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Ensuring the confection strength of adhesives based on SBS copolymers of various topologies

Abstract

This work is devoted to the improvement of a new building material for the road market - connecting bitumen-polymer tapes. The conditions for the use of such tapes have recently been significantly expanded both as a result of the harsh operating temperature conditions of the coating, and due to the need for road construction work at unsatisfactory temperature and humidity. The study attempts to assess the feasibility of using some brands of styrene-butadiene thermoplastic elastomers as a polymer base of adhesives to provide short-term increased strength of fastening of connecting bitumen-polymer tapes to the side surfaces of the crushed stone base for the period before laying a hot asphalt concrete mixture and forming a monolithic asphalt concrete coating in the seam area. The work on the study of a new type of material was hampered by the lack of appropriate technical standards for checking the quality of bitumen-polymer tapes for delamination and separation. Therefore, a high-hardness rubber compound based on styrene-butadiene rubber was chosen as a model solution. The data obtained confirmed the assumption about the effectiveness of various brands of SBS polymers to ensure the confectionery resistance of the base. The established dependencies were distributed and confirmed for highly porous concrete and embedded parts of metal fences.

Keywords: polymer mixtures, adhesion, coatings, connecting bitumen-polymer tapes

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Introduction

Styrene butadiene thermoplastic elastomers (also known as SBS polymers) are widely used to purposefully change the properties of various materials of a wide range of applications. For example, the use of only 3% of such a polymer can significantly change the properties of a multicomponent material based on it.[1] The main binder for the production of asphalt concrete (AC) are petroleum road bitumen (BND), which turn into a brittle state already at a temperature of minus 18 - minus 20°C. At the same time, in a significant part of the northern territories of Eurasia, the minimum temperature level is minus 28°C.[2] At the same time, the widespread use of polymer asphalt concrete observed in recent years has a positive effect on increasing the service life of road surfaces (up to 4 times), while significant problems still remain at the joints of such coatings.[3–5] This is the reason for the formation of low-temperature cracks already in the first years of operation. Cracks spontaneously appear at the junction of the surface layers of asphalt concrete pavement (AC) coatings.

Therefore, in this article, the issues of regulation of adhesive-cohesive interactions at the boundary of technological interfaces of various elements of road surfaces are considered. At the same time, it is necessary to take into account the processes occurring both at the junctions of parallel lanes of the road surface and in the transverse direction. In addition, the unification of road construction materials requires ensuring a satisfactory level of fastening in places adjacent to metal products (embedded parts, lighting supports, manholes and grilles), concrete (curbs, bumpers) and reinforced concrete structures (elements of bridge and support structures), etc.

As an option to improve the ratio of adhesive-cohesive forces in the butt joint zone, it is proposed to use bitumen-polymer tapes capable of ensuring the monolithic connection of two adjacent longitudinal strips of asphalt concrete coatings, even if the temperature of the layers differs significantly (by 80-100°C). It is worth paying attention to the situation in the docking seam (Figure 1) formed: in the absence of a docking layer (a), when applying an adhesive layer only on one side of the existing coating (b) and when forming a seam by placing a bitumen-polymer docking tapes at the contact point zone (c).

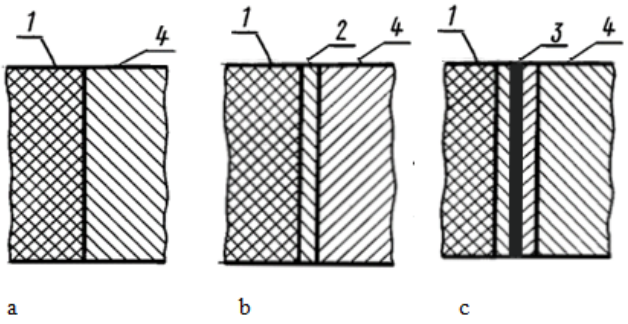

Figure 1 Interface seam.

1 - layer of "old/cold" coating, 2 - bitumen or emulsion layer, 3- bitumen-polymer tape penetrating into both boundary layers of the coating, 4 - "fresh/hot" layer of the coating.

It is obvious that the adhesive system (b), which provides reliable adhesive contact in the zone of boundary layers and at the same time has high cohesive strength, will have optimal strength properties. The presence of polymer components in it increases the cohesive strength of the seam zone and prevents its destruction. When a layer of hot ACM comes into contact with the surface of the docking tape, heat transfer occurs, and the tape material warms up quickly enough for its entire thickness (3-5 mm). After reaching the softening temperature, the bitumen-polymer composition changes from a solid/plastic state to a viscous/fluid one. This allows the components of the tape to envelop the particles of crushed materials protruding from the surface layer and fill the gaps between them in the surface layer AC.[6] Thus, the main contact zone in this case is the macro-roughness in the junction zone of the two layers AC.

The subsequent ramming with a bar and sealing with roller rollers ensure that air is squeezed out of the internal unclosed cavities of

the speaker and filled with a binder, which also has a positive effect on increasing the service life of the connecting seam (by reducing its micro porosity) and the entire road surface generally. The binder fills in the micro-roughness at the interface of the "old/cold" coating, thereby increasing the long-term strength of the joint. However, at the initial stage of preliminary fixing of the tape on the surface of the previously laid AC, there are no external heat sources. A real opportunity to strengthen the contact of the bitumen-polymer tape with the side surface of AC is a short-term impact at the point of contact, carried out by hitting the road worker's foot on the surface of the docking tape. Therefore, a logical option to increase the reliability of fastening the tape to the colder side surface of the speaker is the preliminary application of an adhesive composition that performs the functions of confection glue on the wide surface of the docking tape. It must hold the tape array in an upright position before and during subsequent technological operations. The purpose of this study was to evaluate the possibility of developing and using an adhesive composition for solutions based on Russian-made SBS polymers to improve the adhesive properties of connecting bitumen-polymer tapes. Variants of adhesive compositions for confectionery products based on various grades of SBS polymers are considered.

Experimental part

The issues of using styrene-butadiene thermoplastic elastomers as a polymer base for adhesive compositions for various purposes began to be comprehensively studied long before the start of large-scale industrial synthesis of styrene-butadiene thermoplastics.[2,7–9] Thanks to the active development of industrial production of SBS polymers, the emergence of new production facilities and the expansion of the range of marketable products, it is now possible to compare quality indicators and choose the best brands of SBS polymers to obtain adhesive compositions with the required set of properties. Obviously, this is typical even for areas where the use of fine organic synthesis products seemed unrealistic.

Materials

This study examines three brands of SBS polymers corresponding to various fields of application, the production of which has been mastered at JSC «Voronezhsintezkauchu».[10] Thus, styrene-butadiene thermoplastic elastomers of the DST L 30-01 brand are produced for the longest time and are widely used for modification of petroleum road viscous in accordance with the national standard GOST R 52056-2003 «Polymer-bitumen road binders based on styrene-butadiene-styrene block copolymers. Technical conditions». This polymer has the lowest possible melt flow rate, and the linear structure provides excellent mobility of macromolecules in the bitumen phase (Table 1). This makes it possible to provide the necessary level of cohesive properties of polymer-bitumen binders with a fairly low content of styrene-butadiene thermoplastic DST L 30-01 (about 3-5%).

Block copolymers of the linear structure of the SBS L 7322 brand have excellent elasticity and high elongation values. They differ from the typical DST L 30-01 brand in lower molecular weight and, accordingly, lower viscosity of solutions and melts. The strength characteristics of the adhesive seam, due to a decrease in its cohesive strength, are also affected by the presence of a two-block copolymer in the composition of up to 22% (Table 1), which, in this case, can perform the function of a polymer plasticizer. Due to the presence of the di-block, the manufacturability and the possibility of recycling multicomponent bitumen mixtures are improved.

Even more interesting is the SBS L 7417 brand. Having sufficiently low values of molecular weight (Mw) and especially high (up to 75%) content of two-block structures, it provides low viscosity values and high melt flow rate. At the same time, such a polymer microstructure is guaranteed to provide increased adhesive properties. An additional feature of this type of polymer is the high content of bound styrene in it (up to 37%), which increases the rigidity of the polymer itself and positively affects the final strength of attachment to hard surfaces (Table 1).

In order to ensure the required speed of applying the adhesive composition to the surface of the bitumen base and the qualitative distribution of components in the volume of glue produced at a non-core enterprise, the traditional mortar technology of production and application of the adhesive composition was considered.

In order to visually fix the uniformity of the adhesive layer application, as well as to reduce the cost of commercial products, a natural mineral filler of gray color with an average particle diameter of 10-40 microns, a mixture composition widely used in the practice of road construction, can be used in small quantities as part of the glue. Tallow pitch, a residual product of fractional distillation of natural raw materials in order to obtain tallow oil, can be used as a stickiness agent. It has an affinity for polystyrene blocks of thermoplastics and, due to the mobility of the low-molecular-weight limiting components included in the pitch, improves the adhesive interaction of the adhesive layer with the substrate. To increase the compatibility of the binder with the polybutadiene phase of thermoplastic elastomers, glycerin ether of natural rosin can be used in the composition. It is an ester of triatomic alcohol – glycerin and resin acids of rosin, which is a transparent vitreous mass. Its functional role is to increase the mobility of polybutadiene blocks and improve the adhesive ability of the composition. A rubber compound based on butadiene-methyl styrene rubber of the SCMS-30RP brand of increased hardness was chosen as a substrate modeling the physical and mechanical properties of a bitumen-polymer tape.[11] The main physical and mechanical parameters of the vulcanized rubber mixture obtained during vulcanization for 30 minutes at a temperature of 143°C are presented in Table 2.

Preparation of adhesive blends

Adhesive compositions based on SBS were made according to the traditional method for adhesives.[1] The composition of the adhesive is shown in Table 3.

Mixing of the adhesive components and its final homogenization was carried out using a laboratory dispersant T 25 digital ULTRA-TURRAX® in one stage for 30-40 minutes. Table 3 shows that in order to stabilize the strength properties of the adhesive, taking into account the high content of diblock, the amount of polymer grade SBS L 7417 was increased by 2%. The increase in the amount of plasticizer for DST L 30-01 is explained by the higher molecular weight of the polymer, which significantly exceeds this indicator compared to other polymers under study (Table 1). Consequently, the required level of rheological properties of such a highly viscous composition is possible with a relative excess of plasticizer.

Taking into account the absence of regulated state technical requirements for the definition and classification of elasticity and strength indicators of bitumen-polymer docking tapes, the assessment of the influence of various grades of SBS polymers on the properties of adhesive compositions obtained with their help was carried out on the basis of model rubber systems, the composition of which is presented in Table 3. This made it possible to assess the strength of the adhesive layer of connecting bitumen-polymer tapes by conducting technological tests in accordance with the requirements of the current national standards for rubbers and rubber products.

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As a target flexible fastening object, samples of an industrial batch of bitumen-polymer docking tape were used. With its help, asphalt concrete mixtures were fixed using as bases: a layer of asphalt concrete (complies with GOST 9128-2013 standard), a concrete curb stone (complies with GOST 6665-91 standard) and metal embedded parts of bridge fences (complies with GOST 25772-2021 standard).

Measurements of the mechanical properties

The bond strength for delamination and exfoliation was determined in accordance with the requirements of GOST 6768-75 and GOST 411-77. All tests were carried out at a temperature of 25 °C. The results of five measurements for each sample were averaged. The production of rubber mixtures of increased hardness on a laboratory mixer is extremely difficult. But only the most solid mixture could to a certain extent reliably simulate a real glued substrate. Therefore, a typical rubber of the manufacturer of the increased hardness grade 6272 was used, the properties of which are presented in Table 2.

Table 3 shows the influence of variable component composition on strength indicators for the considered model adhesive systems. Since the main operational indicator is precisely the strength for the two model substrates, it was recorded during the experiment and its averaged data for each variant are presented in Table 3.

The data in Table 4 allow us to illustrate the value of the adhesion strength of the substrate modeling asphalt concrete mixture with real substrates found in road construction: cement concrete, metal parts of signs, fences and connecting bitumen-polymer tapes. The noted data no longer characterize the properties of the adhesive, but illustrate the strength indicators of the entire adhesive compound as a whole, taking into account the properties of substrates of various natures.

Table 1 Physical and mechanical parameters of the used SBS polymers

The name of the indicator	SBS L 7417	SBS L 7322	DST L 30-01
Density at 20 °C, g/cm^3	0,93-0,95	0,93-0,95	0,93-0,95
Molecular weight (typical values), Mw 103	47	60	80
Content of 1.2-links, %	15-Nov	15-Nov	15-Nov
Content of the diblok,%	67-75	22-Dec	13-17
Bound styrene content, % (typical values)	37	30	30
Melt flow rate at 190°C, load 49.05 N (5000 kgf), g/10 min	16-25	9-Mar	0-1
Shore A hardness, conventional units	80-92	69-81	67-77
Elongation at break, % not less	250	800	700
Conditional tensile strength, MPa not less	1,7	10,0	14,7
Dynamic viscosity of 25% toluene solution at a temperature of (25 ± 0.1) ° C , sPs (typical values)	300	1200	5500
Kinematic viscosity of 5.23% solution in toluene at a temperature of (25 ± 0.1) ° C (typical values), mm^2/sec	5,0	7,0	14,0
Ash, % StCa/SiO_2	≤ 0,3/≤ 1,2	≤ 0,3/≤ 1,2	≤ 0,3/≤ 1,0

Table 2 Physical and mechanical parameters of the rubber compound

Brand of rubber compound	Type of rubber	Conditional tensile strength, MPa	Elongation at break, %	Shore A hardness
6272	SCMS-30RP	4,4	200	70-85

Table 3 Ratio of strength indicators of model adhesive systems depending on the composition of adhesive compositions

Brand of polymer adhesive base	Content of components, mass. %			Bond strength of model adhesive systems, kN/m
	SBS	Tallow pitch	Glycerin rosin ether	
SBS L 7417	10	2	2	0,85
DST L 30-01	8	3	2	1,36
SBS L 7322	8	2	2	1,38

Table 4 Indicators of the bond strength of the model substrate with substrates of various nature, kN/ m

Brand of polymer adhesive base	Bond strength of the model substrate with substrates of various nature, kN/m		
	Bitumen-polymer tape	Highly porous concrete	Rough metal
SBS L 7417	1,4	1,6	0,75
DST L 30-01	2,5	2,0	1,25
SBS L 7322	1,8	1,5	0,85

Results and discussion

The need to ensure the manufacturability and safety of using glue of a certain composition in the process of making the tape is extremely important. The use of methylene chloride as a solvent, which is simultaneously a solvent for the polystyrene and polybutadiene phases of thermoplastic elastomers, provides the glue of this composition with incombustibility due to the presence of chlorine atoms in the solvent. At the same time, the toxicological parameters of methylene chloride are significantly lower than that of toluene, traditionally used in this capacity. A feature of mortar compositions is the ability to ensure the penetration of high-molecular components of the adhesive into the surface layer of real bases. In the case under consideration, it was the inner part of the wide plane of the bitumen-polymer tape with a size of 50 * 5. The highly porous concrete surface and uneven edges of the metal embedded parts of the fences indicate the traditional level of fastening for this composition (Table 4).

Previously, it was found that for adhesives based on SBS polymers, polystyrene prevails in the volume of the adhesive film, while polybutadiene prevails on the surface.[9] This circumstance, as well as the structure and properties of the substrate surface, can be explained

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by the increased surface interaction for compositions based on all the studied polymer grades with both a highly porous concrete surface and a plastic surface of a bitumen-polymer composition. It is also known that for structural-mechanical transition zones, the primary feature of the classification is the thermodynamic incompatibility of the adhesive and the substrate; the secondary is the micro and macro relief of the contacting surface of the substrate; the tertiary is the viscous state of the adhesive, which can be solutions and melts of polymers and oligomers. Finally, the third feature is the curing processes of the adhesive after its penetration into the porous structure of the substrate (Figure 2).

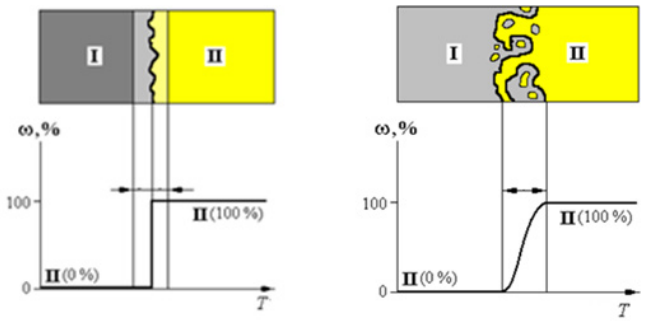

Figure 2 Diagrams of the contact area in the transition zone of incompatible (I) and compatible materials (II) and concentration profiles.[12]

T - the thickness of the boundary layer in conventional units, ω- the percentage of saturation of the boundary layer with adhesive components.

These processes can be associated with: a decrease in temperature and, as a consequence, the crystallization of the glue, with the transition of the glue to a vitreous state, with chemical reactions of glue curing, etc.[12] At the same time, the number of crystallizing resins in the composition of the adhesive under study is not sufficient for brittle destruction of the system under study in the temperature conditions of road construction work. It is necessary to further clarify that in the case under study, the need to ensure only a short-term fastening of the docking tape on the surface of an inhomogeneous dusty substrate is considered. The bonding strength in this case is designed to ensure a stable position of the bitumen-polymer tape profile on the side surface of asphalt concrete (more often vertically, but sometimes at an angle of 45-60°) during the time required for the unfolding and distribution of asphalt concrete or polymer asphalt concrete mixture butt-to-butt with bitumen-polymer tape and its rolling in accordance with the traditional road rolling scheme ice rinks. The deformation component of the strength of a complex adhesive joint in this case is associated with interfacial molecular bonds. Obviously, the more energy is spent on deformation and subsequent destruction of the adhesive joint, the stronger the adhesion at the glue/substrate interface will be. Consequently, an increase in the molecular interaction leads to an increase in the deformation component of the adhesive strength. The deformation and strength properties of multilayer joints depend on the deformation properties of each of the components of the adhesive joint. This probably explains the similar bonding strength of adhesive systems formed on substrates of different nature (highly porous concrete and bitumen-polymer tape) and polymers so different in composition and structure (SBS L 7322 and SBS L 7417).

Conclusion

During the research, the polymer base of the adhesive composition used to ensure the strength of the adhesive composition of the bitumen-polymer tape and asphalt concrete coating, as well as concrete and metal parts, was varied. Due to the lack of regulatory and technical documentation for testing bitumen tapes, a rubber mixture based on styrene-butadiene rubber was chosen as a model system. As a result, the required level of confection strength was provided for adhesive compositions based on all the studied grades of SBS polymers. The composition of the adhesive composition used indicates its effectiveness even at sufficiently low temperatures. Research in this direction will be continued.

Acknowledgments

None.

Conflicts of interest

The author declares that there is no conflict of interest.

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