (PET) waste, as modifiers. Our works (Aitkaliyeva et al., 2022) have previously shown the possibility of their use for modifying bitumen binders. High compatibility of PET with bitumen due to the formation of new oxygen-containing functional groups in the composition of new materials, as well as an increase in the content of asphaltene components in the composition of coatings, which indicates hardening with bituminous binders and an improvement in their viscosity-temperature stability.

Possible directions for further improvement of the properties of bitumen and polymer-modified bitumen are functionalization and development of new additional additives by Zhu J. et al. (2014). In this direction, developments in the field of new additives for compatibility and improvement of adhesion are useful.

Mineral clays composed of tetrahedral layers of silicate and octahedral sheets of hydroxide Martínez-Anzures et al. (2018) were studied as modifiers in the polymer of bituminous binders by Zapién-Castillo et al. (2016). These include montmorillonite, rectorite and kaolinite, which are

Introduction



important due to their effective use in asphalt modification technology (Hernandez et al., 2006; Yu et al., 2007). To reduce the cost of the resulting modified bitumen, some waste and by-products of production can be of great success, which can be used as an alternative to kaolinite and montmorillonite. Therefore, studies aimed at studying the possibility of the potential use of industrial waste as mineral additives for the modification of polymeric bitumen are relevant.

To fill a gap in research, as well as to improve the efficiency of using the bitumen/PET flex system described in our previous work (Aitkaliyeva et al., 2022), here we report the use of coke production waste as a mineral additive for modifying a polymer-bitumen binder.

Materials

Asphalt binder was road oil bitumen with 70/100 penetration- grade. The properties of the binder are presented in Table 1.

| Property | Requirement | Value |
|--|-------------|-------|
| Needle penetration depth, 0,1 mm at 25 °C | 70-100 | 73 |
| Ductility, mm | 70 | 70 |
| Softening point, °C | 43-52 | 52 |

Fig. 1 Laboratory material SCA

at room temperature

Methods

Table 1

The main

properties of bitumen

Table 2 Physical properties of the additive

| Physical properties | Value |
|--------------------------------------|-------------|
| Silicon content | 20,06 |
| Appearance | Bulk mass |
| Color | Light gray |
| Solubility in water | Not soluble |
| Mass fraction of moisture, % no more | 3,5 |

Polyethylene terephthalate (PET) waste, the main characteristic of which is presented in Aitkaliyeva G. et al. (2022) served as a polymer for bitumen modification.

A silicon-containing additive (SCA) as a modifying agent for the polymer of bituminous binders was studied. SCA - a production waste from the calcination of crude petroleum coke at the "UPNK-PV LLP" plant of Kazakhstan.

The sample, designated as SCA, can be seen in Fig. 1, the physical properties are presented in Table 2.

Study of the composition of the siliconcontaining additive

The composition of the additive was studied on an X-Ray Innov-X Systems instrument and an X' Pert MPD PRO (PANalytical) diffractometer.

The infrared spectra of the sample were obtained on a Bruker Tensor II IR Fourier instrument.

Preparation of modified bitumen and study of its main properties

To carry out the modification of bitumen, the method of physical mixing in one stage was used.

The base bituminous binder was heated to a fluid state of about 160°C, the PET modifier was added in an amount of 4% in the constant mixing mode, and the SCA was added for 10 min and continuously mixed for 3 h to complete the sample preparation. The amount of SCA introduced was 5, 10, 20, 30, and 40%.

The choice of this method is due to the advantage from a practical point of view (Porto M. et al. 2019), as it is simple to implement and involves a single step using classic paving equipment.

To identify the potential for practical application of the silicon-containing additive, the physicchemical and performance properties of initial and modified bitumen were investigated.

Study of the composition and properties of bitumen

To study the change in the compositions and properties of bitumen before and after modification, the methods Simdist Boiling point characterization/ distribution (ASTM D7169), High temperatures simulated distillation by Gas chromatography.

To study changes in thermochemical and physical parameters during heating of bitumen and its modified sample, the method of differential thermal (DTA) and thermogravimetric (TGA) analyzes were used, performed on a Q-1000D derivatograph in air in the temperature range from 20 to 1000 °C.

The dynamic viscosity and density of samples of initial and modified bitumen were evaluated at 100°C using a Stabinger SVM 3001 viscometer.

Study of the performance properties of bitumen

Studies of the penetration of bituminous binders were carried out according to standard methods in ASTM D-5, softening point (ASTM D36), ductility (ASTM D113).

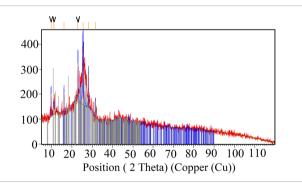
Study of the possibility of using SCA as a bitumen modifier

The main physical and chemical characteristics of SCA are shown in Fig. 2, 3 and Table 3. This additive has a fine grinding, which determines its good adsorbing properties for bitumen.

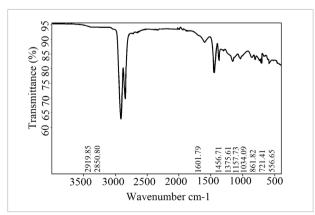
On the diffraction pattern, the silicon-containing additive (Fig. 2) is presented as a predominantly amorphous phase, as evidenced by broad peaks in the range. X-ray phase analysis revealed a high content of silica and aluminum, which is confirmed by the results of X-ray fluorescence analysis (Table 4).

The IR spectrum of coke ash (Fig. 3) contains peaks related to methylene and aromatic functional groups, and the presence of absorption bands in the range of 1100-100 cm⁻¹, characteristic of Si-O-Si, bands in the 665-670 cm⁻¹ can probably be attributed to the content of SiC₆H₅.

The presence of metal oxides in the composition of the SCA will have an activating effect on the formation of structure in bitumen and positively affect the ability of bitumen to adhere to the mineral substrate. In this regard, it was decided to use this additive – a waste product of the coke industry as a modifier for polymer bitumen binders.



| Ref. Code | Score | Scale Factor | Chemical Formula |
|-------------|-------|--------------|--------------------------|
| 96-810-4313 | 23 | 0.553 | Si 8.00 O 24.00 Al 4.00 |
| 96-152-5556 | 8 | 0.410 | 0 81.08 A l8.00 Si 28.00 |



Results and Discussion

Fig. 2

X-ray diffraction pattern of a silicon-containing additive sample

Table 3 XRD analysis

of SCA

Fig. 3

IR spectrum of coke production ash SCA



| _ | | |
|------|------|-----|
| | o b | |
| | - 11 | ~ ~ |
| - 19 | uD | |

Content of metals in SCA

| Element | Content, % | | | |
|---------|------------|--|--|--|
| Si | 20.06 | | | |
| Fe | 9.90 | | | |
| Al | 13.96 | | | |
| Ni | 1.48 | | | |
| Cu | 0.35 | | | |
| Zn | 4.28 | | | |
| Ca | 5,40 | | | |
| Pb | 1.34 | | | |
| Nb | 0.37 | | | |
| Mg | 0.54 | | | |
| Pd | 1.44 | | | |
| Ag | 2.47 | | | |
| Cd | 2.04 | | | |
| Sn | 2.32 | | | |
| Sb | 2.05 | | | |
| Remain | 32.00 | | | |

Sample

Boiling point, C

Fraction 1%

The amount of carbon at the start of boiling

Study of the composition and properties of modified PMB

The influence of the SCA on the properties of the modified bitumen are presented in Tables 5, 6 and Fig. 4. Table 5 represents the results of the distillation temperature distribution by the simulated distillation and gas chromatography (GC) method, which separates the individual hydrocarbon components in the order of their boiling points (Villalanti D.C. et al. 2006). The results show that the introduction of SCA into the composition of the bitumen polymer leads to an increase in the initial boiling point of the binder, which is caused by the weighting of the bitumen from C25 to C28.

When comparing the thermal curves of the decomposition of bitumen samples (Fig. 4), there is a difference in the kinetics of their oxidation in the temperature range of 210–900°C. On the example of thermogravimetric curve readings in the specified interval, a significant change in the weight loss mode of the sample without SCA is observed, losing most of its mass (72.7%), for modified bitumen with SCA, the weight loss decreases to 48.7%, which is consistent with the results of Table 5.

According to the results of the study of the composition and properties of bitumen before and after the introduction of SCA, it can be concluded that with an increase in the content of the additive in bitumen, an improvement in the thermal stability of bitumen is noted, as well as a weighting of its composition.

10%

405,8

435.5

C25

20%

408.3

440.9

C26

30%

418.6

451.9

C27

40 %

429,6

455.3

C28

5%

403.7

433.6

C25

Table 5

Results of hightemperature simulated distillation of bitumen samples

Table 6

Study of the physical and chemical characteristics of bitumen before and after modification

| Property | initial F | PET | SCA content, % | | | | |
|------------------|-----------|--------|----------------|---------|---------|---------|--|
| | | | 5 | 10 | 30 | 40 | |
| Viscosity, MPa*s | 3463,7 | 3894,4 | 8968,7 | 11278,0 | 12946,0 | 13782,0 | |
| Density, g/cm3 | 0,9559 | 0,9762 | 0,9890 | 1,0057 | 1,0098 | 1,0134 | |

Initial

401

425

C25

The results of the **Table 6** shows that the viscosity is related to the content of SCA in bitumen, therefore, the introduction of waste significantly increases the resistance to deformation of bitumen, since high viscosity is an indicator of high resistance to flow (Escobar-Medina et al. 2021).

Escobar-Medina et al. (2021) believe that an increase in viscosity is achieved due to the physicochemical composition of the modifier, its texture and surface area, because of which a network of fibers is formed throughout the bitumen matrix. The data in **Table 6** indicate that starting from a concentration of 5 wt. % viscosity more than doubles; therefore, our applied waste is also able to form a local network structure, serve as a support, reinforcing the bitumen matrix and improving its resistance to deformation. With an increase in the amount of SCA, an increase in viscosity is noted, however, it is important to remember that supersaturation with this additive is possible and the formation of large agglomerates characterized by negative properties in the asphalt mix. Thus, in work of Caro et al. (2016) it was found that at elevated concentrations of the additive, interaction between the fibers is induced, which leads to increased fragility of asphalt concrete mixtures.



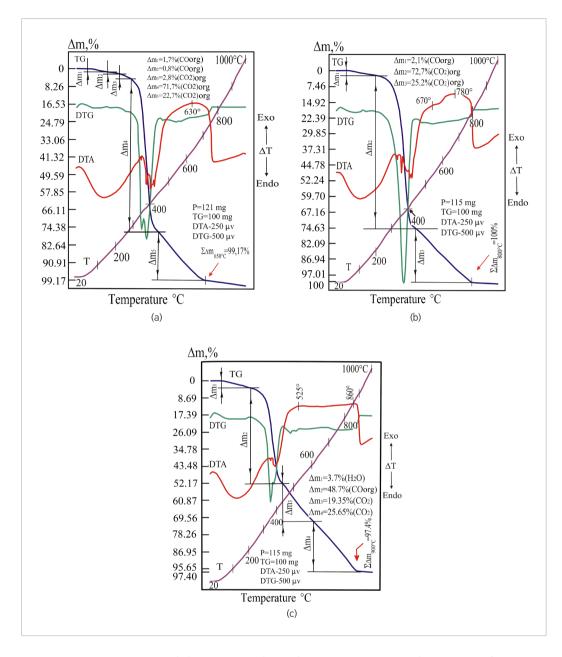


Fig. 4

Derivatogram of a sample of the bitumen: a – original; b – polymer modified bitumen in the amount of 4 %; c – polymer modified bitumen with the addition of SCA in the amount of 5%

An increase in viscosity and density may also indicate an increase in the content of aromatic components in bitumen (Jiang et al., 2020), which certainly has a positive effect on the cohesive strength of the binder. However, it should be noted that the IR spectra show no changes in the functional groups after the introduction of the SCA, which indicates the interaction of the additive and bitumen through physical adsorption.

Zhou et al. (2011); Liu (2009) think that in polymer-modified bitumens, the stabilizer improves the stability of heat preservation due to the formation of a stable adsorption layer at the interface between the bitumen and polymer phases.

Table 7 data indicates a decrease in penetration by about 20% with the introduction of SCA in an amount of 5%. With an increase in the concentration of SCA, the value of penetration decreases, which indicates an increase in the stiffness and hardness of the bitumen. It is known (Jeffry et al.



2018) that high stiffness leads to low susceptibility to high temperature. According to Brown et al. (2009), bitumen exhibits high resistance to cracking at penetrations above 30 mm when properly mixed and compacted.

According to the results of **Table 7**, it can be seen that because of the introduction of SCA, an increase in the ductility values of the modified bitumen is observed, so for a sample with a content of 5% SCA, a value of 40 mm is reached. However, as the concentration of the additive increases, a decrease in the values of plasticity is noted. The authors of Jeffry et al. (2018) attribute this to an increase in the adhesion of bitumen, which is caused by the formation of a strong bond between bitumen and the additive.

Table 7

Change in the main properties of bitumen with the addition of SCA

| Property | Value | | | | | | |
|---|-------|----|----|----|----|----|--|
| | PET | 5 | 10 | 20 | 30 | 40 | |
| Needle penetration depth, 0,1 mm at 25 °C | 62 | 49 | 46 | 41 | 40 | 38 | |
| Ductility, mm | 38 | 40 | 39 | 34 | 33 | 30 | |
| Softening point, °C | 56 | 59 | 60 | 62 | 63 | 64 | |

The softening temperature of bitumen after the introduction of SCA for 5% increased by 3 °C, with an increase in the concentration of introduced SCA, an increase in this indicator is observed. A high softening temperature indicates that bitumen can withstand a certain temperature level before the transition from solid to liquid phase (Jeffry et al., 2018). According to the data obtained, the addition of SCA increases the softening point, provides a low sensitivity of the modified bitumen to temperature.

The authors of Martínez-Anzures et al. (2018) believe that the addition of mineral additives according to Stokes' law can have a positive effect to reduce the difference between the densities of the continuous and suspended phases and increase the viscosity of the system, which improves the aging properties of the bituminous binder at high temperatures and provides improved stability.

Conclusions

In the present work, we studied the possibility of using SCA waste from the coke-chemical industry as a mineral additive for modifying polymer-bitumen binders. Based on the research, it can be concluded that the modification of PmB during the introduction of SCA is achieved through physical interaction, and increased rigidity and temperature resistance, which together indicates their improved performance properties, characterizes the resulting bitumen. The optimal concentration of the introduced additive was 5%, the increase of which will not lead to significant changes in the physical and operational characteristics of the binders.

We hope that the resulting binders based on waste will solve the problems of using PmB technologies in the road industry, while meeting the requirements of sustainable development. This is achieved by using a significant amount of industrial waste as secondary raw materials, processing them into a new product using energy-saving technology, which reduces the consumption of natural resources and prevents the formation of hazardous substances and accidents.

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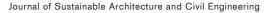
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Synthesis of thermosensitive polymers, hydrogels, production of new materials from production waste

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DANUTĖ VAIČIUKYNIENĖ

Dr. Professor

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Main research area

Synthetic zeolites, special building mixes, use of waste materials in geopolymer materials of cement systems, alkaline activated materials

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GULNAZ IBRAIMBAYEVA

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International Educational Corporation (Kazakh Leading Academy of Architecture and Civil Engineering)

Main research area

The development of composition and technology, the study of the properties of building materials

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