Chem. Chem. Technol., 2023, Vol. 17, No. 3, pp. 674–680 Chemical Technology

USE OF RUBBER CRUMB OBTAINED FROM WASTE CAR TIRES FOR THE PRODUCTION OF ROAD BITUMEN AND ROOFING MATERIALS FROM RESIDUES OF UKRAINIAN OIL PROCESSING

Andriy Nagurskyy^{1,⊠}, Oleg Grynyshyn¹, Yuriy Khlibyshyn¹, Bohdan Korchak¹

https://doi.org/10.23939/chcht17.03.674

Abstract. The process of modifying road bitumen obtained from the residues of Ukrainian oils processing with rubber crumb has been studied. The dependence of the softening temperature, ductility, penetration, and elasticity of the three-component bituminous composition "bitumen : linseed oil : rubber crumb" on its formulation has been examined. Based on the mentioned composition a new type of cold-applied bitumen roofing has been developed. A process flow diagram for the production of modified bitumen and special-purpose bituminous composition has been developed.

Keywords: bitumen, bituminous composition, rubberbitumen binder, rubber crumb, linseed oil, bitumen modification.

1. Introduction

Bitumen is one of the oldest and cheapest thermoplastic building materials. It is mostly used in road construction. Modern highway construction sets high demands on the quality of road construction materials, especially on binders, in particular on bitumen for asphalt concrete. The importance of bitumen in the production of road surfaces is paramount. Such coatings provide strength and safety and are 2-3 times cheaper than concrete ones. Most road surfaces consist of mineral fillers and bitumen, the latter of which is used as a strong waterproof binder. However, the problem of road surfaces quality is still relevant in Ukraine.¹⁻³ Typically, bitumen-based asphalt concrete coatings are not able to provide the necessary physical and mechanical properties of coatings and their durability in the conditions of modern tense and intensive traffic. For example, statistical data show that the service life of road surfaces made of bituminousmineral compositions is only 50-70 % of the standard one.

Low elasticity, insufficient crack resistance values, and narrow performance temperature range limit road surfaces use in the summer and winter, especially in areas with frequent changes in air temperature. The above-mentioned shortcomings indicate that bitumen does not meet the necessary quality requirements.⁴⁻⁶

In many countries of the world and in Ukraine, roofing materials based on bitumen are widely used to protect industrial and civil buildings from atmospheric precipitation. Currently, cold-applied roofing and insulating materials are becoming more and more popular. The advantages of these materials are a short period of preparatory work and ease of installation. The disadvantages of cold-applied materials are insufficient adhesion, poor high-temperature and low-temperature properties.

One of the main ways to increase the service life of road surfaces is to modify bitumen used for the production of asphalt concrete. Bitumen, the properties of which are improved by the addition of certain compounds (polymers, rubber crumb, sulfur, adhesive additives, *etc.*) is called a modified bitumen.⁷⁻¹⁴ Bitumen with the addition of polymers is called bitumen-polymer binder (BPB), with the addition of rubber – bitumen-rubber binder (BRB), and with the addition of rubber crumb – rubber-bitumen binders (RBB).¹⁵⁻¹⁸

Bitumen-polymer binder (BPB) differs from bitumen by higher elasticity typical of polymers (elastomers) both at 298 K and 273 K, higher resistance to cracking, and a wider temperature range of performance. In addition, it is characterized by significantly greater durability under repeated dynamic effects in the area of negative temperatures. In this regard, polymer-asphalt concretes are much more durable and resistant to cracks and landslides than asphalt concretes. By varying the content of plasticizer and polymer, it is possible to control and achieve the required quality characteristics of the finished product. Wide implementation of BPB in the practice of industrial road construction is hindered not only by the complexity and high cost of modern plants for the production of bitumen-polymer binders but also by the relatively high cost of bitumen modifiers.^{2,3}

¹ Lviv Polytechnic National University,

^{12,} Bandery St., 79013 Lviv, Ukraine

[™] nagurskiy@ukr.net

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Apart from known bitumen modifiers such as thermoplastics or copolymers of ethylene with vinyl acetate or synthetic rubbers of various types, rubber crumb obtained by crushing used car tires (UCT), can be used as well. Every year the UCT amount increases and their reuse allows not only to solve the urgent environmental problem of their accumulation but also to save money. Currently, rubber crumb is one of the most widespread production wastes, which is used to modify bitumen.^{1,7,8,19} Nowadays, UCT are used as raw materials for the lowtemperature pyrolysis process or burned.¹⁴ However, their grinding and use in bitumen production is, in our opinion, much more profitable.

This research aims to study the basic regularities of the bitumen modification process obtained by processing residues of Ukrainian oils with rubber crumb in order to obtain high-quality road bitumen and bituminous roofing materials, as well as to develop a process flow diagram of this process.

2. Experimental

Research on the process of obtaining bituminous materials using rubber crumb was carried out in two directions. We studied the main regularities of the road bitumen modification with rubber crumb and determined the optimal formulation of the three-component composition "bitumen-linseed oil-rubber crumb", which will be used for gluing bituminous roofing materials.

Road petroleum bitumen of the BND 90/130 grade with the following characteristics was chosen as the starting material to study the modification process:

- softening temperature 322 K;
- ductility at 298 K 38 cm;
- penetration at 298 K 110 x 0.1 mm;
- elasticity 59 %.

Bitumen under study is produced from residues of the processing of a mixture of paraffinic and highly paraffinic Ukrainian oils. As a modifier, we used rubber crumb obtained by grinding used car tires (fractions of 0.6-0.8 mm and 0.8-1 mm).

The modification process was studied at a laboratory mixing setup. To prepare modified bitumen, preweighed samples of the initial components were loaded into a metal container and heated on an electric stove to a temperature of 433 K, under constant stirring. The mixing process was carried out for 4–6 h. The effect of the granulometric composition of rubber crumb, its percentage content in bitumen, temperature regimes, and mixing time on the quality of the resulting modified bitumen was determined.

Construction petroleum bitumen of the BN 70/30 grade with the following characteristics was chosen as the

starting material to study the process of obtaining coldapplied bituminous roofing material:

- softening temperature 355 K;
- ductility at 298 K 2 cm;
- penetration at 298 K 17 x 0.1 mm;
- elasticity 50 %.

Linseed oil (LO) and rubber crumb (fraction of 0.8-1.0 mm) were also used to obtain the bituminous composition.

The process of obtaining a bituminous composition for gluing cold-applied roofing material was carried out at a laboratory setup at a temperature of 473–493 K. The effect of three-component composition formulation on operational properties was studied. For the resulting compositions, the softening temperature ("ring and ball" method), ductility, penetration, elasticity, heat resistance, water absorption, and flexibility on the bar at 248 K were determined according to standardized methods.^{20,21} Adhesion was determined by measuring the mechanical force during the separation of the metal seal from the surface covered with bituminous material.

3. Results and Discussion

The experimental results of road bitumen modification with rubber crumb are represented in Table 1. An increase in the rubber crumb content increases the modified bitumen viscosity. For example, when modifying BND 90/130 bitumen with 15 wt. % of rubber crumb, a modified bitumen of BMPA 40/60-57 grade is formed, which has boundary values of viscosity. At the same time, the ductility is slightly lower than the requirements of DSTU B B.2.7-135:2014.²² On the other hand, too high content of rubber crumb in bitumen can cause technological difficulties during its modification and create problems in obtaining the asphalt coating itself and its high-quality laying.

The penetration value was found to be decreased with an increase in the rubber crumb amount in modified bitumen. Moreover, a more significant reduction is observed when using a larger fraction of rubber crumb. The ductility of bitumen also decreases due to the increase in the rubber crumb amount but the softening temperature increases. The granulometric composition of rubber crumb is also important for the bitumen modification process. As a result of studying the main regularities of the process, it was found that when a larger fraction of rubber crumb is used to modify bitumen, a binder with lower penetration and softening temperature is obtained. Thus, a binder is formed with higher hardness and infusibility as compared to the original bitumen.

IndKex	Rubber crumb amount, wt. %			
	0	5	10	15
	Mixing time	4 h	•	
	Rubber crumb fraction	n 0.6-0.8 mm		
Penetration at 298 K, 0.K1 mm	110	100	80	60
Softening temperature, K	322	331	336	338
Ductility at 298 K, cm	38	34	27	18
Elasticity, %	59	80	82	86
	Rubber crumb fractio	n 0.8-1.0 mm		
Penetration at 298 K, 0.1 mm	110	94	75	49
Softening temperature, K	322	328	333	336
Ductility at 298 K, cm	38	33	26	17
Elasticity, %	59	79	81	85
	Mixing time			
	Rubber crumb fractio	n 0.6-0.8 mm		
Penetration at 298 K, 0.1 mm	110	95	76	52
Softening temperature, K	322	332	336	339
Ductility at 298 K, cm	38	32	27	17
Elasticity, %	59	81	83	87
	Rubber crumb fractio	n 0.8-1.0 mm		
Penetration at 298 K, 0.1 mm	110	92	73	47
Softening temperature, K	322	330	335	337
Ductility at 298 K, cm	38	31	26	16
Elasticity, %	59	80	81	86

Table 1. Characteristics of BND 90/130 road bitumen modified with rubber crumb

An important aspect of the modification is that, despite the decrease in ductility of the binder obtained with the participation of rubber crumb, its elasticity is significantly increased, which in the long run will positively affect the quality and durability of the asphalt concrete coating based on it.

It has been established that there is no necessity to dissolve rubber crumb in bitumen completely. To ensure appropriate physical and mechanical indices of the binder, it is enough to carry out only surface devulcanization of the rubber crumb. Moreover, heating to high temperatures is associated with additional energy costs.

The minimum mixing time (modification time) was found to be 4 h. With a shorter mixing time, partial layering of the bitumen : rubber crumb system is observed, which is unacceptable. When the mixing time increases, the quality characteristics of the obtained bitumen do not change significantly (Figs. 2-5). Moreover, it is economically feasible to increase the mixing time.

Thus, the principle possibility of using rubber crumb to modify bitumen obtained from the residues of paraffinic oils processing was established. It is shown that the addition of crumb rubber affects the main properties of bitumen: penetration, softening temperature, and elasticity, and makes it possible to replace expensive industrial elastomers in the production of modified bitumen. Polymer-modified bitumen based on oxidized petroleum bitumen was obtained by adding 5-12 wt. % of rubber crumb. The obtained bitumen meets the requirements according to DSTU B V.2.7-135:2014.²²

The modification of BND 90/130 road bitumen with rubber crumb in the amount of 5 wt. % at a temperature of 433 K for 4 h allows to obtain modified bitumen of the BMPA-90/130-50 grade, and modified bitumen BMPA-60/90-53 is obtained under the same conditions when adding 10 wt. % of rubber crumb.

Another option for using crumb rubber is the production of cold-applied bituminous roofing materials. Roofing with such materials does not require gluing or heating, and the material itself is easily applied to the surface and firmly held on it.

To ensure the set of properties required for coldapplied bituminous materials, in particular the combination of high adhesive characteristics (stickiness) with high plasticity at low temperatures, heat resistance, and sufficient hardness, we studied the properties of binary mixtures obtained by mixing BN 70/30 building bitumen and linseed oil (LO), which belongs to the group of "drying" oils and shows the high efficiency compared to other vegetable oils (sunflower, rapeseed, *etc.*).¹⁹ Nevertheless, a bituminous material based on only mentioned two components does not meet all the necessary requirements. It is necessary to add a third component, which would increase the hardness, softening temperature, elasticity, and frost resistance of the bituminous material without deteriorating other indices. This is the effect that is achieved by introducing rubbers into bitumen, in particular, SBS^{18,19} but this product is quite expensive. Instead of SBS, we propose to use rubber crumb, the cost of which is much lower. Therefore, we studied the properties of a threecomponent mixture containing BN 70/30 bitumen, LO and rubber crumb in various ratios. The experimental results are shown in Tables 2-4.

Also, after analyzing previous studies, it can be assumed that the modification of road and construction bitumen with rubber crumb in two cases has a physical nature, since the process is carried out at insufficiently high temperatures for chemical interaction. In our case, the modification occurs as a result of the swelling of the rubber crumb due to the absorption of part of the oil components.²³ It is obvious that with an increase in the rubber crumb amount and a decrease in the LO amount, the softening temperature of the mixture increases but its penetration decreases. The ductility of the three-component mixture increases with an increase in the LO amount. The maximum elasticity is achieved with the maximum content of rubber crumb and the minimum amount of LO.

The investigated mixture stands the flexibility test when LO amount is above 15 wt. % and the amount of rubber crumb is below 10 wt. %. Otherwise, the bituminous material becomes brittle, which does not meet the requirements. The value of adhesion passes through the maximum when the LO amount is 15 wt. % and the addition of rubber crumb decreases adhesion.

Table 2. Characteristics of the three-component mixture "bitumen : LO : rubber crumb" (LO amount is 10 wt. %)

Index	BN 70/30 + 10 wt. % of LO	Rubb	Requirements for		
		5	10	15	bituminous composition
Softening temperature, K	338	345	356	3630	> 343
Ductility at 298 K, cm	4.5	3.5	3.5	3.0	-
Penetration at 298 K, 0.1 mm	48	29.0	25.0	22.0	-
Elasticity, %	23	57.0	71.0	81.0	> 20
Flexibility on the bar at 248 K	fail the test	fail the test	fail the test	fail the test	stand the test
Heat resistance at 333 K	fail the test	stand the test	stand the test	stand the test	stand the test
Adhesion, N/cm ²	5.0	4.0	2.2	1.5	> 5.0
Water absorption, %	0.08	0.11	0.12	0.12	< 1.0

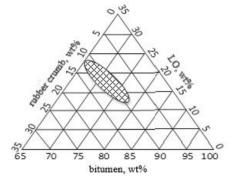
Table 3. Characteristics of the three-component mixture "bitumen : LO : rubber crumb" (LO amount is 15 wt. %)

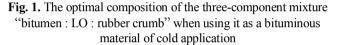
	BN 70/30+	Rubbe	Requirements for		
Index	15 wt. % of LO	5	10	15	bituminous composi-
	10 Wt. 70 01 20	5	10	15	tion
Softening temperature, K	330	339	353	360	>343
Ductility at 298 K, cm	7.0	3.0	3.0	3.5	-
Penetration at 298 K, 0.1 mm	93	33.0	31.0	26.0	—
Elasticity, %	21	50.0	59.0	70.0	> 20
Flexibility on the bar at 248 K	stand the test	stand the test	stand the test	fail the test	stand the test
Heat resistance at 333 K	fail the test	fail the test	stand the test	stand the test	stand the test
Adhesion, N/cm ²	12.0	7.4	6.9	4.5	> 5.0
Water absorption, %	0.27	0.15	0.12	0.12	< 1.0

Table 4. Characteristics of the three-component mixture "bitumen : LO : rubber crumb" (LO amount is 20 wt. %)

	BN 70/30+	Rubber crumb amount, wt. %			Requirements for
Index	20 wt. % of LO	5	10	15	bituminous compo- sition
Softening temperature, K	325	335	347	356	> 343
Ductility at 298 K, cm	7.5	2.5	3.0	3.5	-
Penetration at 298 K, 0.1 mm	131	38.0	35.0	31.0	-
Elasticity, %	20	40.0	48.0	57.0	> 20
Flexibility on the bar at 248 K	stand the test	stand the test	stand the test	fail the test	stand the test
Heat resistance at 333 K	fail the test	fail the test	stand the test	stand the test	stand the test
Adhesion, N/cm ²	9.5	6.6	5.3	4.4	> 5.0
Water absorption, %	0.12	0.12	0.13	0.15	< 1.0

Analyzing the obtained results with the help of a triangular diagram, the region of the mixture optimal composition was established, which ensures the achievement of the required quality characteristics of the cold-applied bituminous material (Fig. 1).





So, the optimal "bitumen : LO : rubber crumb" composition (wt. %) is the following: (65.0-78.5) : (12.5-22.5) : (9.0-12.5). Based on the obtained results, a bituminous composition for cold-applied materials was developed, consisting of 72.5 wt. % of BN 70/30 bitumen, 17.5 wt. % of LO, and 10.0 wt. % of rubber crumb, the characteristics of which are given in Table 5.

The developed bituminous composition for coldapplied roofing material meets the requirements, and due to the increased softening temperature, the developed roofing materials have increased stability in hot climates.

A process flow diagram for the production of bituminous materials modified with rubber crumb is shown in Fig. 2. The initial bitumen from the E-1 tank is fed into the P-1 reactor by the H-1 pump. The amount of bitumen is controlled using a VM-1 flow meter. In the P-1 reactor, the bitumen is heated to the process operating temperature due to the supply of the heat carrier into the reactor's heating jacket and mechanical mixing. After reaching the required temperature, modifiers (or components of the bitumen composition) are fed into the P-1 reactor.

Table 5. Characteristics of the bitumen composition for cold-applied roofing material

Index	Value	Requirements for bituminous com- position
Softening temperature, K	349	> 343
Ductility at 298 K, cm	3.2	_
Penetration at 298 K, 0.1 mm	33	-
Elasticity, %	53	> 20
Flexibility on the bar at 248 K	stand the test	stand the test
Heat resistance at 333 K	stand the test	stand the test
Adhesion, N/cm ²	5.6	> 5.0
Water absorption, %	0.15	< 1.0

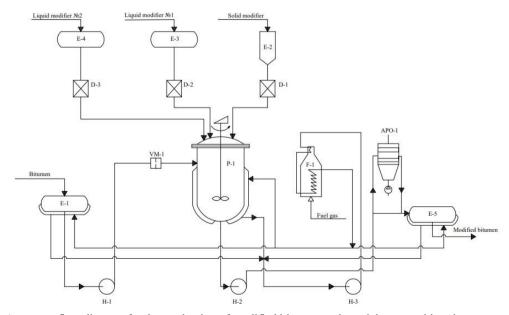


Fig. 2. A process flow diagram for the production of modified bitumen and special purpose bituminous composition

The solid bulk modifier (rubber crumb) is fed into the P-1 reactor from the E-2 tank by self-flow (due to the difference in levels). The amount of the modifier is controlled using the D-1 metering device.

Liquid modifiers are supplied to the P-1 reactor from the E-3 and E-4 tanks by self-flow (due to the difference in levels). The amount of the modifier is regulated using metering devices D-2 and D-3.

Components are mixed in the P-1 reactor at a given temperature for a definite time. After the end of the mixing process, as a result of which the bitumen is modified or the bituminous composition is obtained, the resulting product, which is previously cooled in the APO-1 air cooling refrigerator, is pumped from the P-1 reactor into the E-5 tank by the H-2 pump. If necessary, the resulting product can be pumped to E-5 without cooling. An additional bypass line is provided for this.

The liquid heat carrier used for heating the P-1 reactor, E-1 and E-5 tanks, is fed from the heating jackets of these devices by the H-3 pump to the tube furnace F-1, heated to the operating temperature, and sent back to the corresponding heating jackets.

4. Conclusions

The fundamental possibility of using rubber crumb in bitumen production has been established. It has been proven that the addition of rubber crumb affects the main properties of bitumen: ductility, penetration, softening temperature, and elasticity, and allows the replacement of expensive industrial elastomers necessary for the production of polymer-asphalt concrete.

The modification of BND 90/130 road bitumen with rubber crumb in the amount of 5 wt. % at a temperature of 433 K for 4 h allows to obtain modified bitumen of the BMPA-90/130-50 grade, and modified bitumen BMPA-60/90-53 is obtained under the same conditions when adding 10 wt. % of rubber crumb.

Based on the results of studying the dependence of the three-component system properties on its formulation, a new type of bituminous composition for cold-applied roofing materials was obtained. The optimal formulation of the bituminous composition was established: 65.0-78.5 wt. % of BN 70/30 bitumen, 12.5-22.5 wt. % of linseed oil and 9.0-12.5 wt. % of rubber crumb.

The flow diagram capable of working in different modes was developed for the modification of bitumen with rubber crumb with the possibility of simultaneous addition of liquid modifiers.

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> Received: February 27, 2023 / Revised: April 27, 2023 / Accepted: May 15, 2023

ВИКОРИСТАННЯ ГУМОВОЇ КРИХТИ, ОДЕРЖАНОЇ З ВІДПРАЦЬОВАНИХ АВТОМОБІЛЬНИХ ШИН, ДЛЯ ВИРОБНИЦТВА ДОРОЖНІХ БІТУМІВ ТА ПОКРІВЕЛЬНИХ МАТЕРІАЛІВ З ЗАЛИШКІВ ПЕРЕРОБКИ УКРАЇНСЬКИХ НАФТ

Анотація. Досліджено процес модифікування дорожнього бітуму, одержаного з залишків переробки українських нафт гумовою крихтою. Вивчено залежність температури розм'якшення, дуктильності, пенетрації та еластичності трикомпонентних бітумних композицій "бітум : лляна олія : гумова крихта" від їхнього складу. Розроблено новий тип бітумного покрівельного матеріалу холодного нанесення на основі трикомпонентної бітумної композиції "бітум : лляна олія : гумова крихта". Розроблено принципову технологічну схему установки для одержання модифікованих бітумів та бітумних композицій спеціального призначення.

Ключові слова: бітум, бітумна композиція, гумовобітумне в'яжуче, гумова крихта, лляна олія, модифікування бітуму.