Vol. 7, No. 3, 2013

Chemical Technology

Mohammad Al-Ameri, Oleh Grynyshyn and Yuriy Khlibyshyn

MODIFICATION OF RESIDUAL BITUMEN FROM ORHOVYTSKA OIL BY BUTONAL POLYMERIC LATEXES

Lviv Polytechnic National University 12, S.Bandery str., 79013 Lviv, Ukraine

Received: January 15, 2013 / Revised: March 04, 2013 / Accepted: May 02, 2013

© Al-Ameri M., Grynyshyn O., Khlibyshyn Yu., 2013

Abstract. Modification of residual bitumen from Orhovytska oil by Butonal polymeric latexes has been studied. Using such modifiers the softening temperature and bitumen elasticity increase and penetration decreases. The commercial paving bitumen may be produced from the residuals using Butonal polymeric latexes.

Keywords: bitumen, modification, polymeric latex, Butonal.

1. Introduction

The problem of high-quality petroleum bitumen production meeting the modern requirements has to be solved by two directions simultaneously:

- materials upgrading grounded on refinery technological possibilities. Such bitumen should have the optimum composition and structure ensuring their increased service life and improved properties;

- drastic improvement of bitumen physicomechanical properties *via* complex modification by adhesive or cross-linked additives, polymers most of all.

Orhovytska oil with unique properties is produced in Lviv region [1]. It is heavy high-sulphuric oil with the sulphur content of more than 6.0 mas % and total content of asphaltene-tar compounds of 33.0 mas %. The main product of this oil processing is residual (distillate) bitumen which does not meet the requirements of standards relative to commercial bitumen. Its quality depends only on the amount of distilled fractions from Orhovytska oil. One of the directions aimed at the improvement of bitumen properties is its modification by polymeric materials.

In the world practice various modifiers are used to improve the bitumen properties. Polymers, namely polyethylene, polypropylene, copolymers of styrenebutadiene-styrene type, *etc.* are the most widespread modifiers [2-5].

Among all modifiers the latexes of Butonal type (copolymers of styrene and butadiene crosslinked by sulfur) produced by BASF firm take the special stand [6]. However it is not known how the properties of residual bitumen from Orhovytska oil will be changed when using the mentioned modifiers.

The aim of this work was to study the modification of residual bitumen from Orhovytska oil by Butonal polymeric latexes.

2. Experimental

The raw material for the production of bitumen modified by polymeric latexes was the residual bitumen (> 773 K) from Orhovytska oil with the following characteristics:

-	softening temperature in accordance with "ring and ball" method	310 K
-	ductility at 298 K	> 100 cm
-	penetration at 298 K	106·0.1 mm
-	flash point in the open firepot	531 K
-	mass part of water	traces
-	density at 293 K	998 kg/m ³

Table 1

Characteristic of Butonal type latexes

		Index values	
Normative document	Index	Butonal	
		NS 104	NS 198
BASF product	Appearance	Liquids of white color	
specification:	Polymer content, %	71 ± 1	64 ± 1
ISO 1625,	Concentration of hydrogen ions, pH	10.0-10.5	4.0-4.5
DIN 53189,	Density, g/cm ³	1.0	0.94
ISO 8962	Emulsifier type	anionic	cationic

To modify bitumen we used latexes Butonal NS 198 and Butonal NS 104. They are thermoplastic elastomers representative by themselves the aqueous dispersion of butadiene and styrene copolymers with conglomerated sulfur. The latexes characteristic is given in Table 1.

The bitumen modification was carried out at the laboratory plant allowing to mix the components under definite technological regimes. The operating sequence was as follows:

- bitumen warming to the operating temperature (413–473 K);

- introduction of Butonal necessary amount into bitumen under constant stirring;

- holding of bitumen at operating temperature under constant stirring for definite time of modification necessary to obtain physico-mechanical properties.

Penetration, softening temperature and elasticity of obtained petroleum bitumen were determined in accordance with known methods.

3. Results and Discussion

To achieve the maximal effect of polymeric latexes usage and study their influence on bitumen physico-chenical properties it is necessary to determine optimum operating parameters of the process. Therefore we investigated the effect of preparation conditions for bitumen modified by polymeric anionic latex Butonal NS 104 on its properties.

The results show that introduction of polymeric latex Butonal NS 104 into bitumen decreases its penetration (Fig. 1). The main change in penetration is observed for the first three hours of bitumen modification. After this time the penetration is not changed. Thus 3 h are enough to stabilize the penetration. Then bitumen type may be determined. The increase of temperature from 433 to 473 K slightly increases the penetration (Fig. 2). This result allows to assume that modification proceeds incompletely at 433 K.

The values of penetration decrease of the modified bitumen obtained at 453 and 473 K are almost

the same. It should be noted that the increase in preparation temperature, as well as the increase of polymer amount (to 6%) allows to convert bitumen into more viscous one. This property of obtained modified bitumen must be taken into account while preparing, paving and consolidating polymer-asphalt-concrete mixture.

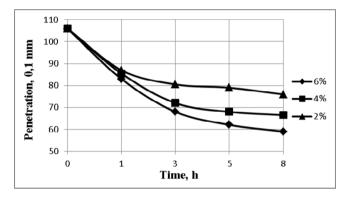


Fig. 1. Bitumen penetration at 298 K vs preparation time at different amounts of Butonal NS 104

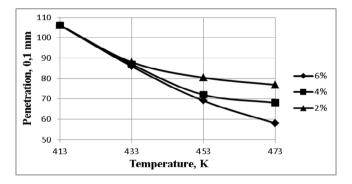


Fig. 2. Bitumen penetration at 298 K vs preparation temperature at different amounts of Butonal NS 104

The introduction of Butonal NS 104 into bitumen increases its softening temperature (Fig. 3). The main increase is observed for the first hour of modification and then the change is insignificant. It should be noted that modification was carried out at 453 K. The change of temperature within the range of 433–473 K shows that 433 K is the insufficient temperature for obtaining the

homogeneous mixture. At the same time the slow increase of heat-resistance at 473 K indicates that aging processes are actually not developed (Fig. 4).

To examine the possible aging of bitumen under the action of high temperatures we studied its elasticity depending on modification time and temperature.

Fig. 5 shows that the introduction of Butonal NS 104 into bitumen increases its elasticity. The increase of modifier amount above 4 % is pointless because it does not affect the bitumen elasticity. The maximum elasticity is observed at the preparation temperature of 453 K (Fig. 6).

The experimental results show that bitumen modified by polymeric latex Butonal NS 104 meets all

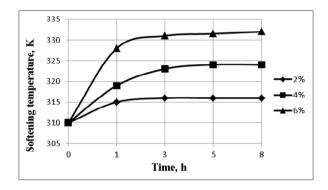


Fig. 3. Softening temperature vs preparation time at different amounts of Butonal NS 104

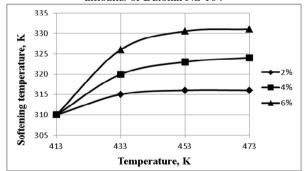


Fig. 4. Softening temperature *vs* preparation temperature at different amounts of Butonal NS 104

requirements for standard bitumen modified by polymers. The short preparation time allows to preserve energy supply and it is the essential advantage of this type of bitumen compared with others. The optimum amount of polymeric latex Butonal NS 104 was found to be $2-3 \mod \%$, optimum preparation time -2-3 h and optimum preparation temperature -443-453 K.

To extend the assortment of polymeric modifiers used for modification of the residual bitumen from Orhovytska oil we also investigated cationic latex Butonal NS 198. The main regularities of the previous modifier (Butonal NS 198) are typical for Butonal NS 198 as well (Table 2).

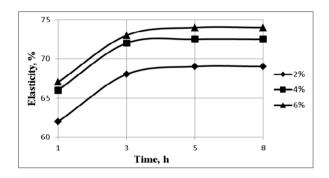


Fig. 5. Bitumen elasticity vs preparation time at different amounts of Butonal NS 104

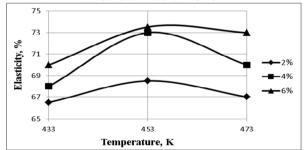


Fig. 6. Bitumen elasticity vs preparation temperature at different amounts of Butonal NS 104

Table 2

Dependence of modified bitumen properties on the amount of polymer Butonal NS 198 and modification time

		1 0				
Modification	Penetration at 298 K, 0.1 mm		Softening temperature, K		Elasticity, %	
time, h	at Butonal NS 198 content of		at Butonal NS 198 content of		at Butonal NS 198 content of	
unite, fi	2 mas %	4 mas %	2 mas %	4 mas %	2 mas %	4 mas %
0	106	106	310	310	31	31
1	101	96	317	318	65	73
2	97	90	320	322	71	75
4	89	83	321	325	72	76
6	77	74	322	327	73	77

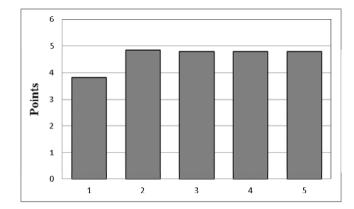


Fig. 7. Dependence of bitumen cohesion with mineral matter on the amount of modifier Butonal NS 198 (%): 0 (1); 2 (2); 3 (3); 4 (4) and 6 (5)

The dependence of bitumen cohesion with the mineral matter upon the amount of polymeric modifier confirms its constancy at both minimal amount of polymer (2%) and maximal one (6%). Thus, for non-modified bitumen the cohesion index is about 3 points, for the modified bitumen it was found to be 5 points (Fig. 7).

The results of investigations show that the polymer-bitumen based on the residual bitumen from Orhovytska oil and cationic latex Butonal NS 198 meets the existing requirements. The improvement of the main properties of the modified bitumen due to the introduction of modifier depending on its amount and preparation temperature indicates the possibility of bitumen properties control under the operational conditions. Cationic latex Butonal NS 198 may be used for the bitumen modification to improve its quality, especially to increase its elasticity.

We compared the operational properties of obtained bitumen with requirements of standard relative to paving bitumen. The modification of the residual bitumen from Orhovytska oil by polymeric latex Butonal (2–3 mas %) at 453 K for 2–6 h allows to produce bitumen corresponding to BND-60/90 type by its properties.

4. Conclusions

The efficiency of the residual bitumen from Orhovytska oil modification by Butonal polymeric latexes has been confirmed. The introduction of 2-3 % of such modifiers allows to increase the bitumen hardness, heat-resistance and elasticity.

The optimal parameters of the modification process are: temperature 443-453 K and stirring time 2-3 h.

We showed the possibility of high-quality commercial paving bitumen production on the basis of residual bitumen obtained during Orhovytska oil processing and Butonal polymeric latexes.

References

Pavlyuh J. and Pavlyuh O.: Naftova i Gazova Prom., 2005, **3** 15.
Leonenko V. and Safonov G.: Khim. i Techn. Topliv i Masel, 2001, **5**, 43.

[3] Zolotarev V.: Avtoshlyahovyk Ukrainy, 2003, 5, 25.

[4] Kishchynskyi S.: Avtoshlyahovyk Ukrainy, 2003, 6, 28.

[5] Shevchenko V., Romanenko S., Rebenok O. *et al.*: Avtoshlyahovyk Ukrainy, 2003, **1**, 27.

[6] Pronyk Yu., Vyrozhemskyi V. and Kishchynskyi S.: Avtoshlyahovyk Ukrainy, 2004, **1**, 41.

МОДИФІКАЦІЯ ЗАЛИШКОВОГО БІТУМУ ОРХОВИЦЬКОЇ НАФТИ ПОЛІМЕРНИМИ ЛАТЕКСАМИ BUTONAL

Анотація. Вивчено процес модифікації залишкового бітуму, одержаного при переробленні орховицької нафти, полімерними латексами Butonal. Встановлено, що при використанні таких модифікаторів зростає температура розм'якшення і еластичність бітумів та зменицується їх пенетрація. Показано, що з використанням латексів Butonal на основі залишку орховицької нафти можна одержувати товарні дорожні бітуми.

Ключові слова: бітум, модифікація, полімерний латекс, Butonal.