

ВЛИЯНИЕ МОДИФИКАТОРОВ НА СВОЙСТВА БИТУМА И АСФАЛЬТОБЕТОНА**А.А. Гувалов, А.Д. Мамедов, Н.Т. Кахраманов**

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Работа посвящена поиску новых эффективных модифицирующих добавок, позволяющих целенаправленно регулировать технологические свойства битума и физико-механические показатели асфальтобетона на основе комбинаций модификаторов и битума, одновременно являющихся структурообразователем многокомпонентного асфальтобетона. Предложено использовать в качестве высокомолекулярных модифицирующих добавок в дорожных битумах блок-сополимер стирола и бутадиена – SBS и сополимер этилена-винилацетата – EVA. Изучено влияние модификаторов Europrene SOL T 6302 на основе бутадиенстирола и этиленвинилацетатный термопласт марки LG EVA EA 28400 на основе этиленвинилацетата на основные параметры полимерно-битумного вяжущего и эксплуатационные характеристики асфальтобетона. По мере увеличения содержания модификатора в битуме увеличивается глубина погружения иглы и повышается температура размягчения до 58 °С, снижается температура хрупкости до -32-35 °С, значительно расширяется диапазон рабочих температур дорожного покрытия. Исследования показывают, что сополимер SBS является термоэластопластом и дает сравнительно лучшие результаты, чем EVA. В результате исследования адгезионных свойств установлено, что добавление полиэтиленполиамин, содержащего 1,5% MasterLife PVA 100, значительно улучшает совместимость и адгезию битума к минеральным материалам. Однако при увеличении содержания MasterLife PVA 100 выше 2% происходят структурные изменения и ухудшается качество асфальтобетонных покрытий. Исследования показали, что применение полимерного модификатора улучшает многие свойства асфальтобетона. Так, в частности, улучшается тепло- и морозостойкость, износостойкость асфальтобетона в совокупности, влияющие на повышение эксплуатационного срока их службы. По результатам исследования выявлены оптимальные соотношения полимербитумных вяжущих (битум 95,5%+СБС 4%+ MasterLife PVA 100 1,5%) в составе асфальтобетонного покрытия, обеспечивающие их высокие эксплуатационные свойства.

Ключевые слова: битум, полимер, модификатор, прочность на сжатие, асфальтобетон**Для цитирования:**Гувалов А.А., Мамедов А.Д., Кахраманов Н.Т. Влияние модификаторов на свойства битума и асфальтобетона. *Изв. вузов. Химия и хим. технология.* 2021. Т. 64. Вып. 10. С. 98–104**For citation:**Guvalov A.A., Mamedov A.D., Kakhramanov N.T. Effect of modifiers on the properties of bitumen and asphalt concrete. *ChemChemTech [Izv. Vyssh. Uchebn. Zaved. Khim. Khim. Tekhnol.]*. 2021. V. 64. N 10. P. 98–104

EFFECT OF MODIFIATORS ON THE PROPERTIES OF BITUMEN AND ASPHALT CONCRETE

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The work is devoted to the search for new effective modifying additives that allow to purposefully regulate the technological properties of bitumen and the physicomechanical parameters of asphalt concrete based on combinations of modifiers and bitumen, which are at the same time a structure-forming agent of multicomponent asphalt concrete. It is proposed to use a block-copolymer of styrene and butadiene - SBS and a copolymer of ethylene-vinyl acetate - EVA as high-molecular modifying additives in road bitumen. The influence of modifiers Europrene SOL T 6302 based on styrene-butadiene and ethylene vinyl acetate thermoplastic brand LG EVA EA 28400 based on ethylene vinyl acetate on the main parameters of polymer-bitumen binder and performance characteristics of asphalt concrete has been studied. As the content of the modifier in the bitumen increases, the immersion depth of the needle increases and the softening temperature rises to 58 °C, the brittleness temperature decreases to -32-35 °C, and the range of operating temperatures of the road surface expands significantly. Research shows that SBS copolymer is thermoplastic elastomer and it gives comparatively better results than EVA. As a result of the study of adhesion properties, it was found that the addition of polyethylene polyamine containing 1.5% MasterLife PVA 100 significantly improves the compatibility and adhesion of bitumen to mineral materials. However, an increase in the content of MasterLife PVA 100 above 2% leads to a structural change and deteriorates the quality of asphalt concrete pavements. Studies have shown that the use of a polymer modifier improves many properties of asphalt concrete. So, in particular, the heat and frost resistance, wear resistance of asphalt concrete in the aggregate improves, affecting the increase in their operational life. According to the results of the study, the optimal ratios of polymer-bitumen binders (bitumen 95.5% + SBS 4% + MasterLife PVA 100 1.5%) in the composition of the asphalt concrete pavement, ensuring their high operational properties, have been revealed.

Key words: bitumen, polymer, modifier, compressive strength, asphalt concrete

INTRODUCTION

One of the promising directions in the modernization of bitumen-based pavements is the application of modifiers [1-5]. The use of polymeric substances such as EVA, Elvaloy, DST-30, butyl rubber and triple copolymer as a modifying additive, first of all, significantly improves their properties and strength [6-10]. However, the use of some high-molecular-weight modifiers causes the following technological problems: first, the uneven distribution of the polymer in the bitumen, and second, the insufficient adhesion of gravel to the bitumen coating, resulting in the collapse of the surface layer [11-16].

Analysis of the literature allows us to select the most widely accepted class of high-molecular com-

pounds for the production of polymer bitumen adhesive (PBA) [17-20]. For example, as a modifying additive, it is convenient to use easily copolymers of olefin and diene in a concentration that ensures that the main parameters of the polymerbitum adhesive are maintained at a technically reasonable level. In connection with the above, the current research is devoted to the possibility of using styrene-butadiene-styrene copolymer as a modifying additive in road bitumen to improve its performance properties.

MATERIALS AND METHODS

In this case, TN AZ 3536601.242-2015 (production of Baku Oil Refinery named after H.Aliyev) from BNB 50/70 oil bitumen; Europrene SOL T 6302

styrene-butadiene block copolymer – from SBS (manufactured in Ravenna, Italy); LG EVA EA 28400 brand ethylene-vinyl-acetate thermoplastic (made in South Korea); MasterLife PVA 100 (manufactured by BASF) was used. Europrene SOL T 6302 styrene-butadiene block copolymer has a specific gravity of 0.94 g/cm^3 , tensile strength of 20 MPa, viscosity of $4.0 \text{ Pa}\cdot\text{s}$ according to Brookfield. LG EVA EA 28400 brand ethylene-vinyl-acetate copolymer has a density 0.945 g/sm^3 , tensile strength of 3 MPa, elongation at break of 900%. The density of MasterLife PVA 100 is 0.93 kg/l , $\text{pH} = 8$, boiling point is $350 \text{ }^\circ\text{C}$, ignition temperature is above $160 \text{ }^\circ\text{C}$. Forms of polymeric substances for bitumen are given in Fig. 1.

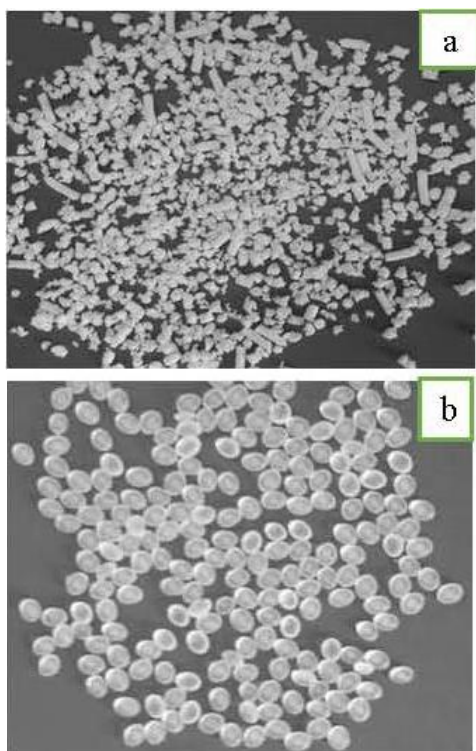


Fig. 1. Polymeric substances for bitumen: of a- Europrene SOL T 6302 brand SBS, b- LG EVA EA 28400 of brand EVA
Рис. 1. Полимерные вещества для битума: а- Europrene SOL T 6302 марки SBS, б- LG EVA EA 28400 марки EVA

In the laboratory, the modifier was mixed with bitumen at a temperature of $160 \text{ }^\circ\text{C}$ in a heating device shown in Fig. 2.

PBA softening temperature is determined by the method of KVS (GOST 11506-73). The brittleness temperature is measured in Fraas (GOST 11507-78). The depth of immersion of the needle at $25 \text{ }^\circ\text{C}$ is determined in accordance with GOST 11501-78. Adhesion with mineral aggregates is determined in accordance with GOST 11508-74. The stability of bitumen, assessed for changes in quality during long-term storage at high temperatures ($163 \text{ }^\circ\text{C}$, 5 h) is carried out in accordance with GOST 18180-72.

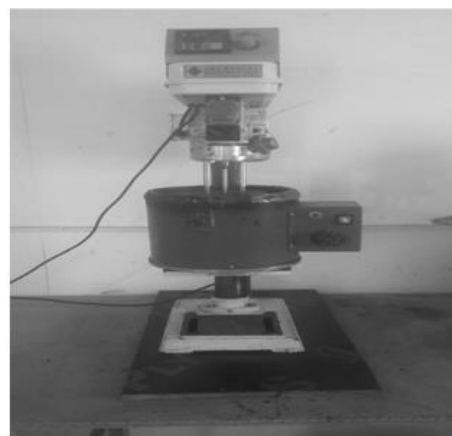


Fig. 2. Mixer with heating
Рис. 2. Смеситель с подогревом

RESULTS AND DISCUSSION

The main physical and mechanical parameters of PBA were studied in the study to determine the role of the polymer in the composition of PBA, as well as in the asphalt concrete mix. First of all, the effect of SBS and EVA content on bitumen performance was considered. As can be seen from Fig. 3, the depth of sinking of the needle, which determines its brand, increases with the amount of modifier in bitumen. In PBA obtained with the application of EVA in the amount of 2% of polymers, the depth of needle immersion is slightly higher than the results of SBS, but SBS shows better results in the range of 3-6%.

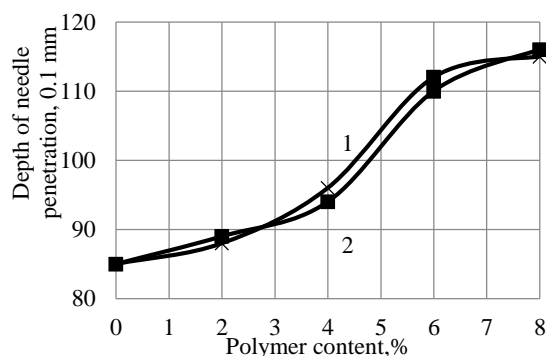


Fig. 3. Dependence of needle sinking on the density of polymers in polymer-bitumen adhesive: 1-SBS, 2-EVA
Рис. 3. Зависимость глубины проникания иглы от концентрации полимера в полимерно-битумном вяжущем: 1-SBS, 2-EVA

Although the main indicator of heat resistance is the softening temperature, which reflects the transition from the elastoplastic state to the viscous state, the effect of the amount of high-molecular compounds added to it has been studied. It was found that when the amount of EVA is 2-6%, the PBA softening temperature is $48\text{-}56 \text{ }^\circ\text{C}$, while the SBS-based PBA softening temperature is $50\text{-}58 \text{ }^\circ\text{C}$ (Fig. 4).

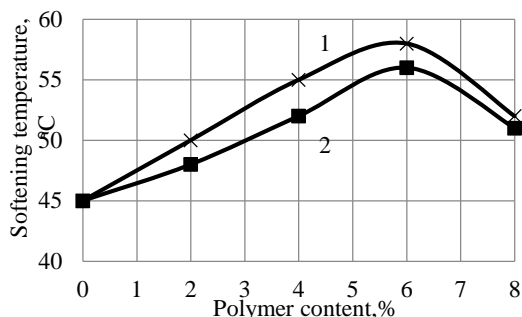


Fig. 4. Softening temperature dependence of the amount of polymers in the polymer-bitumen adhesive: 1-SBS, 2-EVA
Рис. 4. Зависимость температуры размягчения от концентрации полимера в полимерно-битумном вяжущем: 1-SBS, 2-EVA

The observed dependence is probably explained by the formation of a spatial polymer lattice in PBA. Then there is a slight stabilization and decrease. It seems that at first the polymers act as a fine filler, while the softening temperature and viscosity increase. Then, as the mixture reaches a critical concentration in the formation, the peak of the softening temperature due to the saturation of the mixture with the polymer is recorded. Subsequent increase in viscosity leads to excessive saturation of bitumen with high molecular weight compounds, which leads to a decrease in the softening temperature of bitumen. As can be seen from Fig. 5, when using 4-6% of polymers, EVA lowers the brittleness temperature of bitumen to -29-33 °C, while better results (-31-35 °C) are obtained when applying SBS.

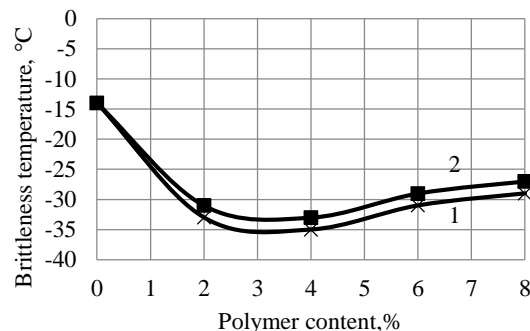


Fig. 5. Dependence of brittleness temperature on polymer viscosity in polymer-bitumen adhesive: 1-SBS, 2-EVA
Рис. 5. Зависимость температуры размягчения от концентрации полимера в полимерно-битумном вяжущем: 1-SBS, 2-EVA

As can be seen from Fig. 3-5, further studies have been performed with SBS because the main properties of the bitumen needle, such as sinking skin, softening and brittleness temperature, are higher than those of EVA when applying SBS.

Table 1 shows the test results of polymer-bitumen prepared with different amounts of SBS. As can be seen, the properties of polymer-bitumen fully meet the requirements of GOST R 52056-2003 for all experimental samples. Given that nitrogen-containing substances are used as additives to enhance adhesion properties [21, 22], BASF's MasterLife PAV-100 surfactant (polyethylene polyamine – PEPA) has also been studied. The "passive adhesion" method was chosen to study the adhesion properties.

Table 1

Physical and mechanical properties of polymer-bitumen adhesive

Таблица 1. Физико-механические свойства полимерно-битумных вяжущих

№	Quantity of PBA components, in % by weight			Penetration at 25 °C, 0.1 mm	Softening temperature, °C	Brittleness temperature, °C	Elongation at 25 °C, cm	Plasticity interval, °C	Adhesion of minerals with fine fillers
	Bitumen	SBS	MasterLife PAV-100						
1	100	0	0	85	45.1	-14	>100	59.1	№3
2	98	2	0	92	54.4	-32.9	>150	97.3	№2
3	96	4	0	112	58.8	-34.7	>150	103.5	№1
4	94	6	0	114	49.8	-30.7	>150	80.5	№2
5	95	4	1	112	49.8	-30.9	103	80.7	№2
6	94.5	4	1.5	116	64.8	-37.8	>150	106.6	№1
7	94	4	2	111	54.1	-34.5	>150	92.6	№1
8	93	4	3	110	50.7	-30.4	>150	84.1	№2
GOST R 52056-2003				91-130	50	-30	>30	-	№2

The results given in Table 1 show that the addition of the SBS modifier to bitumen improves its key performance. First of all, there is a significant increase in the plasticity range, which characterizes the operating temperature range. Bitumens with a wide plasticity range have a higher deformation ability, which increases the resistance to cracking at low temperatures

and the shear strength of the coating at high temperatures. When polymer is added to bitumen, its basic performance improves and, accordingly, the problem of cracking on the road surface is eliminated.

The results show that the adhesion strength of polymer bitumen with fillers is significantly improved,

and the results obtained with the use of 1.5% of MasterLife PAV-100 correspond to the control sample 1 № given in GOST 11508 (Table 1). In this case, other parameters of the adhesive are within the requirements of the standard.

One of the reasons for the premature collapse of the road surface is the wear and tear that occurs during its long-term operation. In this case, complex physical and chemical transformations occur in the structure of the material, which lead to the deterioration of its mechanical properties and a decrease in the serviceability of the coating. As can be seen from Table 2, the softening temperature of 4% SBS and 1.5% MasterLife PAV-100 mixed bitumen increases both before and after heating and is 58 °C and 64 °C, respectively. It is possible that this is due to oxidative dehydration reactions of naphthenaromatic compounds of bitumen with the formation of multi-ring aromatic molecules, which occur under the conditions of aging, and then their association with asphaltenes.

Table 2

The effect of the composition of the polymer-bitumen adhesive on abrasion resistance

Таблица 2. Влияние состава полимерно-битумного вяжущего на устойчивость к старению

№	Quantity of components, %			Softening temperature before heating, °C	Softening temperature after heating, °C
	Bitumen	SBS	Master Life PAV-100		
1	100	0	0	47	48
2	98	0	2	39	48
3	94.5	4	1.5	58	64
4	94	4	2	48	54
5	93	4	3	43	49

In order to determine the deformation resistance of the road surface, asphalt concrete mixtures with the same granular composition with different amounts of modifier additive on the basis of 50/70 bitumen were prepared. The test results obtained during the determination of the compressive strength of asphalt concrete samples are given in Table 3. Experimental results show that the strength characteristics of

PBA asphalt concrete are much higher than 50/70 bitumen-based asphalt concrete. When the amount of Master Life PAV-100 rises above 2%, there is a process of structural failure, which manifests itself as stratification and wear, which leads to a decrease in the quality of asphalt concrete pavement.

Table 3

Compressive strength limit for polymer asphalt concrete

Таблица 3. Предел прочности при сжатии для полимерасфальтобетонов

№	Quantity of components, %			Compressive strength, МПа	
	Bitumen	SBS	Master Life PAV-100	0 °C	50 °C
GOST 9128-97	-	-	-	Not more than 12	Not less than 1.2
1	100	0	0	8.5	1.2
2	96	4	0	10	3
3	99	0	1	5	2.5
4	94.5	4	1.5	10	4
5	94	4	2	12	2.7
6	93	4	3	11	1.4

As can be seen, polymer asphalt concrete with high performance properties is obtained on the basis of polymer bitumen adhesive made of optimal composition (bitumen 95.5% + SBS 4% + Master Life PAV-100 + 1.5%).

CONCLUSIONS

Thus, polymers used as modifiers in road bitumen give better results than Europrene SOL T 6302 styrene-butadiene-styrene (SBS) block copolymer LG EVA EA 28400 ethylene-vinyl-acetate (EVA) thermoplastic. The application of modifiers allows to obtain high-quality polymer-bitumen adhesive that meets the requirements of GOST R 52056-2003. Studies have selected the optimal composition of polymer-bitumen adhesive, which helps to improve the performance of asphalt concrete. In addition to the polymer-bitumen adhesive, the adhesion properties of the composition were increased by the inclusion of polyethylene polyamine, which increases adhesion.

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