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COMPARATIVE ANALYSIS OF THE IMPACT OF SYNTHETIC ADDITIVES AND PHOSPHATIDE CONCENTRATE ON THE ADHESIVE PROPERTIES OF ROAD PETROLEUM BITUMEN

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The adhesive properties of brand BND 60/90 road bitumen with the oxidized phosphatide concentrate additive were studied. It was shown that the oxidized phosphatide increases the adhesive properties of bitumen brand BND 60/90. Oxidized phosphatide concentrate can be recommended as cost-effective alternative to expensive synthetic cationic surfactants, which are now widely used as adhesives for road bitumen. Based on the results of laboratory tests, the mathematical analysis of the obtained data was performed. Comparative graphs were plotted, and Pearson's correlation coefficients and the approximation reliability values were calculated. It was found that the optimal composition for creating effective bituminous compositions with an increased thermo-physical and adhesive characteristics is 0.6 wt.% of oxidized phosphatide concentrate.

Keywords: bitumen, adhesion, surfactant, phosphatide concentrate, wetting angle.

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Introduction

Bitumen is one of the most important materials in road construction. The use of bitumen as a binder, combining the mineral components of mixtures into a single monolith, is possible due to its adhesive properties and thermoplasticity (the ability to become liquid when heated and solidify on cooling). This allows mixing bitumen with mineral materials at the temperature of 120–170^{0}C; and when the mixture hardens up to ambient temperature it turns into a solid material–asphalt concrete.

At the same time, these properties of bitumen can raise problems in the coatings operation. At high summer temperatures, bitumen softens, which under the action of transport leads to the ruts formation on the road coating. At low winter temperatures, asphalt concrete is compressed and the lack of plastic properties and bitumen brittleness causes low-temperature cracks. Bitumen has low strength and is not capable of elastic deformation, therefore there are fatigue cracks with long transport loads on the road coatings. Bitumen ages under the action of technological temperatures, oxygen and weather conditions. Its composition and structure change, it becomes brittle and loses its adhesive ability. As a result, the bearing capacity of the coating is reduced, whereas the cracking and destruction processes are accelerated. Bitumen has a selective adhesive ability (adhesion). It adheres well to alkaline rocks and bad to acidic ones. As a result, there are pits under the water action due to the bitumen film peeling from the mineral grains on the coatings. Thus, of all the asphalt concrete components, bitumen is the most sensitive to transport, technological and weather-climatic factors. Given small bitumen content in asphalt concrete (5–7%), it decisively affects the coatings durability [1,2].

The relevance of this work is related to the need to find effective and cost-effective additives to road bitumens, which will increase their adhesive properties, especially in acidic medium [3,4]. This work is a continuation of several studies on the petroleum road bitumens modification by natural and synthetic substances [5–7].

The aim of this work is to determine the possibility of using the phosphatide concentrate, a by-product of oil extraction production, as an additive for petroleum road bitumen to increase its adhesive properties.

Experimental

The raw material is BND 60/90 – petroleum bitumen with the flash point in an open crucible of 260^{0}C, and the softening temperature (determined by the ring and ball method) of 48^{0}C.

The following adhesive additives were investigated:

1. Phosphatide technical concentrate with the following characteristics: humidity of not more than 3%; mass fraction of phosphatides of not less than 40%; mass fraction of oil of not more than 60%; and substances insoluble in ethyl ether of 5%.

2. Cationic surfactants based on polyamidoamines and imidazolines of fatty acids «DAD-K Premium», «DAD-K», «DAD-C», having the following characteristics: the yield of volatile substances and moisture of not more than 0.5%; and the flash point in an open crucible of 232^{0}C.

Bitumen softening temperature was determined according to the state standard GOST 11506 by the «ring and ball» method.

The adhesive properties of bituminous composites were determined by two methods as follows:

1. Method of determining the binder adhesion to the mineral material surface. The adhesion level is assessed visually by the degree of bituminous binder film preservation on the grains of crushed stone after boiling in distilled water.

2. Adhesion properties of bitumen by the wetting angle method. In fact, adhesion and wetting are two sides of the same phenomenon. Adhesion causes the interaction between the solid phase and the adhesive, and wetting is a phenomenon that occurs due to this interaction. The edge angle is a measure of surface wetting [9]. Depending on the equilibrium wetting angle, there are three main cases:

1) the edge angle is obtuse ($180^0>\Theta>90^0$), the liquid phase does not wet the solid;

2) sharp edge angle ($0^0<\Theta<90^0$), wetting or limited wetting;

3) the equilibrium edge angle is not set, the drop spreads into a thin film (complete wetting) [10].

Results and discussion

When studying the bitumens adhesive properties, experiments were carried out with 7 different samples, including the raw material bitumen brand BND 60/90 (Figs. 1–4). The quantitative results of experiments are shown in Fig. 5.

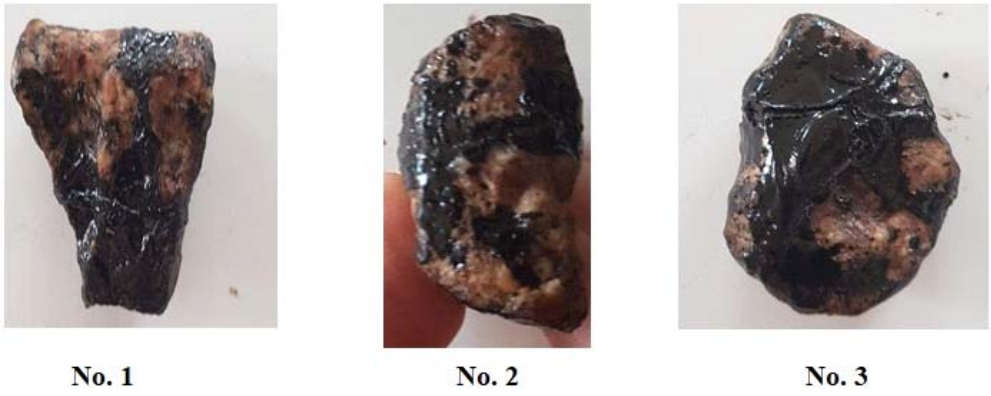

Fig. 1. Samples No. 1, 2 and 3 without phospholipids

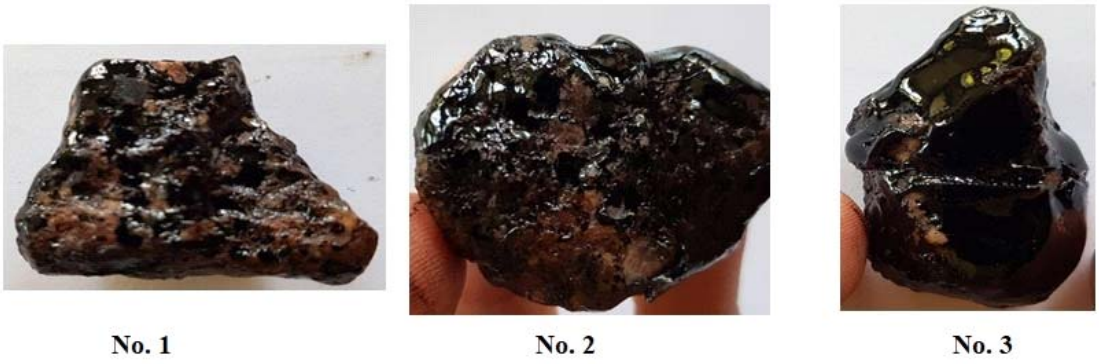

Fig. 2. Samples No. 1, 2 and 3 with the addition of 0.5% phospholipids

Fig. 3. Samples No. 1, 2 and 3 with the addition of 0.7% phospholipids

Fig. 4. Samples No. 1, 2 and 3 with the addition of 1.5% phospholipids

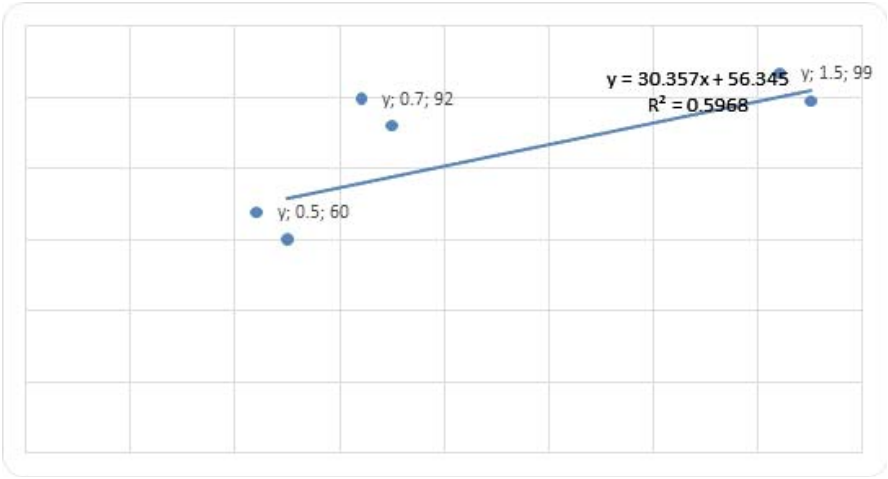

Fig. 5. The ratio between the content of oxidized phospholipid in the composite and the surface area of bitumen. The calculated Pearson correlation coefficient is equal to 0.7697

It can be seen from Fig. 5 that an increase in the content of oxidized phospholipid in bitumen results in an increase in the surface area covered by bitumen, which reaches a value of 99% when the content of oxidized phospholipid is 1.5 wt.%. It was shown that the Pearson correlation coefficients of laboratory experiments can be reproduced in further studies, and the value of the approximation reliability is close to 1, which shows a minimal discrepancy in laboratory experiments [8]. The laboratory study results proved the prospects of using phosphatide concentrate for road bitumen modification in order to achieve the

maximum surface coverage value on crushed stone grains, so its value reaches 99% for red granite and for bitumen modified with phosphatide concentrate.

Experiments were carried out to determine the wetting angle of bitumen with phosphatide (0.75 wt.%) concentrate and with well-known adhesives on black granite, red granite and glass (Figs. 6–8). Here, the sample No. 1 is a bitumen brand BND 60/90 with phosphatide concentrate addition, the sample No. 2 is a bitumen brand BND 60/90 with oxidized phosphatide concentrate addition, the sample No. 3 is a bitumen brand BND 60/90 90 with oxidized phosphatide concentrate with calcium oxide addition, the sample No. 4 is a bitumen brand BND 60/90 90 with oxidized phosphatide concentrate with diethylenetriamine addition, the sample No. 5 is a bitumen brand BND 60/90 90 with oxidized phosphatide concentrate with chlorinated lime addition, and the sample No. 6 is a pure bitumen brand BND 60/90.

The results of determining the wetting angle for bitumen with adhesive additives on red granite and glass are given in Table 1.

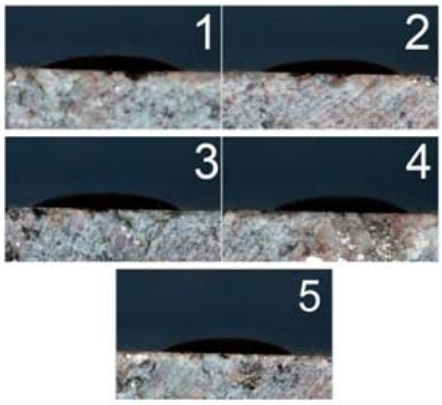

Fig. 6. Drops of researched bitumen mix on red polished granite No. 1

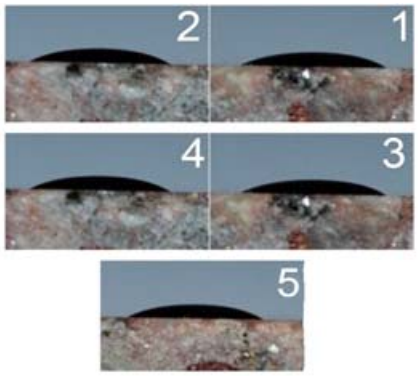

Fig. 7. Drops of researched bitumen mix on red polished granite No. 2

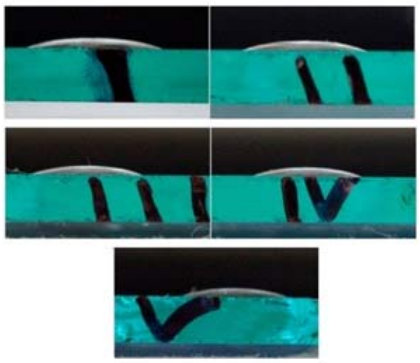

Fig. 8. Drops of researched bitumen mix on glass

Taking into account financial and economic aspects, the sample No. 2 was selected for further experiments. This adhesive additive was studied at the concentrations of 0.3%, 0.4%, 0.5%, 0.6% with respect to the total bitumen weight and the wetting angle was determined on red granite and glass (Figs. 9 and 10).

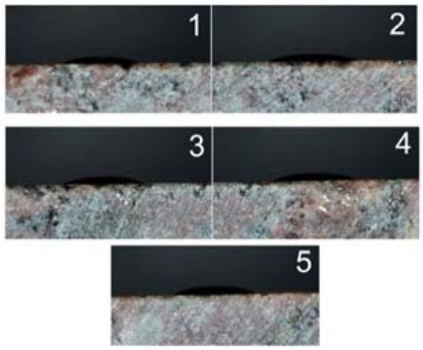

Fig. 9. Drops of the studied bitumen mix on red polished granite: 1 – additive concentration is 0.6%; 2 – additive concentration is 0.5%; 3 – additive concentration is 0.4%; 4 – additive concentration is 0.3%; and 5 – pure bitumen BND 60/90 brand

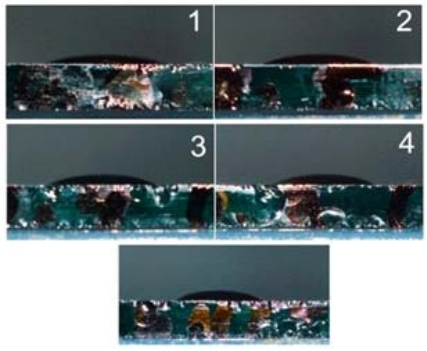

Fig. 10. Drops of the studied bitumen mix on glass: 1 – additive concentration is 0.6%; 2 – additive concentration is 0.5%; 3 – additive concentration is 0.4%; 4 – additive concentration is 0.3%; and 5 – pure bitumen BND 60/90 brand

The results of determining the wetting angle for bitumen with adhesive additives at different concentrations on red granite and glass are given in Table 2.

It can be seen from Table 2 that an increase in the content of oxidized phospholipid in bitumen results in a decrease in the wetting angle, which reaches $19^0$35' and $15^0$31' when the content of oxidized phospholipid in bitumen is 0.6 wt.% for red granite and glass, respectively. The results of laboratory studies proved the prospects of using phosphatide concentrate for the modification of road bitumens in order to achieve the smallest possible value of the wetting angle.

To determine the correlation of this method, the wetting angles of the additives with the already known adhesion value on red granite and glass were measured (Figs. 11 and 12).

Cationic surfactants based on polyamidoamines and imidazolines of fatty acids were used as known additives. This type of surfactant is widely used in asphalt-concrete production, providing a high degree of bitumen adhesion to various mineral materials that have high acidic properties.

The results of determining the wetting angle for bitumen with adhesive additives at different concentrations on red granite and glass are given in Table 3.

Table 1

The results of determining the wetting angle

Experiment	Wetting angle					
	sample No. 1	sample No. 2	sample No. 3	sample No. 4	Sample No. 5	sample No. 6
glass	$15^0$4'	$17^0$13'	$18^0$3'	$17^0$56'	$19^0$12'	$20^0$15'
red granite No. 1	$25^0$55'	$25^0$15'	$26^0$33'	$29^0$18'	$26^0$12'	$26^0$22'
red granite No. 2	$26^0$6'	$24^0$52'	$24^0$31'	$25^0$14'	$25^0$10'	$26^0$10'

Table 2

The results of determining the wetting angle

Experiment	Wetting angle				
	concentration				
	0.3%	0.4%	0.5%	0.6%	pure bitumen
red granite	$19^0$43'	$20^0$9'	$20^0$18'	$19^0$35'	$24^0$31'
glass	$24^0$37'	$21^0$47'	$20^0$46'	$15^0$31'	$28^0$37'

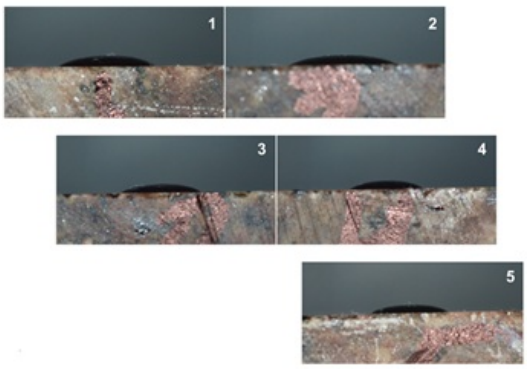

Fig. 11. Drops of the studied bitumen mix on red polished granite: 1 – bitumen with the addition of DAD-K premium; 2 – bitumen with the addition of DAD-K; 3 – bitumen with the addition of DAD-K; 4 – bitumen with the addition of DAD-C; and 5 – pure bitumen

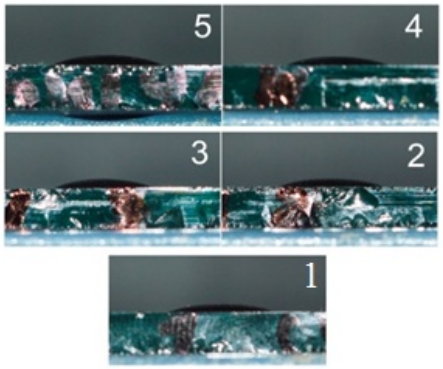

Fig. 12. Drops of the studied bitumen mix on glass: 1 – bitumen with the addition of DAD-K premium; 2 – bitumen with the addition of DAD-K; 3 – bitumen with the addition of DAD-K; 4 – bitumen with the addition of DAD-C; and 5 – pure bitumen

Table 3

The results of determining the wetting angle

Experiment	Wetting angle				
	sample				
	DAD-K premium	DAD-K	DAD-KT	DAD-C	bitumen
red granite	$23^0$4'	$20^0$15'	$21^0$34'	$23^0$46'	$22^0$37'
glass	$15^0$56'	$18^0$10'	$22^0$8'	$19^0$19'	$21^0$46'

Comparing the results given in Tables 2 and 3, we can conclude that regarding the reduction of wetting angle, the studied oxidized phospholipid significantly exceeds known surfactants, for which the wetting angle lies in the ranges of $20^0$15'–$23^0$46' and $15^0$56'–$22^0$8' for red granite and glass, respectively.

As the adhesion of bitumen is known from the literature (Table 4), further studies were conducted to determine the effect of the modification of bitumen with oxidized phospholipid on adhesion. Figure 13 shows the effect of the adhesive concentration on the wetting angle (glass as a substrate). Figure 14 shows the same dependence for red granite as a substrate.

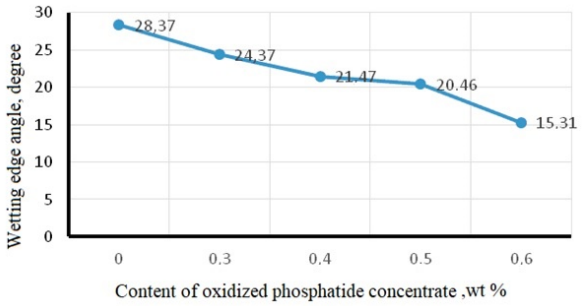

Fig. 13. Dependence of wetting angle on the adhesive additive concentration (glass as a substrate).
Pearson's correlation coefficient is equal to 0.96031

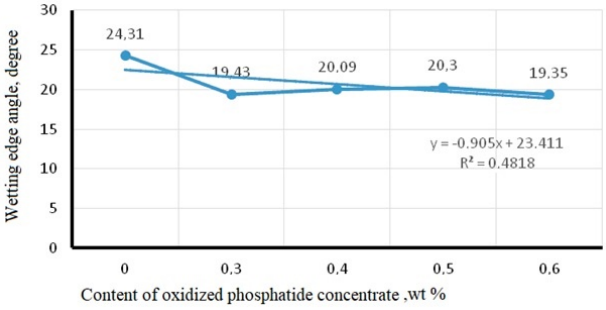

Fig. 14. Dependence of wetting angle on the adhesive additive concentration (red granite as a substrate).
Pearson's correlation coefficient is equal to 0.85746

Table 4

Adhesion value

Sample	Adhesion, %
DAD-K premium	92
DAD-K	82
DAD-KT	92
DAD-C	78
bitumen	30

Next, the correlation between the change in wetting angle and adhesion for the case of modification of bitumen with oxidized phospholipid was investigated. Figures 15 and 16 show the correlation between wetting angle and adhesion determined on glass and red granite, respectively.

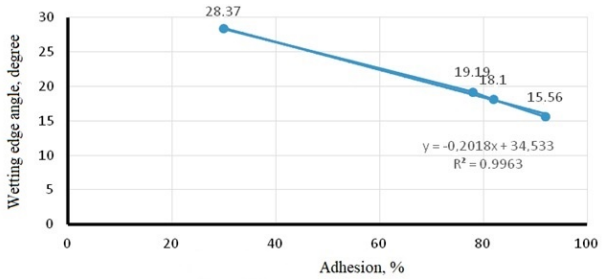

Fig. 15. Correlation between wetting angle and adhesion determined on glass as a substrate.
Pearson's correlation coefficient is equal to 0.99814

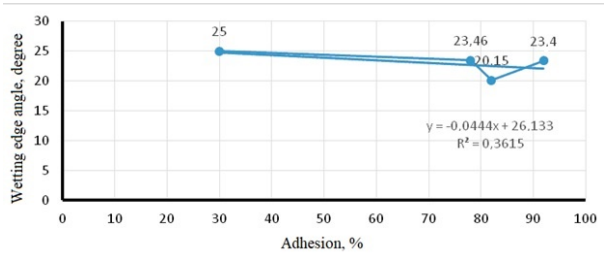

Fig. 16. Correlation between wetting angle and adhesion determined on red granite as a substrate.
Pearson's correlation coefficient is equal to 0.60126

It follows from Figs. 15 and 16 that modification of bitumens with oxidized phospholipid allows significantly increasing adhesion from 30% to 92% due to the reduction of wetting angle. In addition, modification of bitumens with oxidized phospholipid allows significantly increasing their softening temperature from 48^{0}C to 53^{0}C, as evidenced by the «ring and ball» method. Thus, this new bitumen–rubber composition with significantly improved basic physical and mechanical properties can be recommended for use in construction.

Conclusions

Based on analysis of the literature and results of experimental research, we developed a new additive based on oxidized phosphatide concentrate, which can reduce the cost of road bitumen while maintaining efficiency as compared with known additives based on cationic surfactants. The bitumen–polymer composition modified by adding 0.6% of oxidized phosphatide concentrate proved to be optimal; its adhesion is 92% and 96% on red granite and n glass, respectively. According to the results of laboratory tests, mathematical analysis was performed. Comparative graphs were plotted, and Pearson's correlation coefficients and the approximation reliability value were calculated.

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ПОРІВНЯЛЬНИЙ АНАЛІЗ ВПЛИВУ СИНТЕТИЧНИХ ДОБАВОК І ФОСФАТИДНОГО КОНЦЕНТРАТУ НА АДГЕЗІЙНІ ВЛАСТИВОСТІ ДОРОЖНІХ НАФТОВИХ БІТУМІВ

Лаврова І.О., Демидов І.М., Черкашина Г.М., Лебедєв В.В., Забіяка Н.А.

Здійснено дослідження адгезійних властивостей дорожнього бітуму марки БНД 60/90 з добавкою окисненого фосфатидного концентрату. Показано, що окислений фосфатид підвищує адгезійні властивості бітуму марки БНД 60/90. Окислений фосфатидний концентрат можна рекомендувати як економічну альтернативу дорогим синтетичним катіонним поверхнево-активним речовинам, які зараз широко використовуються як адгезиви для дорожніх бітумів. За результатами лабораторних досліджень виконано математичний аналіз отриманих даних. Побудовано порівняльні графіки, визначено коефіцієнти кореляції Пірсона і значення достовірності апроксимації. Встановлено, що оптимальним складом для створення ефективних бітумних композицій з підвищеним теплофізичним комплексом і адгезійними характеристиками є 0,6 мас.% окисленого фосфатидного концентрату.

Ключові слова: бітум, адгезія, поверхнево-активні речовини, фосфатидний концентрат, кут змочування.

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Keywords: bitumen; adhesion; surfactant; phosphatide concentrate; wetting angle.

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