Using of ethylene-vinyl acetate copolymer for manufacture of road surfaces

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Abstract. The trend of European road companies to use an ethylene-vinyl acetate (EVA) copolymer is related to its ability to provide the necessary properties for the road surface, as well as its comparative cheapness and safety. The paper presents a study of bitumen modified by EVA, changes in the physicochemical properties of binders modified by EVA in concentrations from 1 to 10% and the influence of the number of vinyl acetate groups in the polymer composition on the properties of bitumen have been studied. Changes in physicochemical properties of bitumen in dependence on the molecular weight of EVA have been explored, as well as physicomechanical properties of asphalt mixtures with different contents of EVA have been evaluated.

1. Introduction

The regulation of the structural-group, dispersed composition of road bitumen by varying the technological parameters during the oxidation of the oil residue, as well as its compounding with polymers that increase the deformation resistance to high loads, do not allow obtaining a binding material with an acceptable level of adhesion [1, 2]. However, known adhesive additives have a number of disadvantages: high cost, thermal instability, harmful effects on the human body and the environment. Polymer additives, widely used in the practice of the United States and Europe, intended for residual bitumens, do not always have a positive effect on oxidized bitumens produced in Russia, and to combine them is usually a difficult technical task. One way to solve the problem is to introduce surfactants with polymeric materials into oxidized bitumen [3].

In the last decade, road organizations in Europe have shown increasing interest in the copolymer of ethylene and vinyl acetate (EVA) instead of the traditional styrene-butadiene-styrene copolymer (SBS) [4]. This is primarily due to the fact that EVA provides the necessary properties for the road surface: wide operating temperature range, chemical resistance to anti-icing agents, high impact resistance and adhesion. In addition, EVA is safe and relatively inexpensive. However, a methodological approach has not yet been developed to determine the permissible molecular weights and concentrations of EVA, as well as the influence of other groups on the physicochemical properties of bitumen of various natures and asphalt concretes based on them [4].
Direct modification of road bitumen binders with an ethylene-vinyl acetate (EVA) copolymer leads to a decrease in penetration, extensibility, an increase in the softening point, and an improvement in their low-temperature and adhesive properties. An increase in the content of vinyl acetate groups in EVA from 12.5 to 28% leads to an increase in the deformation-strength and adhesive properties of the copolymer, which is reflected in the properties of polymer-bitumen compositions: the higher the concentration of EVA and the content of vinyl acetate groups in it are, the lower the penetration is [4].

2. Methodology

In the conducted studies, bitumen with a group composition (wt %) was selected as the initial binding material: oils - 66.4, resins - 22.2, asphaltenes - 11.4. The physicochemical properties of bitumen are as follows: the depth of needle penetration (×0.1 mm) at 25°C - 120, at 0°C - 20; softening point 43°C, Fraas brittleness point - 14°C; extensibility at 25°C - 100 cm, at 0°C - 4.5 cm; adhesion with Pavlovsk-granite rubble - 3, with Pervouralsky rubble - 3, with reference standard sand - 3.

In order to obtain a homogeneous polymer-bitumen composition, a planetary mixer equipped with a thermostatic container was used. EVA was injected in a finely dispersed state (up to 20 microns) into dehydrated and heated to a temperature of 100°C bitumen in an amount of 1 to 10% by weight, then the temperature of the mixture was adjusted to 150°C. The content of vinyl acetate groups in the polymer varied from 12.5 to 28%. Complete dissolution of EVA in bitumen took place within 0.5-1 hour depending on the amount and molecular weight of the polymer. The advantage of EVA can be attributed to its ability to quickly and easily come unstiched in bitumen without the participation of a solvent, such as industrial oil.

3. Results and Discussions

When a modifier is introduced into oxidized bitumen, the copolymer particles absorb part of the maltene fraction, forming a spatial polymer structure. That happens due to associative relationships between oxygen-containing modifier fragments and existing structural quinone and asphaltene bitumen fragments. The homogeneity of the resulting composition was evaluated by the penetration values at 25°C in 4 sections of each sample [5]. With an increase in the copolymer concentration above 10%, phase inversion occurs with subsequent delamination, which was noticeable by the difference in penetration rates between sections.

The introduction of a low-molecular copolymer with a molecular weight of 2500 atomic mass units in bitumen leads to the formation of a spatial polymer structure that provides stability of the newly formed heterogeneous system [6]. With the content of 28% vinyl acetate in EVA, modified binder is a polydisperse system consisting of copolymer particles of different sizes. The introduction of an excessive amount of oxygen-containing copolymer groups into the bitumen can create a field of additional associative forces that provide sedimentation stability of the newly formed dispersed polymer system conjugated with asphaltenes. At a copolymer concentration of 2% with a vinyl acetate content of 12.5%, polymer particles of smaller sizes with their uniform distribution over the entire volume of bitumen are observed. Changes in the physicochemical properties of binders modified by EVA in concentrations from 1 to 10% have the same tendency, the difference is only in the absolute values of these indicators as shown in Figure 1.
Modification of an organic binder with an EVA copolymer leads to improvement of low-temperature and adhesive properties of bitumen, reduction of penetration, extensibility, and increase in the softening point. An increase in the content of vinyl acetate groups in EVA from 12.5 to 28 % leads to an increase in the deformation-strength and adhesive properties of the copolymer, which is reflected in the properties of polymer-bitumen compositions [7]. There is a sharp change in the penetration of bitumen: the higher the concentration of EVA and the content of vinyl acetate groups in it are, the less penetration is. On the other hand, the process of forming a spatial polymer structure in bitumen with the introduction of a copolymer is characterized by the effect of "interstructural plasticization" associated with significant swelling of EVA, which leads to the absence of a clear correlation between the values of the softening point and other bitumen properties.

**Figure 1.** Dependence of bitumen properties on the concentration of EVA with different content of vinyl acetate groups: a) softening temperature; b) depth of needle penetration at 25°C; c) brittleness point; d) extensibility at 25°C
point and penetration. An increase in the copolymer content of more than 2% in bitumen leads to a change in its structure from the sol-gel type with a penetration index from -1 to +1 to the gel type structure with a penetration index of more than 1. The possibility of such a structural transition by changing the concentration of the copolymer can be a useful and effective way to regulate the properties of the binder when it is used in different climatic conditions. Apparently, a large number of vinyl acetate links in the copolymer binds the maltene part of the bitumen in such a way that the actual values of mass loss and softening point after heating do not change much and do not exceed 1% and 3°C, respectively, which indicates a high resistance of the modified bitumen to thermal aging.

The resulting modified binders have a wider plasticity interval - more than 72°C compared to the original bitumen with a plasticity interval of 60°C. However, samples of polymer-bituminous compositions with a high content of vinyl acetate groups in EVA are a bit inferior in terms of softening point and extensibility to binders modified by EVA with a low content of vinyl acetate.

From the analysis of experimental data using the STATISTICA software package, the dependence of the main physicochemical properties of modified binders on the amount of EVA and the content of vinyl acetate in the copolymer was revealed. A significant contribution to the value of the softening and brittleness points is made by the EVA content (equations are presented in Figure 1). On the contrary, the penetration and extensibility of binders are greatly influenced by the content of vinyl acetate in EVA.

An important role in modifying an organic binder with a polymer is played by the molecular weight of the latter [8]. Introduction of 5% EVA with a molecular weight of 2500 into bitumen leads to a decrease in the softening point by 4°C and an increase of 15 units of penetration. The introduction of low-molecular-weight EVA into the bitumen also leads to a decrease in the extensibility and brittleness point, while the plasticity interval of these compositions is 5-15°C higher than that of the original bitumen. With an increase in the molecular weight of the copolymer to 12500, the softening point of the bitumen increases up to 51°C with a simultaneous decrease in penetration. Introduction of EVA with a molecular weight of 25000 into bitumen is manifested in a slight increase in the softening point (up to 55°C) and a favorable decrease in the brittleness point to -22°C.

The results of studies have shown that the improvement of the performance properties of binders is observed when introducing EVA containing 19% of vinyl acetate groups. This is due to the formation of a spatial polymer structure of polymer-bituminous compositions with a smaller size of copolymer particles and their uniform distribution over the volume.

For testing asphalt concrete mixtures prepared using modified bitumen as a binder with an ethylene-vinyl acetate copolymer as a prototype, the composition of hot dense asphalt concrete of type “B” grade II was selected as the most widely used in Russia. Mineral composition (wt %) of a sample of hot fine-grained asphalt concrete accepted for further research, is as follows: crushed stone 5-20 mm – 45 %, crushing screening - 22 %, river sand - 27 %, mineral powder - 6%, binding agent - 4 %. The optimal content of the binding agent in the mixture was determined based on the analysis of physicomechanical parameters of asphalt concrete [9].

The results of the effect evaluation of EVA content with a molecular weight of 25000, containing 19% of vinyl acetate groups in the binder on the performance properties of asphalt concrete [10] are shown in Table 1. From the analysis of the obtained data, it can be concluded that samples of asphalt concrete based on a modified binder have plastic deformation at low temperatures, the index of compressive strength decreases at 0°C, which will favorably affect the resistance to crack formation in the winter period of the coating’s operation.
Table 1. Physicomechanical properties of asphalt concrete mixtures based on bitumen modified with an ethylene-vinyl acetate copolymer

| Indicators                                             | EVA content in the binder, wt % |
|-------------------------------------------------------|---------------------------------|
|                                                       | 0   | 1   | 1.5 | 2   | 3   |
| Tensile strength, MPa, at temperature of               |      |      |      |      |      |
| 0°C                                                   | 7.6  | 7.1  | 6.3  | 6.6  | 6.8  |
| 20°C                                                  | 3.0  | 3.6  | 3.8  | 4.2  | 3.3  |
| 50°C                                                  | 1.08 | 1.32 | 1.36 | 1.40 | 1.15 |
| Water saturation, vol.%                               | 3.5  | 3.3  | 2.8  | 2.7  | 3.0  |
| Tensile strength of water-saturated samples, MPa      | 3.0  | 3.5  | 3.8  | 4.0  | 3.3  |
| Crack resistance                                      | 3.8  | 2.5  | 3.8  | 4.8  | 2.8  |
| The coefficient of temperature sensitivity            | 6.8  | 5.0  | 4.3  | 4.5  | 5.8  |
| Shear Resistance, MPa                                 |      |      |      |      |      |
| with shift at 50°C                                    | 0.22 | 0.25 | 0.26 | 0.27 | 0.24 |
| the coefficient of internal friction                   | 0.91 | 0.93 | 0.93 | 0.94 | 0.88 |
| The coefficient of water resistance                   | 1.00 | 0.93 | 0.94 | 0.94 | 0.96 |

With an increase in the concentration of EVA in bitumen to 2%, the strength of asphalt concrete mixtures increases at 20 °C. Thus, the degree of destruction of the binder structure, modified by EVA, when deformed under equal conditions is less than that of traditional bitumen. The coefficient of water resistance and water saturation of asphalt concrete based on a modified binder is higher in comparison with the unmodified one. The decrease in the strength of asphalt concrete containing more than 2% EVA is due to an increase in the viscosity of the binder. Since the newly formed polymer structure makes it difficult to wet the mineral particles of the composite material, there is a significant aggregation of the mineral powder, and most of the filler remains unbound by the binder.

A sharp decrease in the temperature sensitivity coefficient occurs when the content of EVA in bitumen increases to 1.5 %. A further increase in the EVA content increases the temperature sensitivity coefficient. Samples of asphalt concrete mixes with an EVA content of 1.5-2.0 % are characterized by low temperature sensitivity coefficients, i.e. they are equally well resistant to the formation of shear deformations at high temperatures and cracking in the cold season. The shear resistance of asphalt concrete mixtures based on a modified binder is higher than the shear resistance of a sample based on traditional bitumen, which will have a positive impact on the durability of the road surface.

4. Conclusions

Thus, modification of bitumen with EVA copolymer improves low-temperature properties, increases the temperature range of plasticity, strength, hardness and crack resistance of asphalt concrete. Increasing the content of ester groups up to 19% in the copolymer improves the adhesion of modified bitumen to the surface of the stone material, increasing the strength, water resistance, heat resistance, shear resistance and frost resistance of asphalt concrete mixtures.

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