UDC 665.6
DOI: 10.15587/1729-4061.2022.257782

THE USE OF POLYETHYLENE TEREPHTHALATE WASTE AS MODIFIERS FOR BITUMEN SYSTEMS

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This study examined the suitability of polyethylene terephtal (PET) waste to provide an alternative modification to bitumen and reduce waste accumulation. The chemical structure and physical properties were evaluated for modified bitumen with different PET flakes content from 2 to 10 %. The effect of PET waste content on bituminous systems was analyzed using IR and 1H NMR spectroscopy, differential thermal (DTA) and thermogravimetric (TGA) analyses. The study of change in the microstructure as a result of bitumen modification was carried out using atomic force microscopy (AFM). Regularities of changes in the structural-group composition of bituminous binders after their modification with PET waste have been established. It is noted that the chemical interaction of the base bitumen with PET flakes occurs due to the formation of associative bonds between the oxygen-containing components of the modifier and bitumen.

The influence of the modifier on the physical and mechanical properties was evaluated using standard methods (penetration, extensibility and softening point). Based on the physical and mechanical properties of bitumen, such as needle penetration depth (penetration), ductility and softening point, it was found that the optimal dosage of PET waste in terms of asphalt binder characteristics is 3 %. It has been established that PET waste and original bitumen interact both at the chemical and physical levels and can be considered as a suitable alternative for changing the properties of bituminous binders. Thus, the obtained samples of modified bituminous binders have improved physical and mechanical properties, which makes it possible to produce high-strength asphalt concrete pavements based on them

Keywords: PET waste, municipal solid waste, bituminous binder, polymer modified bitumen, green chemistry

Received date 21.03.2022
Accepted date 25.05.2022
Published date 30.06.2022

***How to Cite**: Aitkaliyeva, G., Yelubay, M., Yerzhanova, D., Ismailova, A., Massakbayeva,S.(2022). The use of polyethylene terephthalate waste as modifiers for bitumen systems. Eastern-European Journal of Enterprise Technologies, 3 (6 (117)), 6–13. doi: https://doi.org/10.15587/1729-4061.2022.257782*

1. Introduction

Today, it is estimated that billions of tons of plastic have been produced [1] and due to the progressive accumulation of plastic waste over the past decades [2], the problem of their disposal has received much attention. Landfilling and incineration, which are commonly used to treat plastic waste, contribute significantly to CO_2 emissions and, thus are associated with serious environmental problems [3]. Pollution of the marine environment with microplastics, caused by the slow degradation of plastic waste, is another global environmental problem [4]. Robust methods of converting plastic waste into useful materials are increasingly needed to ensure the green and sustainable use of fossil resources, but the available chemical technologies are still limited to basic synthetic processes.

Polyethylene terephthalate (PET), the world's fourth most produced polymer [5], is a thermoplastic polymer, whose recycling has attracted widespread attention to ensure efficient waste management. At the moment, there is no existing effective technology for processing this type of waste, and therefore, the development of an effective method for their processing is an urgent task, the solution of which will make the production of polyethylene terephthalate and products based on it economically profitable and environmentally safe.

There is no effective recycling technology for PET waste at the moment. Therefore, research on the development of an effective method for the disposal of PET flakes is relevant.

2. Literature review and problem statement

The paper [3] presents the results of the mechanical processing of PET, where it is physically crushed and melted to obtain textiles or used as a raw material at low loads to obtain primary PET materials. However, only 15 % of waste is processed by this method [5].

Chemical methods associated with depolymerization to reproduce PET have become widespread. In this regard, it is necessary to pay due attention to the suitability for further processing of the resulting polyester, which limits the use of processes by mechanical processing, hydrolytic and thermal decomposition [6].

Among chemical methods, of interest is the process of glycolysis [7], in which transesterification occurs in PET chains with diols to form monomers. The work [8] describes a method for further processing of depolymerized PET monomers by amminolysis, where depolymerization is carried out with nitrogen-containing compounds. Despite all the advances in the production of new materials based on depolymerized PET, chemical recycling remains an expensive method due to the large volumes of chemicals and the necessary labor-intensive product purification steps, which indicates the need to find new efficient and cheap methods for processing PET waste.

A review of the literature indicates a very strong interest in the possibility of using PET waste to modify building materials, especially concrete and mortar. A number of researchers have used this waste as a replacement for aggregate in cement concrete or as fiber reinforcement. Numerous publications describe the possibility of synthesizing polyester resin based on PET waste; some research centers have demonstrated the possibility of obtaining epoxy resin in this way [9].

An interesting solution to the problem of recycling this plastic is the production of cementless composites based solely on PET waste and aggregate. There are much less numerous works both with PET waste and resin-based composites [10]. It is mainly aimed at a resin-based mortar obtained by partially replacing the aggregate with the described recyclate [11]. It is also noted that PET waste is one of the most intensively studied plastic wastes, which can replace natural aggregate in cement composites [12].

All this allows us to state that it is expedient to conduct research on the study of PET flakes as a component of road surfaces.

It is known [13] that one of the most promising and widely used methods for processing polymers and their wastes is the production of modifying additives for bituminous binders. In this regard, this study explores the potential of PET waste as a BND 70/100 bitumen modifier.

3. The aim and objectives of the study

The aim of the study is to develop a polymer-bitumen material with an improved set of physical, mechanical, and operational properties. It will solve the practical problem of PET waste disposal.

To achieve the aim, the following objectives were set:

– to study the structure of the composition based on bitumen and PET waste;

– to study the properties of polymer-bitumen compositions based on bitumen and PET waste.

4. Materials and methods

4. 1. Method of samples preparation

Polyethylene terephthalate waste used in this study was obtained from water bottles, separated, washed, and ground. IR and ^{1}H-NMR spectra of PET waste are shown in [14].

Modification of bitumen with PET waste was carried out as follows: bitumen was constantly mixed and heated at 150 °C for 40 min, after which pre-dried PET waste was introduced in an amount of 2, 3, 8, 10 wt%.

4. 2. Methods of investigating the samples' characteristics

A study of the structural-group composition of bitumen before and after modification with PET waste in an amount of 2 to 10 wt% was carried out by IR, ^{1}H NMR spectroscopy.

To register changes in thermochemical and physical parameters during heating of bitumen before and after its modification, the method of differential thermal (DTA) and thermogravimetric (TGA) analysis was used, performed on a Q-1000D derivatograph in air, in the temperature range from 20 to 1000 °C.

To better understand the mechanism of change in chemical components, the method of atomic force microscopy (AFM) was used on the Solver Spectrum setup.

Studies of the penetration of bituminous binders were carried out according to GOST 11501–78 "Method for determining the depth of penetration of a needle", ductility according to GOST 11505–75.

5. Results of the study of the composition and properties of polymer-modified bitumen

5. 1. Study of the structure of polymer-bitumen compositions

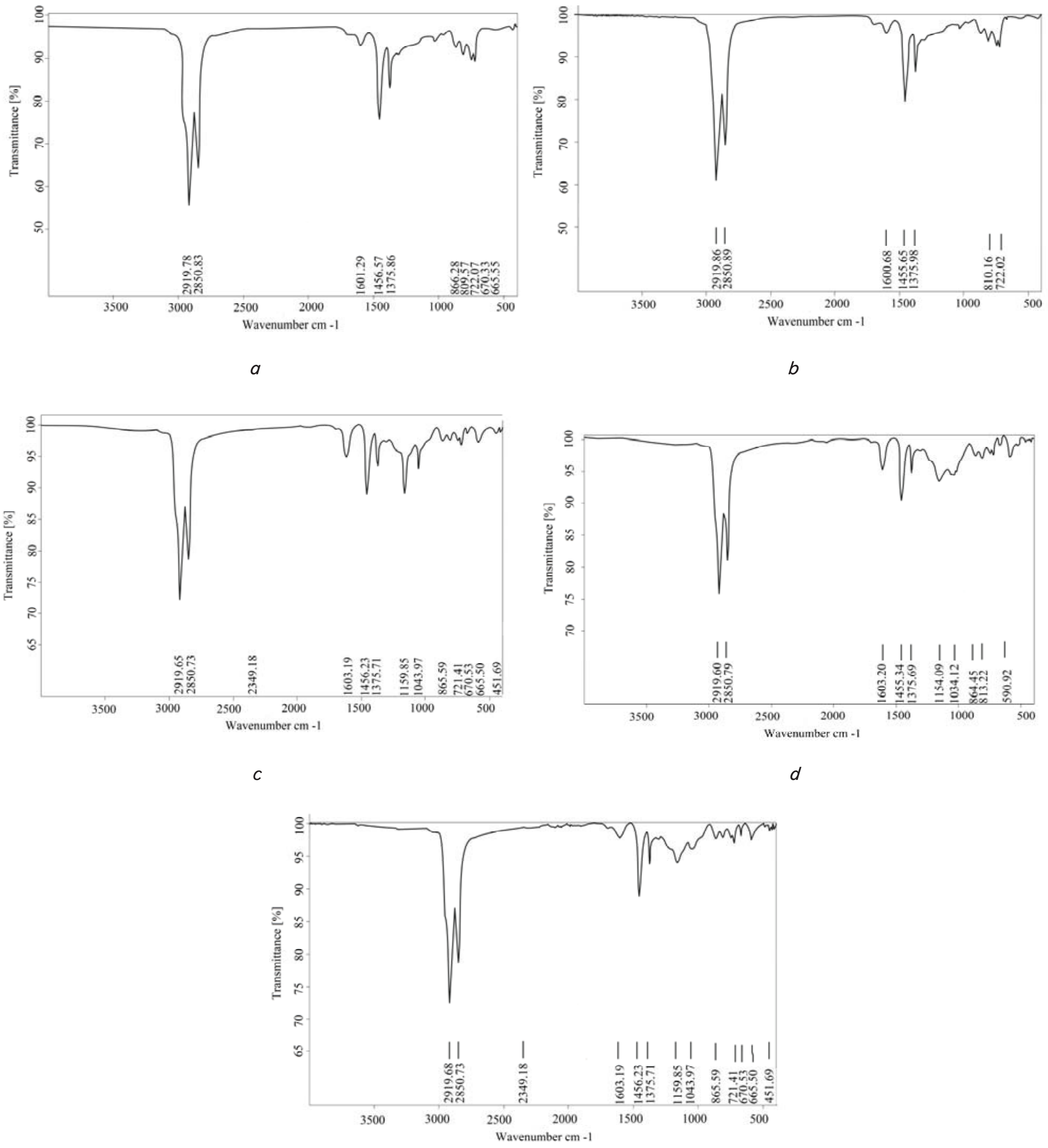
IR spectra of the original and modified bitumen are shown in Fig. 1.

On the IR spectrum of PET flakes [14], high-intensity stretching vibrations of the carbonyl group are observed in the range of 1,715 cm^{-1}. It is known that the stretching vibrations of the C–O group give a number of strong bands in the range of 1,300–1,000 cm^{-1}: in the spectrum of the PET flakes, one can distinguish the average vibration frequencies νC–O for several esters with a long hydrocarbon chain with strong peaks at 1,270 and 1,130 cm^{-1}. The peak in the region of 1666 cm^{-1} is typical for double C=C bonds in aromatic compounds, and the bands of out-of-plane bending vibrations of the unsaturated C-H bond lie in the region of 1,000–800 cm^{-1}, which confirms the presence of a benzene ring in the PET sample.

To establish the nature of the interaction between the PET components and the bituminous binder matrix, the IR spectroscopy method was used.

The IR spectra of the PET-modified samples (Fig. 1) with the initial bitumen and PET flakes are practically the same, except for an additional band at 1130 cm^{-1} in the modified bitumen. This indicates the presence of the carbon-oxygen functional group C-O, which is characteristic of PET flakes.

The ^{1}H NMR spectrum of PET (Fig. 2, *a*) showed a singlet at 8.1 ppm corresponding to the aromatic ring proton. The triplet at 4.7 ppm was assigned to the methylene group ($-CH_2-$) adjacent to the ester bond [15]. The 3.52 ppm singlet refers to the methylene groups referred to as protons in the methylene group in $-CH_2-OH$.

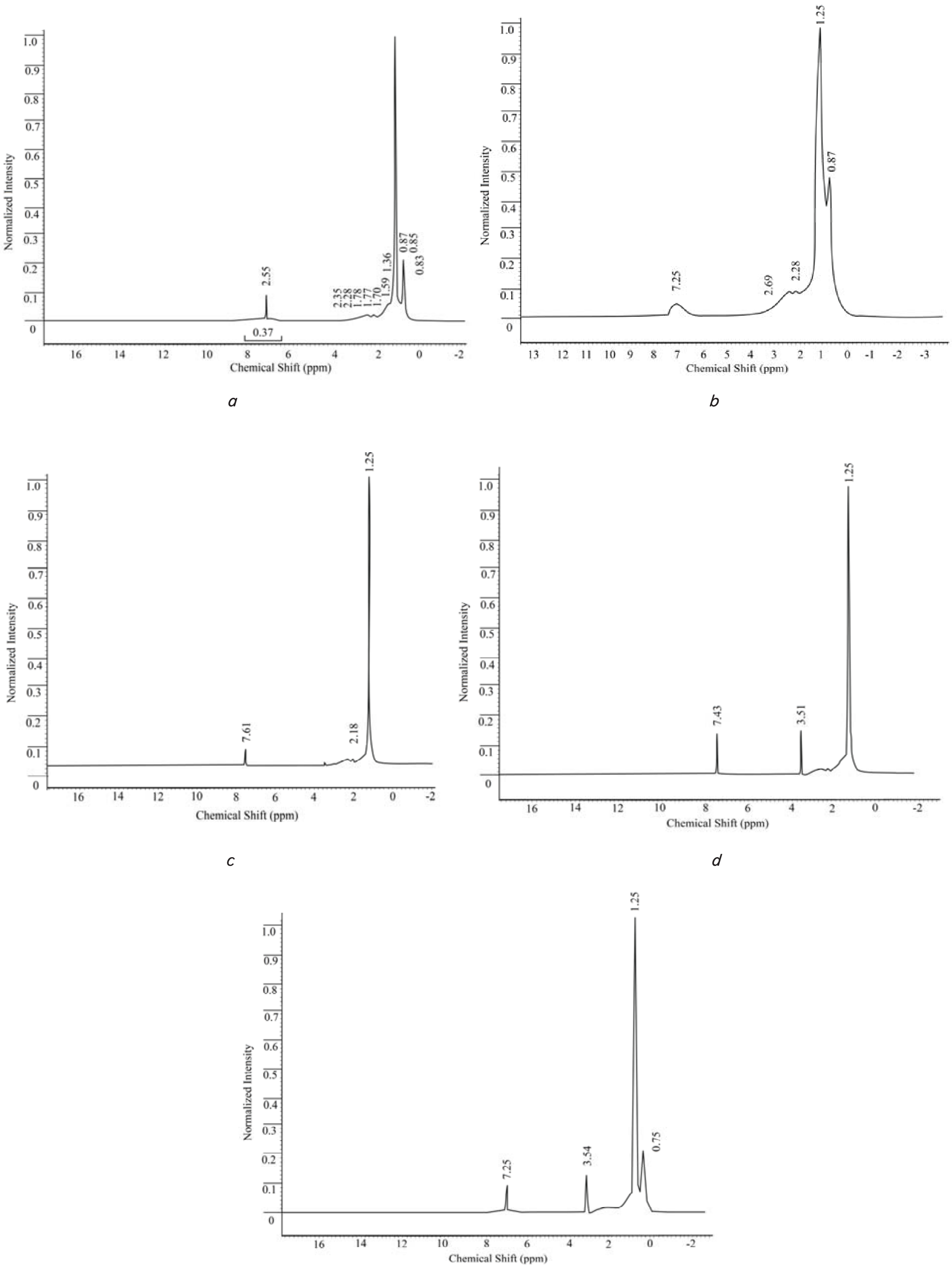


a *b* *c* *d* *e*

Fig. 1. Infrared spectra of the bitumen samples: *a* – original; *b* – polymer-modified in the amount of 2 %; *c* – polymer-modified in the amount of 3 %; *d* – polymer-modified in the amount of 8 %; *e* – polymer-modified in the amount of 10 %

The results of the ^{1}H NMR spectrum of the initial bitumen show that it contains several chemical shift signals from 0.83 to 2.55 ppm. These signals can be divided into aliphatic proton signals from alkanes (0.8 to 1.4 ppm) and protons of alkenes, alkynes, or aliphatic compounds with a carbonyl group such as a ketone, aldehyde, ester, and carboxyl group (1.5 to 2.5 ppm) [13]. The presence of a chemical shift at ~7 ppm corresponds to the proton of the aromatic group [16].

Fig. 2, *b*, *e* shows the NMR of bitumen modified with various amounts of PET. In the modified bitumen, a new signal was found at 3.5 ppm, which indicates the proton of the $-CH_2-OH$ group. The formation of this signal indicates a chemical interaction between bitumen and PET. An increase in the intensity of this signal corresponds to an increase in the PET content in the samples.



a

b

c

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Fig. 2. ^{1}H Nuclear magnetic resonance spectra of the bitumen samples: *a* – original; *b* – polymer-modified in the amount of 2 %; *c* – polymer-modified in the amount of 3 %; *d* – polymer-modified in the amount of 8 %; *e* – polymer-modified in the amount of 10 %

5. 2. Study of the properties of polymer-bitumen compositions based on bitumen and PET waste

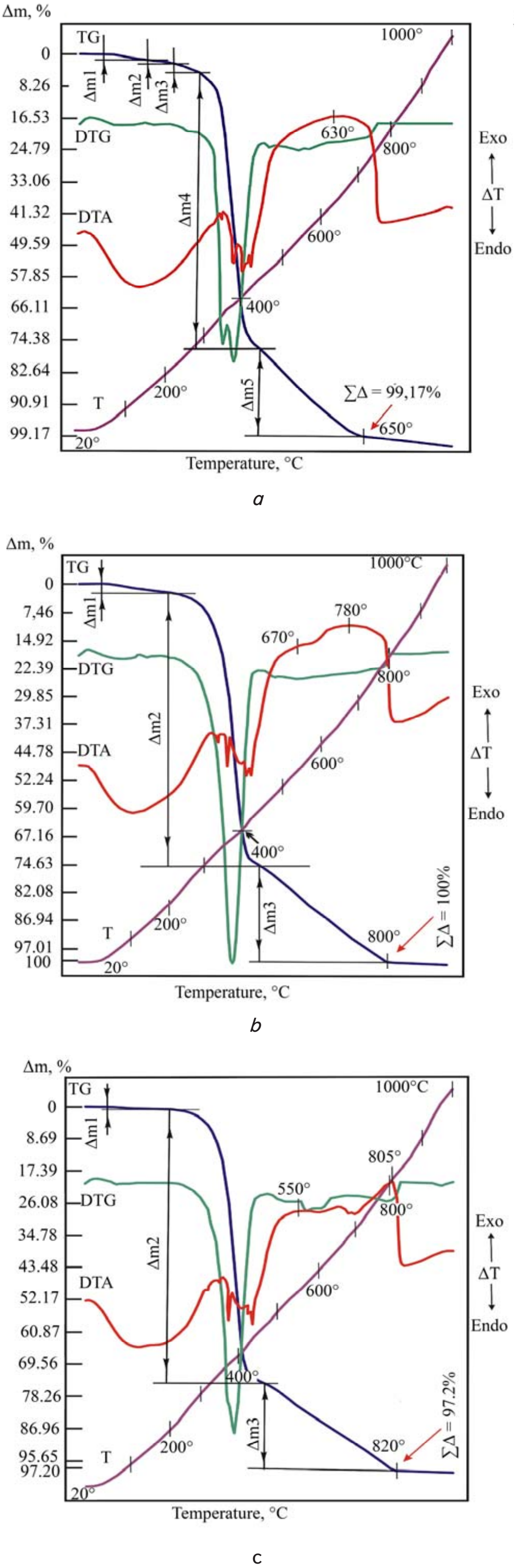
To establish the thermal behavior of bitumen samples before and after adding PET to its composition, the method of differential thermal and thermogravimetric analysis was used, the results of the studies are presented in Fig. 3.

During the heating of bitumen samples, it is possible to conditionally determine the stages of their decomposition on the TG curve in the temperature range of 20–280 °C, 280–450 °C and 450–720 °C (Fig. 3, *a*). It is noted that for the original bitumen sample in the low-temperature heating range, three depressions formed on the DTG curve (at 90, 170 and 260 °C). They indicate the presence on the TG curve in the range of 20–280 °C of three weight loss steps of 5.3 %, corresponding to the emissions from the system of three portions of weakly bound aromatic inclusions of bitumen and partly of its combustion product (CO_2).

The next stage of decomposition proceeds in the range of 280–720 °C. The thermal oxidation process proceeds in two stages within 280–450 °C and 450–720 °C. In the range of 280–450 °C, combustion takes on an explosive character, in which more than 71 % of the bitumen is lost. The second stage of the interaction of bitumen with oxygen (450–720 °C) occurs at a moderate rate, when the less active part of the organic matter enters the reaction. In the specified temperature range, the remaining part of 22.7 % of bitumen is carried away in the form of CO_2 into the atmosphere. The total weight loss of all emissions in the range of 20–720 °C was 99.7 %, which corresponds to 0.3 % ash content of the tested system.

For the modified samples of bitumen (Fig. 3, *b*, *c*) there is a decrease in the content of light fractions. On the thermal curves of the modified sample containing 6 % PET (Fig. 3, *b*) in the low temperature range (20–200 °C), the bitumen weight loss is 2.1 %. Within the same temperature ranges, the differential thermogravimetric curve recorded two weakly pronounced depressions at 80 and 140 °C, indicating a separate two-stage emission of weakly bound carbon oxides into the atmosphere. In relation to the original sample, the modification of bitumen also led to minor changes in its weight loss in the main heating temperature ranges (20–200, 200–405 and 405–800 °C).

A sample of bitumen with a content of 10 % PET (Fig. 3, *c*) is characterized in the range of 20–190 °C by a weakly expressed degree of weight loss (0.7 %), caused by the removal of loosely bound particles of organic origin from the system. At higher temperatures of 190–825 °C, the DTA curve registers a powerful exothermic manifestation caused by the oxidation of organic matter with the release of carbon dioxide into the atmosphere. The removal of CO_2 is carried out in two stages. The first stage of OM combustion proceeds within the range of 190–470 °C with the intensive formation of carbon monoxide and its removal from the system. In the specified temperature range, the bulk of the sample burns out. This is evidenced by the presence on the TG curve (in the marked temperature range) of a sheer step, the weight loss equal to 73.5 % of the total mass of the sample. The second stage of bitumen oxidation takes place in the range of 470–825 °C, and at this stage of the interaction of oxygen with organic matter, the system loses another 23.0 % of its mass, bringing the total weight reduction of the sample to 97.2 %.



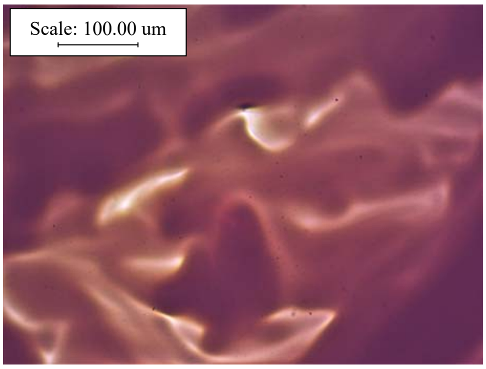
a

b

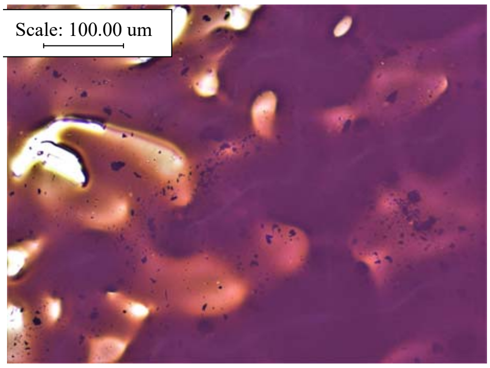
c

Fig. 3. Derivatogram of the bitumen sample: *a* – original; *b* – polymer modified bitumen in the amount of 4 %; *c* – polymer modified bitumen in the amount of 10 %

To study the microstructure and deeper understand the complex mechanism of changes in the chemical composition of bitumen, the method of atomic force microscopy (AFM) was involved. Fig. 4 shows AFM images of bitumen before and after the introduction of the modifier.



a



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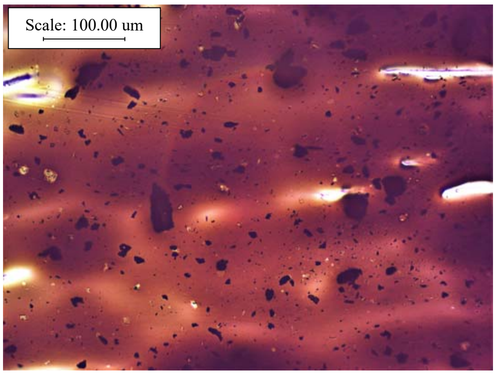

c

Fig. 4. Atomic-force microscopy images of bitumen samples: *a* – original; *b* – polymer modified bitumen in the amount of 4 %; *c* – polymer modified bitumen in the amount of 10 %

Based on the images obtained, it can be seen that the modification of bitumen leads to a change in its structure at the microlevel. The formation of "bee" structures for modified bitumen samples is noted. The authors of [17] explain the formation of these structures by the aggregation of asphaltene components, which indicates an increase in their content in bitumen samples.

The impact of PET waste on modified bitumen properties was evaluated based on physical performance including penetration depth, ductility, and softening temperature (Table 1).

Based on the data obtained, it was found that the improvement in the performance properties of bitumen occurs when a polymer modifier is introduced, even in an amount of 2 %. From the analysis of Table 1, it can be seen that the penetration depth of the needle was significantly reduced at a PET concentration of 3 %, indicating a decrease in fluidity and an improvement in the high temperature consistency of the bitumen.

Table 1

Physical and mechanical properties of bitumen BND 70/100 after adding a modifier to its composition

Indicator	Value					
Modifier quantity, %	Without modification	2	3	4	8	10
Needle penetration depth, 0.1 mm at 25 °C	73	65	61	62	64	64
Extensibility, mm	70	49	38	38	43	43
Softening temperature, °C	52	54	57	56	55	55

The extensibility of samples of modified bitumen noticeably decreases with the addition of polymer waste from 3 to 4 wt%, then an increase in this indicator is noted. It is known that penetration is an indirect characteristic of the degree of hardness of bitumen samples. In particular, when entering the modifier in the amount of 3 wt%, there is a decrease in the needle penetration depth (at 25 °C) by 12 mm.

One of the most important characteristics of bitumen is its softening temperature. As expected, the bitumen softening point value is increased by the addition of the PET modifier, as shown in Table 1. This fact indicates that the PET waste/bitumen mixtures will show a significant improvement at high pavement service temperatures. So, the softening temperature of the original bitumen was 52 °C, and its modification with PET waste in the amount of 3 wt% PET has an increase in softening point up to 57 °C.

6. Discussion of the results of studying the composition and properties of polymer-modified bitumens

The combination of IR and ^{1}H NMR spectra of the original and modified bitumens indicates the formation of a new oxygen-containing functional group. There is a tendency to increase the intensity of the polyfunctional group with increasing concentration of PET. The formation of new oxygen-containing fragments can occur due to associative bonds between the oxygen-containing components of the modifier and bitumen [18].

When analyzing ^{1}H NMR spectra of bitumen samples, scientists [13] divided the signals into two regions: 0.8–1.4 ppm in the region for protons of aliphatic alkanes, and the region 1.7–3.5 ppm is a proton of alkenes, alkynes. Based on the ^{1}H NMR spectra of samples of the original and modified bitumen (Fig. 2), it can be concluded that, when bitumen is modified, the oxygen-containing group of PET waste interacts with alkenes in bitumen, since the area at 2.2 and 2.5 ppm disappears in the composition of the original bitumen. The appearance of new functional groups in the modified bitumen indicates the chemical interaction of the base bitumen with the polymer during mixing, which indicates their compatibility.

DTA/TG and AFM studies revealed an increase in the content of asphaltene components in the composition of modified bitumen 8 % and 10 % PET flakes. It can be assumed that the transformation of aromatic compounds and resins into asphaltenes occurs and leads to the strengthening of bitumen [19], and is also favorable for viscosity and temperature stability.

When studying the physical and mechanical characteristics of modified bitumen, it was found that the most effective concentration of PET flakes is the PET content in the amount of 3 %. In this case, there is a significant decrease in penetration and ductility, as well as an increase in the softening temperature of bitumen from 52 to 57 °C. The increase in softening temperature after modification of bitumen with PET waste is explained by the internal structure formed by the polymer [19] and the increase in softening temperature suggests that the addition of PET causes significant resistance to deformation at both moderate and high temperatures. It is assumed that the increase in softening point after polymer modification is beneficial since bitumen with higher softening points is generally considered to be less prone to deformation [20].

The limitation of the use of research results is the use of bitumen of a certain brand, namely with BND 70/100, obtained under the conditions of Pavlodar Petrochemical Plant LLP. When using bituminous binders with other penetration indicators, it is necessary to take into account the concentration of the modifier.

The disadvantage of the study is the lack of data on the rheological properties of the modified bitumen. Since during the period of operation of coatings, a change in the internal structure of bitumen under the action of external deforming forces is essential.

A promising direction of research on the modification of bituminous binders with PET waste is to determine the structure of bitumen depending on external conditions, which will allow a more objective analysis of the behavior of bituminous material in asphalt concrete.

7. Conclusions

1. The structure of a composition based on bitumen and PET waste was studied by IR and ^{1}H NMR spectroscopy. The formation of a new absorption band at 1,130 cm^{-1} on the IR spectra of modified bitumens, as well as a new signal in the ^{1}H NMR spectrum at 3.5 ppm, which indicates the formation of a carbon-oxygen functional group CO, was revealed. On the ^{1}H NMR spectrum of the polymer of the bitumen composition, the signals disappear at 1.77 and 2.33 ppm, which indicates the chemical interaction of bitumen alkenes and PET.
2. Based on the thermophysical characteristics and microstructure of bitumen samples, an increase in the content of asphaltene components was revealed. An analysis of the studies of the physical and mechanical properties of bitumen BND 70/100 before and after its modification shows that the use of polymer waste leads to an improvement in the characteristics of bitumen. At a PET waste concentration of 3 wt%, there is a significant improvement in the characteristics of the modified bitumen: a decrease in penetration by 12 units, an increase in the softening point by 5 °C.

Acknowledgments

This research has been funded by the Science Committee of the Ministry of Education and Science of the Republic of Kazakhstan (Grant No. AP09259187 “Development of an innovative way of using hard-to-use household waste in the production of environmentally friendly building materials”).

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