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Technology for the production of road bitumen modifier using aluminosilicate microspheres extracted from ash and slag energy waste

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Abstract. The results of research on the development of technological solutions for the production of a new rubber-bitumen binder for asphalt concrete based on the use of industrial waste: used car tires, used car oil and microdispersed aluminosilicate spherical particles obtained from the processing of ash and slag waste (ASW) from thermal power plants are presented. The proposed technological solutions make it possible to obtain high-quality polymer additives for modifying the properties of road bitumen. The elements of novelty of the developed approach include the use, to obtain a granular modifier, of micro-sized hydrophobized aluminosilicate spheres, which are extracted as an additional product during the complex processing of ASW. The positive economic efficiency of technological solutions is ensured by the use of large rubber crumb (more than 8 mm) or rubber chips, their devulcanization together with hydrocarbon fractions of used engine oil and petroleum bitumen at a given temperature to form a gel-like mass, which is further subjected to mechanical grinding in a mill.
Keywords: rubber bitumen binder, modifier, road bitumen, aluminosilicate microspheres, hydrophobization

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1. INTRODUCTION

Bitumen is an important organic component of asphalt concrete mixtures used for highway pavement construction. Bitumen use as a binding material is due to its availability as a large tonnage refinery waste, as well as its ability to provide bonding of the mineral components of the mixture to improve the performance of the road pavement. At the same time, petroleum bitumens have a number of significant technological disadvantages: relatively low adhesion properties and lack of elasticity, which negatively affects the quality and durability of road surfaces. Which is especially important for areas with difficult climatic conditions and increasing traffic intensity [1, 2] (Fig. 1).

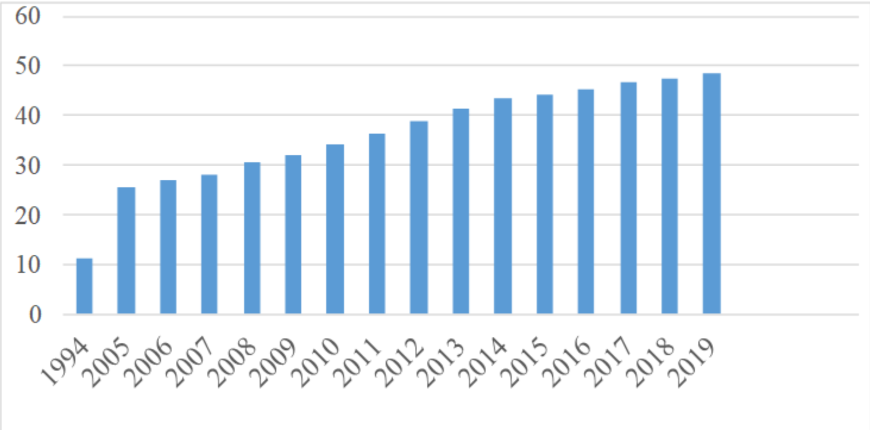

Fig. 1. Number of passenger cars in Russia, mln.

Analysis of researchers' works [3-7] shows that improvement of quality and durability of asphalt concrete pavements is possible with the use of modified petroleum bitumen having high adhesion, strength and deformation characteristics.

The most common types of such modifying additives are some types of high molecular weight compounds - elastomers, thermoplastics, thermoplastic elastomers. Bituminous binders using polymers are widely used in the territory of the European Union countries [8-12]. For example, in Germany and France, polymer-bitumen binders with the use of SBS type thermoplastic elastomers are used in road construction. Their share in the market of modifiers reaches 40%. The Czech Republic and Slovakia have also successfully used this type of binder: for example, the Czech company Paramo uses Carifex SBS thermoplastic elastomers. In addition, Motoplast-65 ethylene vinyl acetate copolymers as well as polyethylene and propylene (Appolobit, Slovakia) are used as modifying additives in bitumen. In Italy, elastomers (PR-PLAST type) and atactic polypropylene are generally used in modified petroleum bitumen. Shell produces a modifier "Kraton D", which is also used in the Russian Federation. However, the use of modifying additives is constrained for a number of reasons: the complexity of the technological process; relatively high cost of production; instability of the quality of the initial binder, negatively affecting the final product [13-16].

An alternative technical solution to polymer bitumen is the use of crumb rubber as an additive in road bitumen mixtures. The priority of the use of crumb rubber in the composition of asphalt concrete is provided by an almost unlimited supply of raw materials obtained from the processing of waste car tires; simultaneous solution of problems of both resource conservation and improvement of the environmental situation; relatively low cost [17]. The introduction of crumb rubber into the bitumen mixture is possible by "dry" and "wet" methods. The first one is considered obsolete and consists of adding crumbs in the form of mineral powder. More progressive are "wet" technologies, in which crumb is introduced into bitumen either in chemically modified form in the presence of certain reagents, such as lead and zinc oxides; or by thermomechanical method with preliminary devulcanization of crumb rubber and subsequent mixing of the mixture at high temperature. The result is a rubber-binding compound with high elasticity and a wide operating range [18].

The size class of crumb rubber used in Russia and other countries to modify road bitumen varies significantly. For example, the "UNIREM" technology uses a fraction up to 0.9 mm, "BITREK" - up to 3.0 mm, "BRK-IGU" – 5-7 mm. In Europe, crumb rubber with a size class of 1.25-2.0 mm is mainly used. With regard to the use of smaller fractions of crumb rubber (less than 1 mm), the opinions of experts differ: studies show both positive and negative results of the effect of the additive of such size on the quality of the final composite [19-22].

In a number of countries (USA, European Union, South America, etc.), a significant number of studies have been devoted to the issues of improving the quality of roadbeds based on petroleum bitumen modified with crumb rubber [23-27]. A number of companies, such as "Viafrance" (France), "Massenza" (Italy), "Crafko" (USA), demonstrate successful practical experience in the use of crumb rubber in the construction and repair of highways [28].

The experience of research and production association "Infotech" in using BITREK technology, which confirmed the advantage of the "wet" method, is interesting. But this solution also has a number of disadvantages associated with the impossibility of long-term storage and reheating of modified bitumen, which geographically limits the use of this composition; the presence of inhomogeneous inclusions in the composition of the resulting bitumen, which leads to clogging spray nozzles at the filling plant; as well as a relatively high initial cost of the resulting modified bitumen, which limits mass application. Despite this, the technology has found widespread use. Thus, about 15 million square meters of top layers of street and highway pavements in Moscow and about 0.5 million square meters in the cities of Moscow region and other Russian regions were repaired with rubber asphalt concrete of various types based on BITREK binder. A number of bridges were paved, including the Avtozavodsky Bridge, Khlebnikovsky Bridge over the Klyazma Reservoir (Moscow Canal) and Oktyabrsky Bridge over the Volga River in the center of Yaroslavl. In 2012, a section of the R-112 road near the settlement of Rastovtsy (Taldomsky District, Moscow Region) was inspected, where in the summer of 1998, a B-1 type mix with BITREK binder was laid. The Commission had found the site to be in good condition after 14 years of road use. There is damage in the form of temperature cracks with minimal opening (less than 1 cm). The levelness of the pavement meets the requirements of Russian building regulations. At the same time, the traffic intensity exceeds the norm of the road of the 3rd technical category on average 2-3 times.

Technological research of "Infotech" in recent times was aimed at finding the optimal bitumen/rubber ratio, which allows to maximize the use of rubber properties and, on the other hand, to avoid complications in the technology of asphalt concrete mixes (ACM) paving, according to the standards of GOST 9128-2013. When the modifier was added up to 30 % instead of equal part of bitumen, the strength of the specimens at 20ºC increased and reached 4.0 MPa. When the modifier proportion was further increased, the porosity of the asphalt concrete specimens increased, resulting in a decrease in strength. The tests were performed on ACM of B2 grade according to GOST 22245-90 (Table 1).

Table 1. Characteristics of the composite binder (according to "Infotech").

Characteristics	Bitumen / modifier ratio					
	100/0	90/10	80/20	70/30	60/40	50/50
Strength at 0ºC, MPa	11.3	11.0	10.3	8.1	8.2	8.2
Strength at 20ºC, MPa	2.4	2.7	3.4	4.0	3.8	3.2
Strength at 50ºC, MPa	1.3	1.4	1.6	1.8	1.7	1.7
Water resistance, units.	0.86	0.87	0.88	0.88	0.86	0.86
Porosity, %	17	16	16	15	17	19
Penetration at 25ºC, 0.1 mm	102	110	119	125	130	137
Penetration at 0ºC, 0.1 mm	32	36	68	90	100	111
Ring and Ball Apparatus, ºC	44	45	46	49	52	54
Brittleness temperature, ºC	-18	-22	-25	-28	-29	-30

When the modifier content in the binder increases more than 30%, the viscosity of ACM increases significantly, which does not provide normal compaction during the formation of samples, and, consequently, increases porosity and reduces strength.

In addition to the above, the use of modifiers based on crumb rubber is associated not only with the problems of increasing the cost of bitumen due to the need to obtain crumbs of significantly small fractions in the mechanical grinding of worn-out tires, but also with the difficulty of achieving an effective mixing of crumb rubber with bitumen to obtain a consistent quality (primarily in terms of elasticity) rubber-bitumen binder.

Thus, the discussed scientific and technological task remains relevant, which makes it necessary to continue the research.

2. METHODS AND MATERIALS

The novelty and peculiarity of our research is the combination of three components used for the first time in the developed bitumen modifier MD-01: used machine oil; enlarged fraction of rubber crumbs obtained at utilization of used automobile tires; aluminosilicate microspheres extracted from ash and slag of coal electric power plants. Since all the proposed components are products of waste recycling, at the same time the actual ecological problem of reducing the level of environmental pollution is also solved.

Technological processes were worked out on the experimental stand in the laboratory of the development of secondary resources utilization technologies at the Polytechnic Institute of Far Eastern Federal University (FEFU) in accordance with the general scheme presented in Fig. 2.

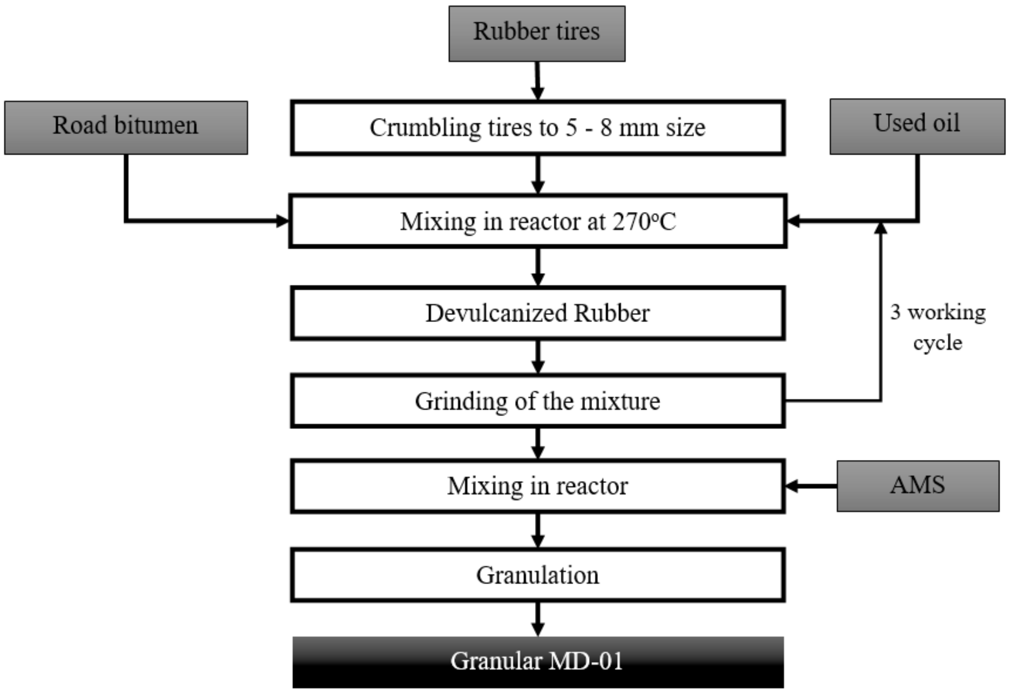

Fig. 2. Technological scheme for production of bitumen modifier MD-01.

The main elements of the technological scheme are:

- mechanical shredding of tires to obtain rubber crumb of 5-8 mm coarseness class (we used such a coarse fraction for the first time, which not only simplifies the technological process, but also leads to a significant reduction in the cost of production of the modifier);
- removal of metal or textile cord;
- mixing of crumb rubber with waste automotive oil and fusible bitumen in a heated reactor for 3 hours at a given temperature for devulcanization of rubber;
- grinding of the heated mixture on a disk eraser to obtain fine rubber devulcanizate;
- use of aluminosilicate spheres in obtaining granular modifier of long storage (application of this component in obtaining modifier is the author's novelty of technological approach).

3. RESULTS AND DISCUSSION

Lime flour, activated mineral powder used in cement production, and aluminosilicate microdispersed spheres (AMS) were tested in the selection of microdispersed additive in liquid modifier – Fig. 3.

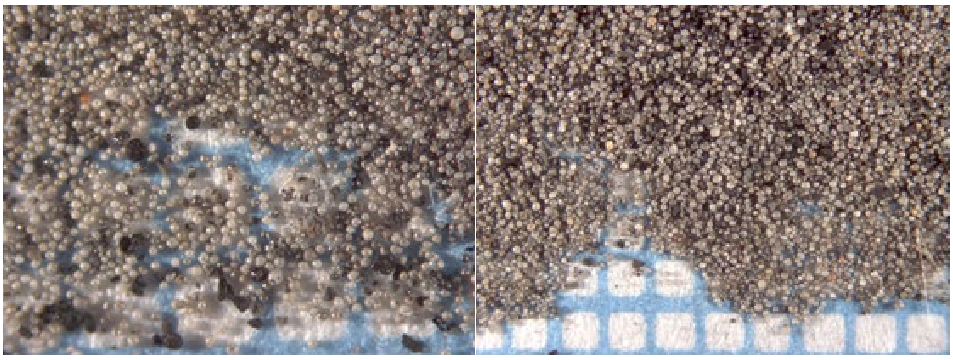

Fig. 3. Aluminosilicate microspheres of size grade from 0.071 to 0.1 mm.

AMS were obtained from ash and slag waste generated by coal combustion at thermal power plants. Compared to other fillers, microspheres were preferred in terms of particle size distribution uniformity, mix plasticity, cost and scarcity in the local market. The method of electron microscopy revealed a variety of glass spherical grains (spherules, microspherules), differing in size (from 11 to 125 microns), color, surface micromorphology and composition (Fig. 4). The obtained microspheres have predominantly aluminosilicate composition (> 75 %) with admixture of Ca, Mg, K, Na, Mn, Fe, Ti, S.

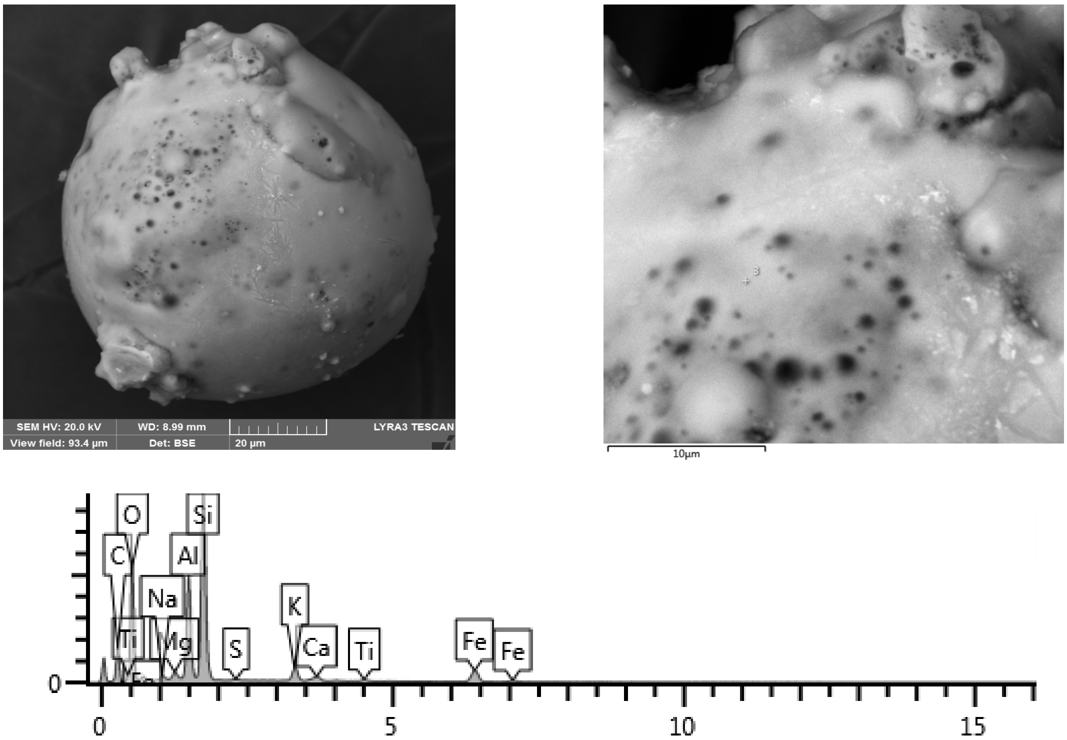

Fig. 4. SEM image of aluminosilicate microspheres, surface fragment and EDS spectrum of spherulas composition.

Previous studies have shown the possibility of effective extraction of AMS from fine fractions of ash and slag wastes from coal-fired TPPs by flotation method (Table 2).

Table 2. Results on obtaining concentrate of AMS from ash and slag wastes of coal-fired TPPs.

Class coarseness, mm	Fractional weight		Mass of fraction after flotation, g	Microsphere content in the fraction after flotation	
	g	%		g	%
+2.0	27.5	2.8	-	-	-
–2.0 +1.0	64.0	6.4	-	-	-
–1.0 +0.5	73.0	7.3	-	-	-
–0.5 +0.2	216.5	21.8	0.02	17	<0.01
–0.2 +0.1	186.5	18.8	0.04	80	0.03
–0.1 +0.071	90.0	9.1	0.04	75	0.03
–0.071	336.5	33.8	<0.01	85	<0.01
Total	994.0	100	0.11	257	0.06

Two types of microspheres, dry and hydrophobized, were tested. Hydrophobization of microspheres was carried out on a special unit in hot gases of heavy hydrocarbons. After hydrocarbon gas treatment, the microspheres were mixed with the liquid modifier uniformly throughout the volume in a shorter time and without forming non-impregnated modifier pellets. Additionally, the use of hydrophobized AMS as microdispersed filler in asphalt has been tested. Preliminary positive results on reduction of bitumen consumption and improvement of asphalt characteristics in comparison with the use of lime flour, activated mineral powder and cement in its production were obtained.

To exclude the introduction of modifier into the initial bitumen used at asphalt concrete plants (ACP), the technology of its introduction directly into the mixer of the plant was proposed. For this purpose, the resulting modifier is granulated with the addition of AMS obtained from ash dumps of thermal power plants. On the one hand, they reduce the caking of granules, and on the other hand, they are part of the mineral powder introduced into the asphalt-bitumen mixture. The optimum amount of mineral powder necessary to fulfill all the requirements for it has been selected experimentally.

The traceability of the pellets was tested as follows: Tube, with a cross-sectional area of 1.75 cm^2, was fixed in a vertical position on a stand. The modifier was backfilled approximately 3.0 cm in height, a piston with a pad on top was inserted and loaded to a total mass of 0.5 kg. Thus, a pressure of 2.85 m of water column was created. All pellets were incubated for 24 h at 25ºC. When the tested modifier is removed from the tube, the granules may stick together, but they should crumble when lightly pressed. If the granules fall apart individually, the caking is considered to be satisfactory. This test shows that the modifier in big bags can be stored in a 2-tier stack.

Due to obtaining the modifier in granular form there is no need for long storage in hot form, which positively affects the preservation of bitumen properties. Also, in the case of granular material, the geography of use is significantly increased.

In order to exclude inhomogeneities and full disclosure of rubber properties, in contrast to the technology of introduction of fine rubber into bitumen used by NPO "Infotech", we proposed a method of primary heating of rubber crumb in bitumen medium and preliminary devulcanization with further grinding in a disk mill. As a result, the cost of homogenizing rubber in bitumen is significantly reduced, as it is possible to use rubber crumbs with different size ranges. Pre-crushing is necessary to remove the metal cord and prevent it from entering the mill. The maximum size of rubber particles is limited by piping openings and mill clearance. The use of large rubber particles can significantly reduce grinding costs.

The technological methods described above make it possible to achieve a total cost of the modifier lower than the cost of bitumen. And the increase in the performance characteristics of the resulting asphalt concrete allows us to count on increased demand for this modifier. In this case, complete utilization of used tires is carried out (without secondary production of rubber waste, as in the production of sports surfaces), i.e. the environmental problem is solved.

To solve the problem of grinding devulcanized rubber crumb in the medium of hot bitumen, a plant was developed that allows heating the rubber-bitumen mixture to a working temperature of 240ºC and feeding it into the mill for further grinding. The scheme is shown in Fig. 5.

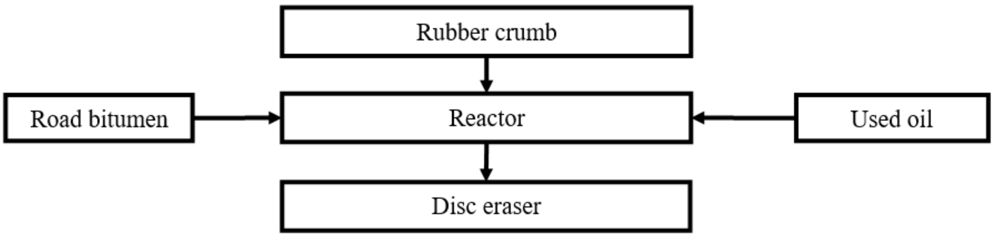

Fig. 5. Scheme of the plant for grinding devulcanized rubber crumb.

Fig. 6 shows a photograph of the manufactured granular bitumen modifier. Such modifier can be stored for up to six months (tested in the laboratory conditions) without loss of technological characteristics, as well as transported over considerable distances and form production stocks for road works.

Fig. 6. Granular bitumen modifier MD-01.

Table 2 shows the results of application of the modifier in comparison with the characteristics of road oil bitumen of BND 90/130 grade (used for regions with cold climate) widespread in the Russian Federation, when it is added to the mixture in the volume of up to 30% instead of an equal part of bitumen.

According to the results presented in Table 3, the following bitumen parameters can be improved (compared to BND 90/130):

- increase in softening temperature by 18 %;
- increase of the brittleness temperature by more than 60 %;
- increase in elasticity of rubber-bitumen binder by an order of magnitude (10 times);
- decrease of the noise level by 50%;
- decrease of rutting formation by more than 80%;
- increase in the strength of the resulting asphalt by 20%.

Inter-repair interval of road pavement increased 2 times. In addition, the cost of binder with modifier MD-01 was reduced by 30-36 % compared to the original bitumen.

Table 3. Cost and physical-mechanical parameters of road oil bitumen modified with MD-01 additive.

Indicators	BND 90/130 (initial)*	0.7 BND 90/130 + 0.3 MD-01**
Cost of binder, USD/kg	0.25	0.16
Softening temperature, °C	45	55
Brittleness temperature, °C	-18	-30
Elasticity, %	0,5	50
Noise level, dB	40	20
Rutting formation, mm	30	5
Asphalt strength at 20°C, MPa	25	30

Note:
* – petroleum road bitumen with penetration from 9 to 13 mm using a special calibrated needle;
** – a mixture of bitumen and modifier in the proportion of 70% to 30%.

The proposed technology allows to create bitumen modifiers for regions with different climates by varying the proportion of modifier and its components in the mixture with bitumen. Increasing the proportion of bitumen allows for a higher softening point, higher strength, and stable roadway performance at high temperatures. Increasing the proportion of rubber increases ductility, adhesion and viscosity, and extends the temperature range. Increasing the proportion of oils lowers viscosity, increases plasticity and provides stable roadway performance at low temperatures.

To test the properties of the modifier in real conditions, a pilot plant for the production of modifier MD-01, with a capacity of 300 kg per day, was manufactured. In 2018, a pilot paving of 300 m^2 of cold asphalt-concrete mix made with the use of modifier was carried out during the repair of driveways along a residential building in Vladivostok. The inspection in January 2023 of the technical condition of the paved asphalt showed that the pavement has no cracks and potholes and is in excellent condition, which testifies to the correctly chosen proportions in the preparation of the binder and to the good performance properties of the pavement.

4. CONCLUSIONS

The technology of obtaining a universal road bitumen modifier, having characteristics comparable to polymer bitumen, applicable to any road bitumen and doubling the service life of road pavement without increasing the cost of asphalt has been developed.

For the first time a combination of three components in the modifier composition was used: coarse-fractional rubber crumb (a priority technical solution of the research group of FEFU), which allows to simplify the technological process of obtaining the modifier and reduce the cost of its production; used machine oil and aluminosilicate microspheres extracted during the processing of ash and slag from coal power engineering.

The obtained modifier was tested in real conditions and confirmed high performance characteristics in the production of asphalt with specified properties for regions with different climates.

All raw components are industrial and household wastes, which allows to simultaneously solve the ecological task of reducing pollution of the environment.

5. ACKNOWLEDGEMENTS

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