UDC 691.162

S. N. Shabaev[1]

FACTORS INFLUENCING THE EFFICIENCY OF THE PROCESS OF MODIFICATION OF OIL BITUMENS WITH RUBBER CRUMB

Kuzbass State Technical University Named after T. F. Gorbachev
Russia, Kemerovo, tel.: (3842) 39-63-22, e-mail: isa.ad@kuzstu.ru
[1]PhD in Engineering, Assoc. Prof., Head of the Dept. of Highways and Urban Cadastre

Statement of the problem. In global practice the greatest distribution was gained by technologies of modification of Asphalt Rubber and Terminal Blended bitumens, each of which possesses the shortcomings. Therefore it is necessary to combine them on the basis of the synergy effect, and for this purpose it is necessary to know what factors influence the efficiency of modification.
Results. The factors (the size, a specific surface and the chemical compositions of a rubber crumb, speed of rotation of the mixer, structure of the dispersive environment, temperature and duration of process) determining the efficiency of process of modification of bitumens by a rubber crumb with their systematization and breakdown on a version (basic active and passive, integrated) as well as justification of the recommended range of definition are defined.
Conclusions. As a result of the executed theoretical research, the use of bitumen of a rubber crumb for modification with the developed specific surface and big scope of the sizes of particles of the average value, rather small on value, is justified and the rational interval of a variation of temperature of process is established.

Keywords: rubber crumb, bitumen, modification, factors, process.

Introduction. The technology of the modification of rubber crumb bitumen has long been developed and there are studies going on to improve it. However, there is no comprehensive theoretical model to account for the influence of various factors on the efficiency of the process. This can be explained by inconsistency of the results of studies and different suggested technological parameters of combining rubber crumb with oil bitumen, which puts constraints on the use of similar technology in actual economics sector.

In world practice two major technological modifications of rubber crumb bitumen are considered: the technology with a relatively low (160…200 ºC) temperature of the thermal mechanical process with no special requirements for mixing equipment [23, 24, 28—34], i.e. the *Asphalt Rubber* method, and the technology with a high (200…260 ºC) temperature of the thermal mechanical process using special mixers, i.e. the *Terminal Blended* method [26, 27, 36].

Combined methods are also suggested when the temperature of the process corresponds with a range typical of the *Terminal Blended* method when no high-speed mixers are supposed to be employed or the temperature of the process is accepted to range from 160…200 ºC with simultaneous use of high-speed mixers [1, 7, 16].

Studies dedicated to the combined method are caused by the fact that each of the basic methods has certain disadvantages. For the *Asphalt Rubber* methods they are:

— there is constant mixing of a binder in order to avoid sedimentation of rubber particles that can be stored for no more than 8 hours;

— a binder is a heterogeneous product that hinders pumping and use.

The disadvantages of the *Terminal Blended* method are the following:

— in order to be obtained complex costly equipment with the rate of rolling of a mixer of around 8000 rotations a minute is necessary;

— a binder has a low high-temperature stability.

Therefore the most urgent issue is not to improve these methods but to combine them based on the synergy effect in order to eliminate the above disadvantages while keeping advantages. However, in order to address the task, it is necessary to know what factors and in what way impact the efficiency of combining rubber crumb abd oil bitumen.

Generalizing the data of the previous studies and our experience, we assume that during combining rubber crumb and bitumen the former swells in low-molecular fractions of bitumen and (or) plastifier (a combining agent) forming a disperse medium followed by its partial or complete solidification with some of linear chains of rubber macromolecules breaking away when they are relatively evenly distributed in a liquid phase. However, the rate of swelling and solidification (these might not take place) of rubber crumbs will be largely dependent on the factors (parameters) of a technological process.

All the technological factors can be classified in two groups:

— the factors that cannot be changed or only following changes in a technological process of obtaining rubber crumb or designing of industrial setups for obtaining rubber bitumen binders, i.e. passive basic factors (size, specific surface of particles and chemical composition of rubber crumbs, designing and rotation rate of a mixer);

— the factors that can be changed when a technological process of combining rubber crumb and bitumen is controlled, i.e. active basic factors (composition of a disperse medium, temperature and time of the thermal mechanical process of combination).

1. Influence of the size of particles of rubber crumb on the efficiency of modification of bitumens. The size of particles is considerable in swelling and solidification of rubber crumb.

Let change in the size of rubber crumb (swelling or solidification) take place within the time Δt by $\Delta D_ч$, then assuming that rubber crumb has the shape of a ball, change in the volume of this particle is (Fig. 1):

$$\Delta V_ч = \frac{\pi}{6}\cdot\left[D_ч^3 - \left(D_ч - \Delta D_ч\right)^3\right] = \frac{\pi}{6}\cdot\left(3\cdot D_ч^2\cdot\Delta D_ч - 3\cdot D_ч\cdot\Delta D_ч^2 + \Delta D_ч^3\right), \quad (1)$$

where $\Delta D_ч$ is an average weighted diameter of rubber particles.

At $\Delta t \to 0$ $\Delta D_ч \to 0$, thus the second and third summands in the formula (1) for the ultimate accuracy can be neglected. In the simplified form it will look as follows respectively (1):

$$\Delta V_ч = 0.5\pi\cdot D_ч^2\cdot\Delta D_ч. \quad (2)$$

Let the total volume of rubber particles that are swollen or solidified be Q, then the amount of the particles will be given by the equality:

$$N_ч = \frac{Q}{V_ч} = \frac{6\cdot Q}{\pi D_ч^3}. \quad (3)$$

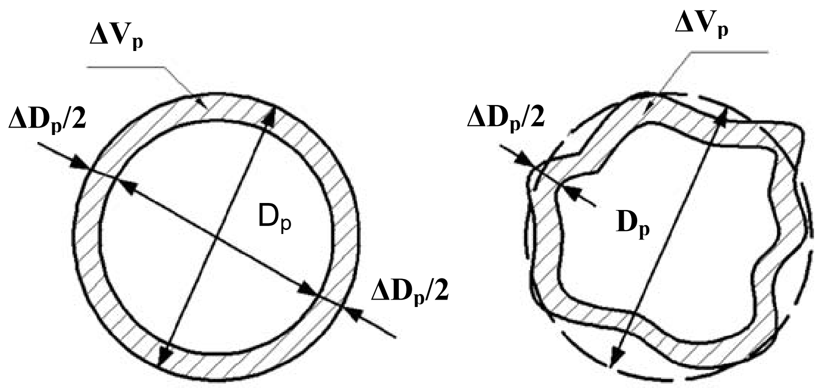

Fig. 1

Change in the volume of all the rubber particles that are swollen and solified over the time Δt is

$$\Delta V_{общ} = \Delta V_ч\cdot N_ч = \frac{3\cdot Q\cdot\Delta D_ч}{D_ч}. \quad (4)$$

The analysis of the dependence (4) shows that for the same volume of rubber particles (assuming that $\Delta D_ч \approx$ const) a decrease in their size causes an increase in their volume during swelling and solidification over the same time Δt. Thus the smaller particles of rubber crumb are, the higher the rate of swelling and solidification is (for the same volume of rubber particles).

Also note that during combination of rubber crumb with bitumen there is generally no complete solidification, i.e. a rubber bitumen binder is a heterogeneous system containing some rubber crumb. The smaller rubber particles are in the same content in the overall volume, the more evenly they are distributed in a system, i.e. as the size of rubber particles decreases, so does heterogeneity of a rubber bitumen binder causing a binder to be more stable. Besides, based on the Stokes equation, the rate of sedimentation of a disperse phase (rubber particles) in a disperse medium (a bitumen rubber binder along with a combining agent or without) depends on the size of a disperse phase and has an increasing parabolic dependence, i.e. as the size of rubber particles decreases, the rate of their sedimentation will drop considerably. Given that there are association forces between rubber particles and a disperse medium during thermal and mechanical combination, for some small particles of rubber crumb the association force is higher than the gravitational force and thus there is no sedimentation. These are supported by studies by I. A. Dibrova [4].

An average weighted size of rubber particles gives an overall idea of the significance and efficiency of using such rubber crumb. However, more objective data can be obtained if extra analysis is carried out of a range in the size of all the particles in relation to an average weighted value. A range in the size of the particles of rubber crumb in relation to an average weighted value can be described by a scattering coefficient given by the dependence:

$$K_{dis} = 1 - \sum_{i=1}^{n} \left(ЧO_i \right)^2 , \quad (5)$$

where n is a number of sieves including a tray where particles of rubber crumb are retained; PO_i is a residual in the i-th sieve or tray, units.

The dependence (4) allows the evaluation of total changes in the volume of rubber particles of the same size per a unit of time Δt. However, in fact particles of rubber crumb have the same size, i.e. it can be stated that rubber crumb is mostly a mix of different fractions.

Then the equation (4) is rather written as:

$$\Delta V_{общ} = 3 \cdot \Delta D_ч \cdot \sum_{i=1}^{n} \frac{q_i}{\left(D_ч \right)_i} , \quad (6)$$

where q_i is the volume of particles of the i-th fraction in rubber crumb; $(D_ч)_i$ is an average size of the particles of the i-th fraction determined by a half-sum of the size of the cells of this and previous sieves.

It is known that a particular residual in the i-th sieve is determined by a ratio of the mass in this sieve to the total mass of the sample for analysis. Given that an average density of all the rubber particles is almost identical, the particular residual can be given by the dependence:

$$ЧO_i = \frac{q_i}{Q}, \text{ thus, } q_i = Q \cdot ЧO_i .$$

Then the formula (6) can be written as follows:

$$\Delta V_{общ} = 3 \cdot Q \cdot \Delta D_ч \cdot \sum_{i=1}^{n} \frac{ЧO_i}{\left(D_ч\right)_i}. \quad (7)$$

If the total volume of rubber crumb used to modify bitumen is assumed to be constant, $3 \cdot Q \cdot \Delta D_ч$ in the formula (7) is constant, i.e. change in the volume of all the rubber particules over the time Δt can be accepted to be expressed by means of a total relative change in their volume:

$$\Delta V_{общ}^{отн} = \sum_{i=1}^{n} \frac{ЧO_i}{\left(D_ч\right)_i}. \quad (8)$$

Therefore the total change in the volume of rubber particles over the time Δt is determined exclusively by their granular metric composition and thus knowing the granular metric composition of rubber crumb, the efficiency of its combination with oil bitumen can be predicted.

Let there be nine different samples of rubber crumb. The granular metric compositions of these samples as well as the results of calculating an averaged weighted size of the particles, the scattering coefficient as well as the total relative change in the volume of rubber particles over the time Δt are presented in Table 1. Graphical presentation of the dependencies of the total relative changes in the volume of rubber particles over the time Δt on the average weighted size of the particles and scattering coefficient is given in Fig. 2.

Table 1

Characteristics of different samples of rubber crumb

Original data									
Size of the cells of the sieves, mm	Particular residual in the sieve, %. for the sample №								
	1	2	3	4	5	6	7	8	9
5	—	—	4	—	—	—	—	—	—
2.5	6	9	9	—	—	4	—	—	—
1.25	42	38	21	6	9	9	—	—	4
0.63	52	44	36	42	38	21	6	9	9
0.315	—	9	25	52	44	36	42	38	21
0.16	—	—	5	—	9	25	52	44	36
0.05	—	—	—	—	—	5	—	9	25
Less than	—	—	—	—	—	—	—	—	5
Total	100	100	100	100	100	100	100	100	100

End of Table 1

Calculation data									
Calculation index	Calculation index for the sample №								
	1	2	3	4	5	6	7	8	9
Dcp, mm	1.50	1.51	1.50	0.75	0.75	0.75	0.38	0.38	0.37
Kdis	0.55	0.65	0.75	0.55	0.65	0.75	0.55	0.65	0.75
$\Delta V_{общ}^{отн}$, mm^{-1}	0.793	0.886	1.265	1.585	1.763	2.566	3.124	3.595	6.445

The analysis of the above data suggests that for a small scattering of the size of rubber particles of a relative average weighted value (*Kdis* < 0.65) a decrease in the average weighted size of particles of rubber crumb causes a corresponding increase in the total relative change in the volume of rubber particles over the time Δt. For a larger scattering of the size of particles of a relatively average weighted value, a decrease in the latter causes a more significant growth of the total relative change in the volume of rubber particles over the time Δt.
Scattering of the size of rubber crumb of a relatively averaged weighted value has a considerable effect on the total relative change in the volume of rubber particles over the time Δt and if for a relatively large rubber crumb change can be about 50 %, i.e. for smaller rubber crumb this might be 100 % and beyond.

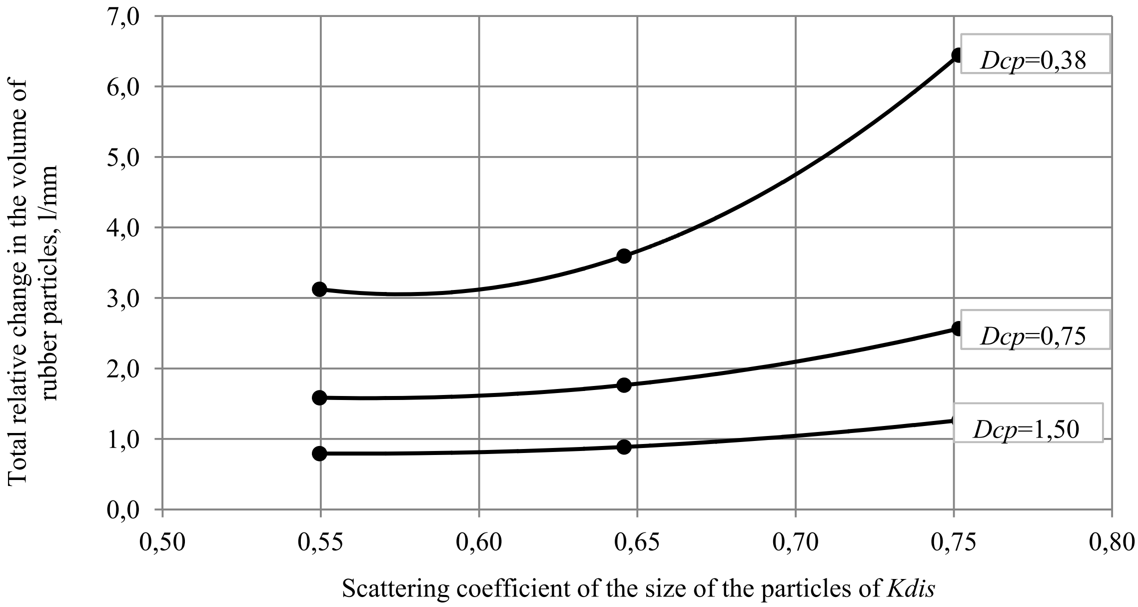

Fig. 2. Dependencies of the total relative change in the volume of rubber particles overt the time Δt on an average weighted size of particles and scattering coefficient

2. Influence of a specific surface of particles of rubber crumb on the effectiveness of the modification of bitumens. Besides the size of rubber particles, their specific surface is important as well. For a larger area of the surface of a rubber particle with the same average size change in the volume of this particle as a result of thermal and mechanical impact over the same time Δt is more considerable (Fig. 1) and thus the rate of swelling and solidification is higher. This is proved by the data by V. G. Nikolskiy, V. S. Prokopets, A. A. Khristoforova [6, 13, 19]. In addition, according to the physical and chemical concepts set forth by P. A. Redinberg, in order to improve the strength of materials, their temperature stability and elasticity, an extremely large surface of their impact and tight contact of the phases taking part in the reaction [14, 15]. A high specific surface of rubber particles can be reached by means of high-temperature shift grinding of larger rubber particles or mechanic activation of rubber crumb.

3. Influence of the chemical composition of rubber crumb on the efficiency of modification of bitumens. The chemical composition of rubber has an unquestionable effect on the rate of swelling and solidification of rubber crumb as the temperature of failure of different rubber varies. This factor is not only passive but also heterogeneous. In order to make it more homogeneous, it is advisable that rubber crumb is obtained using wornout car tyres of the same brand and production. This can only be achieved if tyres are processed in the same large enterprise (e.g., an angle cut).

One of the most preferable rubber components of rubber crumb employed to modify bitumens is natural rubber with a higher adhesive capacity, elasticity and strength compared to synthetic rubber. However, natural rubber has a severe disadvantage, i.e. a low resistance to atmosphere (oxidation). Therefore tyre rubber crumb obtained only based on natural rubber is not acceptable as a bitumen modifier. This is why restrictions in the amount of natural rubber in rubber crumb used to modify organic binders in the USA [7]. In fact manufacturers of tyres that are designed to operate under excessive loads (aviation tyres, mining truck tyres) generally employ a mix of natural rubber with synthetical one, which allows rubber bitumen binders to be obtained based on them with al arge elasticity range, high adhesive capacity and elasticity on the one hand and a long life cycle on the other. Therefore it is necessary to use rubber crumb from wornout aviation and heavy-duty tyres to modify bitumens.

4. Influence of the rate of rotation of a mixer on the efficiency of modification of bitumens using rubber crumb. The structure and rotation speed of a mixer are the factors that impact the intensity of solidification of rubber particles. When a solid body (a rubber particle) is in motion in a liquid (a disperse medium), it is affected by the resistance force of the medium that apart from its viscocity, depends on the velocity and has a non-linear increasing nature, almost parabolic one. The higher the velocity is (the number of rotations of a mixer) and thus the resistance force, the more likely the bond *–S–S–*, *–S–C–* и *–C–C–* is to break away per a unit of time Δt thus causing rubber crumb to solidify at a faster rate. This is the effect that is utilized in the *Terminal Blended* method.

5. Influence of the composition of a disperse medium on the efficiency of modification of bitumens using rubber crumb. A disperse medium is oil bitumen, plastifier or a mix of oil bitumen with a plastifier (a combining agent). As the latter different authors use polyhydron [7], gas tar and its particular fraction [9, 18, 20, 25, 36], rubber [9, 35], divinylsterol thermoelastolayer [16], hydrocarbon oil [2, 3, 5], etc. The use of combining agents is first of all due to the fact that most pyrobitumens in bitumens tend to absorb aromatic hydrocarbons [3]. Therefore a disperse medium of bitumens is a system exhausted by free aromatic hydrocarbons that are the major components influencing the intensity of swelling and solidification of rubber crumb. Despite enhancing swelling and solidification of rubber crumb, oils that bitumens contain are less active solidifiers. Hence it is understandable why different authors recommend that plastifiers should be introduced for catalyzing swelling, solidification as a result of combining rubber crumb and (or) solidification byproducts (rubber macromolecules) with bitumen components.

The combining agents that are employed should not:

— dissociate under the effect of planned temperature impacts during combination of rubber crumb with oil bitumen;

— be classified as more hazardous than oil bitumen;

— cause an increase in the viscocity of the final product as the introductuion of rubber crumb increases the viscocity of rubber bitumen binders;

— be costly.

6. Influence of the temperature on the efficiency of modification of bitumens using rubber crumb. Temperature has a large impact on swelling and solidification of rubber crumb in a disperse medium and the higher it is (within an acceptable range), the faster these are. E.g., in [2] it is proved that swelling of rubber particles in bitumen at the temperature 160 °C does not depend on how long a process is does not exceed 50 % and solidification 25 %. At the same time at a higher temperature rates of swelling and solidification can be higher. However, at a high temperature oxidation of a binder is considerably more intense, which causes degradation of its properties and intensive aging. Therefore at the temperature of thermal and mechanical combination of rubber crumb of over 160 ºC it is crucial that there is as little oil bitumen as possible so that the entire rubber crumb is processed in a plastifier solution (e.g., mineral oil) with bitumen or without [12], or for a small amount of a combining agent by introducing some of rubber crumb. The first method is technically less challenging and time of thermal and mechanical impact on the entire volume of rubber crumb is constant. However, it generally has 1.25 times as much plastifier along with bitumen or without as the mass of rubber crumb as otherwise rubber crumb will not absorb humidity and become sticky.

The temperature of a thermal and mechanical process should have an upper acceptable range that is determined by the intensity of failure of rubber that rubber crumb contains in the bonds –*C*–*C*– resulting in low-molecular hydrogens including volatile ones. It is known that natural rubber starts decomposing at the temperatures of over 200 ºC and at the temperatures of over 250 ºC radical depolymerization is increasing and almost uncontrollable. Experimental data suggests [21] that at the temperature of 260 ºC solidification of rubber crumb in a disperse medium occurs with an intense emission of volatile hydrocarbons and there is hardly any modification of a binder taking place even if the process takes as long as a few minutes. Given that in wornout car tyres particularly aviation and heavy-duty ones mostly used for rubber crumb, natural rubber is a major component, the upper acceptable temperature range of a thermal and mechanical process of combining rubber crumb with bitumen should not be over 250 ºC.

Given that the maximum rate of swelling and solidification of polymers is reached at the temperatures close to those of failure, while combining rubber crumb with oil bitumen there are two contradictory conditions: the temperature of combining should not be over 160 ºC so that the properties of an original binder are retained as much as possible; the temperature of combining should be close to that of failure of rubber and be no less than 190…200 ºC (for natural rubber). Therefore in order to achieve maximum combination of rubber crumb with bitumen binders, it is necessary that one of the factors (e.g., solidification of rubber crumb in a palstifier followed by combining the resulting solution with bitumen) is excluded or external physical fields (e.g., ultrasound waves, excessive pressure, etc.) are employed to activate combination of rubber crumb with bitumen at low temperatures.

7. Influence of time on the efficiency of modification of bitumens using rubber crumb. Swelling and solidification of rubber crumb in a disperse medium has an influence on the time of a thermal and mechanical process. Theoretical studies [22] suggest that for each set of basic factors there is its own time of obtaining rubber bitumen binders where the amount of resulting rubber structuring an original binder is maximum and the final product has the best physical and chemical properties. Therefore it is more viable to use not the time factor but that of optimal time of a thermal and mechanical process. This allows this factor to be regarded separately from the other basic factors, which would also enable the number of experiments involved in modeling a process to drop.

Conclusions. As a result of the study to identify the factors that influence the efficiency of combining rubber crumb with bitumen, for the first time:

— the results of previous studies have been summarized as well as those obtained with the author's participations, which allowed all the factors influencing the efficiency of the process to be considered as well as recommended according to their application range;

— the use of rubber crumb which is as fine as possible with the most developed specific surface to modify bitumens has been theoretically justified;

— it has been found that the more scattered the size of particles is in relation an average weighted size (the scattering coefficient is over 0.65), the larger a modification effect that can be achieved is, i.e. a modified bitumen will have improved properties;

— it has been established that a rational range of the temperature should be from 190 to 250 °C.

All the factors as well as a recommended application range are presented in Table 2.

Table 2

Factors of modification of bitumens using rubber crumb and their recommended application range

№	Factor	Recommended application range (based on a literature review and results of our own studies)
		Basic passive factors
1	Total relative change in the volume of rubber particles over the time Δt, mm^{-1}	Up to 1.5 — acceptable, 1.5…3.0 — good, over 3.0 — excellent
2	Specific surface of particles of rubber crumb, m^2/g	0.01…0.1 — acceptable, 0.1…0.5 — good, over 0.5 — excellent
3	Chemical composition of rubber crumb	Total volume of rubber compounds is 40…65 %; content of natural rubber is 20…40 %
4	Rate of rotation of a mixer, rotations/min	Up to 500 — acceptable, 500…2500 — good, over 2500 — excellent

End of Table 2

№	Factor	Recommended application range (based on a literature review and results of our own studies)
Basic active factors		
5	Composition of a disperse medium	Rubber crumb should be solidified in a combining agent or a solution of a combining agent with bitumen. The ratio “a disperse medium: a disperse phase” should be no less than 1.25 : 1
6	Temperature of a thermal and mechanical combination, ºC	No less than 190 and no more than 250 (for rubber crumb containing natural rubber)
Integral factor		
7	Optimal time of a thermal and mechanical combination, h	Experimentally determined for each set of values of basic factors

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